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Occurrence and Distribution of Persistent Trace Organics in Rainwater in an Urban Region (India)

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Abstract The rainwater samples collected from the Lucknow city (India) were analyzed for selected organochlorine pesticides (OCPs) and polycyclic aromatic hydrocarbons (PAHs). HCH-isomers contributed most to the ΣOCPs with the highest levels of β-HCH isomer. The OCPs, which are currently banned in the country, were also observed. The residue levels of ΣOCPs in rainwater samples ranged between BDL and 447.17 ng L^{-1}. Endrin, β-endosulfan, heptachlor epoxide B and methoxychlor could not be detected in any of the sample. The levels of ΣPAHs in rainwater samples ranged between 19.32 and 11,112.09 ng L^{-1} and the most abundant hydrocarbon was acenaphthylene. Further, the lower molecular weight PAHs dominated over the high molecular weight compounds.

Keywords Rainwater · Trace-organics · Pesticides · PAHs

Many man-made organic compounds are very persistent and once they enter the environment, they remain there for years (Kawamura and Kaplan 1983). Organic substances brought to the atmosphere due to their evaporation from the Earth's surface, or emission from human activities, may be subsequently transported with the masses of air over long distances (Grynkiewicz et al. 2002). Polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) are important classes of persistent organic pollutants (POPs) that are commonly found in the environment. POPs are long-lived organic compounds, and originate almost entirely from anthropogenic activities such as chemical industry, combustion processes and agricultural practices (UNECE 1999). PAHs are a group of compounds of great environmental concern because of the documented carcinogenicity and widespread occurrence of several of its members (IARC 1991). Environmental Protection Agency of the USA (USEPA) has listed the 16 PAHs, some of which are considered as being possible or probable human carcinogens by International Agency for Research on Cancer (IARC 1991), as priority pollutants. PAHs are introduced into the environment mainly via natural and anthropogenic combustion processes. Volcanic eruptions and forest and prairie fires are the major natural sources of PAHs to the atmosphere. Anthropogenic sources include automobile exhaust and tyre degradation, industrial emissions from catalytic cracking, air blowing of asphalt, coking coal, domestic heating emissions from coal, oil, gas and wood, refuse incineration and biomass burning (Manoli et al. 2000). Due to their resistance to degradation, OCPs have remained major global pollutants, with numerous investigations reporting the continued and ubiquitous presence of OCPs in the global atmosphere (Hung et al. 2002). The emission of pesticides to the atmosphere is caused by drop and vapour drift during application, by evaporation from plants, soil and water after application, and by recirculation of fine dust particles by the wind. The intensity of the emission is dependent on the application technique and meteorological circumstances, among other factors.

Both PAHs and OCPs are of particular concern due to their environmental and health effects. These micro organic pollutants are semi-volatile under atmospheric conditions, and may occur both in the gas phase and as attached to particles depending on the vapour pressure of

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the compound (Wania and Mackay 1996). The major removal mechanism of the semi-volatile organic compounds from the atmosphere is through deposition, which can take place either wet or dry. The prevailing removal mechanisms depend on the physico-chemical properties (aqueous solubility, Henry's law constant, vapour pressure) of the compound, its vapour-to-particle portioning and meteorological parameters (Grynkiewicz et al. 2002). Organic pollutants emitted to the atmosphere can return back to land, water and vegetation surfaces through dry deposition of airborne particulate matter, or wet deposition by precipitation, which may subsequently interact with the aquatic and terrestrial life including the human beings causing serious health hazards. Thus, wet precipitation can be responsible both for reduction of the atmospheric load of pollutants as well as contamination of the receiving compartments (soil, water, vegetation). There are several studies reporting levels of trace organic pollutants in the rainwater/wet precipitation in different regions of the world (Manoli et al. 2000; Grynkiewicz et al. 2001; Van Maanen et al. 2001; Grynkiewicz et al. 2002; Gaga and Tuncel 2003), however, data on the distribution pattern and levels of organic pollutants in the rainwater of India are rare. This study reports the levels and distribution pattern of some OCPs and PAHs in the northern Indo-Gangetic alluvium plain, which due to fertile soils, is the region of high agriculture and industrial activities.

