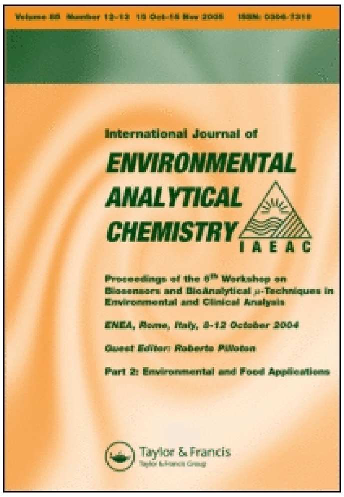

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Calcutta Pollutants: Part II. Polynuclear Aromatic Hydrocarbon and Some Metal Concentration on Air Particulates During Winter 1984

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Calcutta Pollutants:

Part II. Polynuclear Aromatic Hydrocarbon and Some Metal Concentration on Air Particulates During Winter 1984

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The concentration of 25 polynuclear aromatic hydrocarbons (PAH) and 12 metals on