Materials and Methods

Lucknow, situated on the banks of the Gomti River, is the state capital of Uttar Pradesh, in the northern part of India (26°45′–27°N latitude; 80°50′–81°05′E longitude). The city covering an area of about 400 km^2 has a population of about 3.5 million (Census 2001). The town is encompassed by agricultural land on its outskirts, orchards, and different types of industrial units such as metals based, chemicals, pharmaceuticals, brick-kilns, etc. In a peripheral area of about 100 km around the city, there are heavy industrial townships, known for heavy industries of fertilizers, pesticides, chemicals, tanneries, distilleries, sugar mills, coal-fired thermal power station, etc. The region, experiences three distinct seasons, winter (November–February), summer (March–June), and monsoon (July–October). The average annual rainfall in the region amount to about 852 mm and the major fraction of precipitation is received during the monsoon months. The atmosphere is loaded with dust by the time the monsoon sets on. Therefore, the first monsoon precipitation washes away the atmospheric dust load cleaning the atmosphere.

A sampling network for collection of rainwater samples (monsoon 2005) was comprised of total 50 locations in the Lucknow city (Fig. 1). The network was designed in such a way as to cover areas representing the semi-urban at the outskirt of the town, highly populated in the old congested city, industrial zones, near highways and newly established colonies through out the town. First monsoon precipitation water samples (single event on a given day or as multiple events in a day) were collected from all the fifty sites at about 10 m height above the ground. The wet only events were collected manually in brown coloured glass bottles using a large diameter glass funnel, pre-flushed with acetone and distilled water. The sampling funnel was uncovered only during precipitation events thereby avoiding contribution from dry fall out. The sample collectors were deployed just before onset of rainfall and were withdrawn immediately after they were filled. All the samples were immediately transferred to the laboratory. The samples were stored at low temperature (4°C) in a refrigerator until analyzed. The collected samples were analyzed for the PAHs and OCPs.

Standards for all pesticides and PAHs used in this work were purchased from Sigma-Aldrich, USA. Standard stock solutions containing 21 OCPs (Aldrin, α- and γ-chlordane, dieldrin, endrin, α-endosulfan, β-endosulfan, endosulfan sulphate, hexachlorobenzene (HCB), α-HCH, β-HCH, γ-HCH, δ-HCH, heptachlor, heptachlor epoxide A, heptachlor epoxide B, pp-DDE, op-DDT, pp-DDD, pp-DDT, and methoxychlor) and 16 PAHs, viz., naphthalene (Nap), acenaphthylene (Acen), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phen), anthracene (Anthra), fluoranthene (Fluo), pyrene, benz[a]fluoranthene [B(a)F], chrysene (Chr), benzo[b]fluoranthene [B(b)F], benzo[k]fluoranthene [B(k)F], benzo[a] pyrene [B(a)P], indeno[1,2,3-cd]pyrene (IP), dibenz[ah]anthracene [D(ah)A] and benzo[ghi]perylene [B(ghi)P] were used in this study. GC grade solvents (Spectrochem, India; 99%) were used throughout the study.

For OCPs analysis, 1 L of the sample (unfiltered) was triply extracted by liquid–liquid extraction in a separatory funnel using 50, 30 and 30 mL of *n*-hexane solvent. The combined solvent extracts were demoisturised using anhydrous granular sodium sulphate and concentrated in a rotary evaporator with a final volume of 1 mL. Then, all the samples were analysed on Varian CP-3800 Gas Chromatograph equipped with fused silica CP-Sil 19 CB capillary column (30m × 0.32mm i.d. × 0.25 µm film thickness) and Ni^{63} ECD. The instrumental analyses were made employing nitrogen (IOLAR) as carrier gas (2 mL min^{-1}) and operating temperatures were: 300°C for the injector port and detector and 220°C for column oven. The split ratio of 1:2 was used. The minimum detection limit for aldrin, α-chlordane, γ-chlordane, dieldrin, op-DDT, pp-DDT, pp-DDE, pp-DDD, endrin, α-endosulfan, β-endosulfan, endosulfan sulfate, HCB, α-HCH, β-HCH, γ-HCH, δ-HCH, heptachlor, heptachlor epoxide-

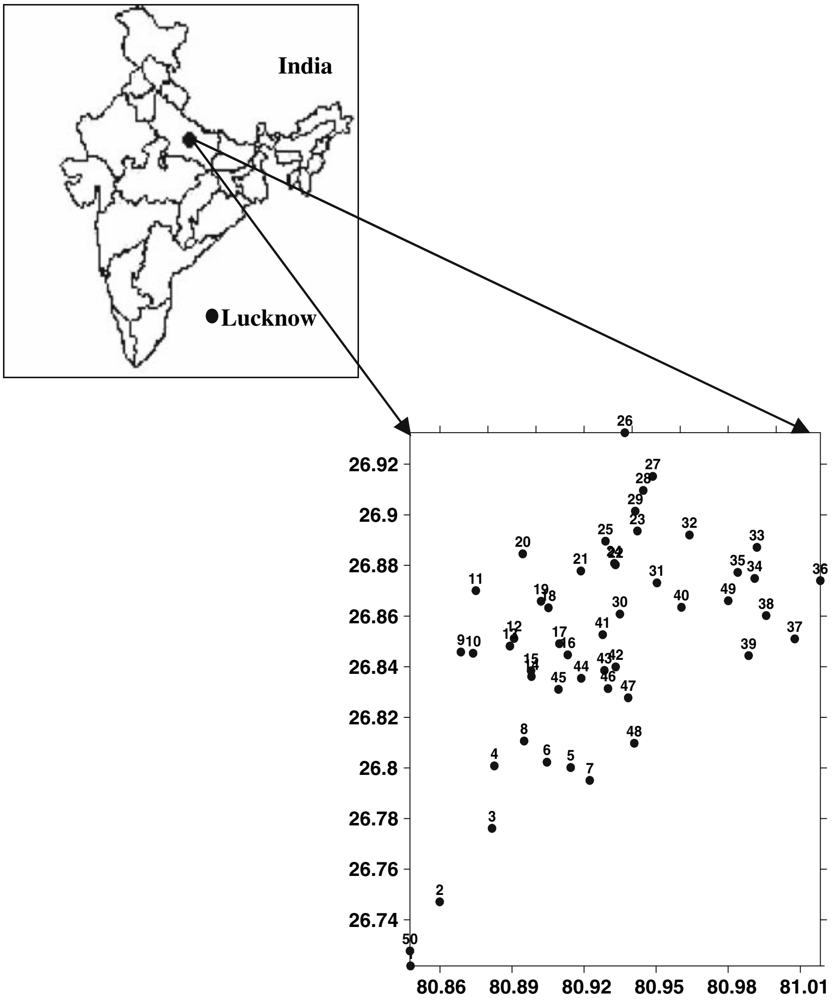

Fig. 1 Map of the Lucknow city showing sampling sites

A, heptachlor epoxide-B and methoxychlor are 100, 100,100, 50, 100, 100, 30, 60, 50, 40, 70, 50, 10, 50, 100, 100, 50, 60, 20, 20 and 100 ng L^{-1}, respectively. A further lower detection of these pesticides was achieved through 1,000-time concentration of the samples. All analyses were carried out in duplicate and the recoveries of individual pesticides were determined through spiked sample method, which were found between 80 and 104% (except for *β*-endosulfan; 72%). Recovery correction factors were applied to the final results.

For PAHs analysis, 1 L of the sample (unfiltered) was triply extracted by liquid–liquid extraction in a separatory funnel using 50, 30 and 30 mL of dichloromethane solvent. The combined solvent extracts were demoisturised using anhydrous granular sodium sulphate and concentrated in a rotary evaporator and the solvent was exchanged to acetonitrile with a final volume of 1 mL. Then the extracts were analysed for PAHs using water–acetonitrile solvent system on HPLC (Metrohm, Switzerland) equipped with a Metrohm IC-709 programmable pump, Metrohm IC-733 separation center, and Metrohm IC-753 UV–vis detector. The separation was achieved on a C-18 column (4.6 × 75 mm). The flow rate was adjusted to 1 mL min^{-1} with a sample injection volume of 100 μL. The detection limit for all the PAHs was 1 ng L^{-1}. All the analysis were carried out in duplicate and the recoveries of individual PAHs were determined through spiked sample method, which were found between 93 and 98%. Recovery correction factors were applied to the final results.

Results and Discussion

The basic statistics of the OCPs residues analyzed in rainwater samples is presented in Table 1. The residue levels of ΣOCPs (sum of 21 OCPs) in rainwater samples ranged between BDL and 447.17 ng L^{-1}. HCH-isomers were the more frequently detected OCPs as compared to others in the rainwater samples, and it was the *β*-HCH, which occurred most frequently followed by *δ*-HCH. On the other hand endrin, *β*-endosulfan, heptachlor epoxide B, methoxychlor and pp-DDT could not be detected in any of the sample. The average concentration of individual OCPs along with the detection frequency is presented in the Fig. 2a. There was found a good correlation ($R = 0.63$, $p < 0.05$) between the detection frequency and residue levels of OCPs. In terms of residue levels again these were the HCH-isomers dominating over the others. The highest mean residue levels (13.13 ng L^{-1}) were observed for the β-HCH followed by the γ-HCH. The median values were BDL for all the OCPs except γ-chlordane, HCH-isomers and heptachlor epoxide-A. Among the aldrin, dieldrin and endrin, residue levels and detection frequency of dieldrin were higher than aldrin, whereas endrin could not be detected in any of the samples. This may be due to the extensive earlier use of dieldrin and aldrin as insecticides for cotton, corn, and citrus fruit and as termiticides for wooden structures (Jorgensen 2001). Further, dieldrin is also a major metabolite of aldrin (Jorgensen 2001; ATSDR 2002). In the rainwater samples of study area, residue levels of aldrin and dieldrin were between BDL and 2.48 ng L^{-1} (mean value 0.28 ng L^{-1}) and BDL–24.40 ng L^{-1} (mean value 2.41 ng L^{-1}), respectively. It is notable that aldrin, and endrin are banned since 1996 and 1990, respectively, whereas dieldrin is under complete ban since 2003 only. Moreover, dieldrin has higher water solubility as compared to aldrin. Although, differences in sampling and analytical methods do not permit a direct comparison among data, however, residue levels of individual OCPs in our study are compared with that reported in the precipitation of other regions. In the rainwater of the Lucknow city (study area) aldrin residues were relatively higher than that in the rainwater of Poland (range 0.05–3.28 ng L^{-1}, mean 0.31 ng L^{-1}) and lower than that in the rainwater of Singapore (range BDL–0.06 ng L^{-1}, mean 0.04 ng L^{-1}) as reported by Grynkiewicz et al. (2001) and Basheer et al. (2003), respectively. The dieldrin residues were higher than that observed in the rainwater of Singapore (Basheer et al. 2003). HCB was detected in 14% samples with a mean value of 0.94 ng L^{-1}. The principal

Table 1 Basic statistics of OCPs residues (ng L^{-1}) in rainwater of Lucknow city

OCP	Range	Median	Mean ± SD	25th Percentile	75th Percentile
Aldrin	BDL–2.48	BDL	0.28 ± 0.51	BDL	0.42
α-Chlordane	BDL–4.15	BDL	0.48 ± 0.86	BDL	0.85
γ-Chlordane	BDL–24.34	1.07	2.66 ± 4.13	BDL	3.79
Σ-Chlordane	BDL–28.49	1.54	3.14 ± 4.81	BDL	3.79
Dieldrin	BDL–24.40	BDL	2.41 ± 5.24	BDL	1.86
Endrin	BDL	BDL	BDL	BDL	BDL
α-Endosulfan	BDL–6.71	BDL	0.15 ± 0.96	BDL	BDL
β-Endosulfan	BDL	BDL	BDL	BDL	BDL
Endosulfan sulfate	BDL–63.69	BDL	2.84 ± 11.00	BDL	BDL
Σ-Endosulfan	BDL–63.69	BDL	2.99 ± 11.00	BDL	BDL
HCB	BDL–20.13	BDL	0.94 ± 3.69	BDL	BDL
α-HCH	BDL–62.26	0.55	3.30 ± 10.59	BDL	1.08
β-HCH	BDL–93.03	5.22	13.13 ± 19.34	1.01	14.82
γ-HCH	BDL–129.66	0.71	3.95 ± 18.39	0.17	1.54
δ-HCH	BDL–43.14	1.49	3.09 ± 6.47	0.55	3.15
Σ-HCH	BDL–328.08	9.92	23.48 ± 49.02	2.29	21.78
Heptachlor	BDL–8.45	BDL	0.77 ± 1.62	BDL	0.69
Heptachlor epoxide A	BDL–7.35	0.83	1.43 ± 1.78	BDL	1.87
Heptachlor epoxide B	BDL	BDL	BDL	BDL	0.00
Σ-Heptachlor	BDL–15.80	1.27	2.20 ± 3.00	BDL	2.85
pp-DDE	BDL–4.51	BDL	0.21 ± 0.79	BDL	BDL
op-DDT	BDL–2.80	BDL	0.06 ± 0.40	BDL	BDL
pp-DDD	BDL–6.22	BDL	0.26 ± 1.18	BDL	BDL
pp-DDT	BDL	BDL	BDL	BDL	BDL
Σ-DDT	BDL–6.22	BDL	0.53 ± 1.42	BDL	BDL
Methoxychlor	BDL	BDL	BDL	BDL	BDL
Σ-OCPs	BDL–447.17	16.23	35.99 ± 69.18	4.22	31.00

Lower detection achieved through 1,000-time concentration of samples

BDL below detection limit

source of HCB to the atmosphere is believed to be volatilization from contaminated soils. It is evident that HCB has never been registered as pesticide in India, however, use of HCB has been reported in India till 1997 (UNEP 2003). HCB has very long atmospheric residence time, which allows it to become widely distributed in the global atmosphere. It has long atmospheric degradation lifetimes (940 days) (Brubaker and Hites 1998). Among the chlordane isomers, γ-chlordane dominated over the α-chlordane both in terms of frequency of detection and residue levels (Fig. 2a). The γ-isomer is more persistent in nature, which may be present in the environment after several years of its ban for use. The chlordane is banned in India since 1996 (UNEP 2003). Among the isomers/metabolites of endosulfan (α-, β-endosulfan, and endosulfan sulfate), endosulfan sulphate predominated with the residue levels ranging from BDL–2.84 ng L^{-1}. The residue levels of α-endosulfan ranged between BDL and 0.15 ng L^{-1}, whereas β-endosulfan could not be detected. Total heptachlor residues (sum of heptachlor and heptachlor epoxides) ranged between BDL and 2.20 ng L^{-1}. Heptachlor is metabolised to heptachlor epoxides in the soils, plants and animals, which is more stable. Heptachlor epoxide-A occurred more frequently (Fig. 2a, ranging between BDL and 1.43 ng L^{-1}) as compared to heptachlor (range BDL–8.45 ng L^{-1}, mean 0.77 ng L^{-1}), whereas heptachlor epoxide-B was absent. This may be attributed to the relatively high stability of the heptachlor epoxides and ban on the use of heptachlor since 1996 in the country (UNEP 2003). Among the HCH isomers β- and δ-HCH were detected most frequently. On an average β-HCH contributed 56% to the total HCH concentration. In our case, both the high frequency of detection and residue levels of β-isomer, which is more persistent as compared to other isomers, suggest for earlier usage of the pesticide. Basheer et al. (2003) have also observed relatively higher levels of β-HCH as compared to α-HCH and γ-HCH in the rainwater of Singapore. Further, predominance of β-HCH has also been reported in the soil, surface water and groundwater samples in vicinity of the studied area (Singh et al. 2005, 2007). The γ-HCH was detected in 76% of the samples ranging from BDL to 129.66 ng L^{-1} with a mean value of 3.95 ng L^{-1}. The average residue levels of γ-HCH in the rainwater of the studied area are relatively higher than that found in rainwater of Poland (range 0.01–5.06 ng L^{-1}, mean 0.37 ng L^{-1}; Grynkiewicz et al. 2001) and Singapore (range BDL–0.03 ng L^{-1}, mean 0.02 ng L^{-1}; Basheer et al. 2003). Technical HCH is reported to have a ratio of α-HCH/γ-HCH of 4–7, while lindane contains approximately 99% γ-HCH (Iwata et al. 1993). In the rainwater samples, a median value of 0.60 for α-HCH/γ-HCH ratio suggests use of lindane in the region. Among the DDT metabolites, pp-DDE, op-DDT, and pp-DDD were detected in 8, 2 and 6% samples only, whereas pp-DDT was absent in all the samples. This may be due to the lower aqueous solubility of the DDT-metabolites.

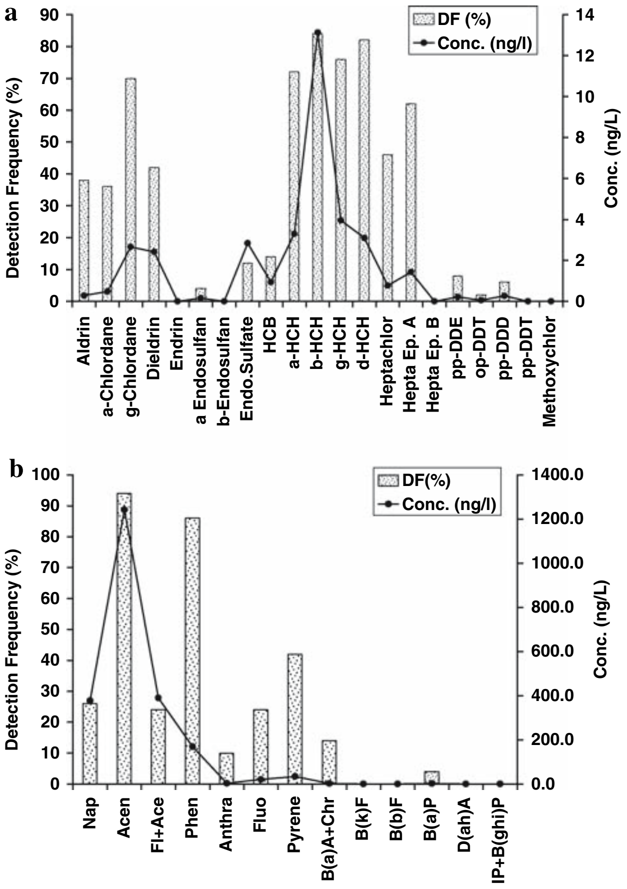

Fig. 2 Detection frequency and concentration of individual **a** OCPs and **b** PAHs in the rainwater samples of the study region

The basic statistics of the concentration levels of 16 USEPA PAHs in rainwater samples are presented in Table 2. The levels of ΣPAHs (sum of 16 PAHs) in rainwater samples ranged between 19.32 and 11,112.09 ng L^{-1}. In the rainwater samples the most abundant hydrocarbon was acenaphthylene followed by phenanthrene and pyrene with detection frequencies 94, 84, and 42%, respectively (Fig. 2b). There was found a good correlation ($R = 0.72$, $p > 0.05$) between the detection frequency and concentration of ΣPAHs in the rainwater. The median values for the PAHs were BDL except acenaphthylene and phenanthrene. The highest concentration level was observed for the acenaphthylene. In general, acenaphthylene, fluorene + acenaphthene, naphthalene and phenanthrene contributed more (>97%) to the ΣPAHs burden of the rainwater, with the highest contribution from acenaphthylene (>55%). In rainwater the lower molecular weight PAHs were found to be more prominent than the higher molecular weight compounds. This may be due to the

Table 2 Basic statistics of PAHs (ng L^{-1}) in rainwater of Lucknow city

PAH	Range	Median	Mean ± SD	25th Percentile	75th Percentile
Nap	BDL–4,684.95	BDL	377.79 ± 900.39	BDL	151.25
Acen	BDL–5,164.12	706.30	1,242.31 ± 1,150.62	365.41	2,163.99
Fl + Ace	BDL–6,673.92	BDL	389.42 ± 1,193.21	BDL	BDL
Phen	BDL–628.02	155.70	169.16 ± 148.87	55.07	219.47
Anthra	BDL–102.58	BDL	3.06 ± 15.12	BDL	BDL
Fluo	BDL–433.54	BDL	20.83 ± 65.67	BDL	BDL
Pyrene	BDL–353.61	BDL	34.08 ± 65.21	BDL	40.43
B(a)F + Chr	BDL–30.19	BDL	2.03 ± 5.92	BDL	BDL
B(k)F	BDL	BDL	BDL	BDL	BDL
B(b)F	BDL	BDL	BDL	BDL	BDL
B(a)P	BDL–56.70	BDL	1.61 ± 8.63	BDL	BDL
D(ah)A	BDL	BDL	BDL	BDL	BDL
IP + B(ghi)P	BDL	BDL	BDL	BDL	BDL
Σ-PAHs	19.32–11,112.09	1,601.10	2,240.27 ± 2,186.94	566.63	3,259.14

BDL below detection limit

physico-chemical properties of the compounds such as water solubility which is much higher for the lower molecular weight PAHs as compared to the higher molecular weight PAHs. These results are in accordance with that reported by Sahu et al. (2004) in the rainwater of Mumbai city (India) and in the rainwater of Turkey (Gaga and Tuncel 2003). Although, differences in sampling and analytical methods do not permit a direct comparison among data, levels of individual PAHs in our study are comparable with that reported in the precipitation of Mumbai city of India (Sahu et al. 2004), however, relatively lower levels have been reported in the rainwater of Singapore (Basheer et al. 2003) (Table 3).

In terms of the number of the fused rings present in the chemical structure of the polycyclic aromatic hydrocarbons, it was observed that the three-ringed hydrocarbons were the most abundant ones followed by the two-ringed hydrocarbons. The occurrence of PAHs in precipitation could be hypothesized due to a combination of the removal processes, viz., particle scavenging by rain (within or below cloud), vapour dissolution in rain droplets, dry-gaseous deposition and dry-particle deposition (Manoli et al. 2000). The abundance of three-ringed hydrocarbons in the rainwater of the study area may be attributed to the partitioning of these hydrocarbons from the dissolved to organic carbon-rich settling particles, whereas higher molecular weight PAHs exist primarily in the particulate phase in both the atmosphere and water (Bidleman 1988). Among the carcinogenic PAHs identified by the IARC (1991) only Benzo(a)anthracene (with chrysene) and Benzo(a)pyrene could be detected [detecton frequency 14 and 4% for Benzo(a)anthracene + chrysene and Benzo(a)pyrene, respectively]. However the values for median, 25th and 75th percentile were BDL for both the PAHs.

The rainwater samples at Lucknow city (India) contained several OCPs and PAHs. Among the analysed OCPs, HCH-isomers contributed most to the total burden of OCPs with the highest levels of β-isomer. The OCPs

Table 3 Literature data of PAH concentration in rainwater (ng L^{-1})

PAH	Lucknow, India Range (Mean)	Turkey Range (Mean)	Singapore Range (Mean)	Mumbai Range (Mean)
Nap	BDL–4,684.95 (377.79)	–	0.008–0.162 (0.060)	–
Acen	BDL–5,164.12 (1,242.31)	–	0.005–0.044 (0.013)	–
Phen	BDL–628.02 (169.16)	–	0.006–0.021 (0.012)	30.10–440.90 (86.7)
Anthra	BDL–102.58 (3.06)	13.96–492.3 (202.10)	0.009–0.041 (0.016)	14.40–124.50 (54.8)
Fluo	BDL–433.55 (20.83)	36.63–645.0 (271.10)	0.005–0.027 (0.015)	22.90–379.80 (62.3)
Pyrene	BDL–353.61 (34.08)	57.97–687.2 (260.70)	0.005–0.049 (0.016)	20.80–293.80 (35.8)
B(a)P	BDL–56.70 (1.61)	21.74–47.69 (28.24)	0.021–0.165 (0.071)	2.20–25.60 (8.9)
Reference	Present study	Gaga and Tuncel (2003)	Basheer et al. (2003)	Sahu et al. (2004)

which are currently banned in the country were also observed. Three-ringed PAHs dominated among the individual PAHs in the rainwater of the Lucknow city. However, the sources and their regions were not apparent. Further, investigations regarding the sources of organic pollutants are important; this study will contribute to the present knowledge of environmental monitoring of trace organics and their risk assessment.

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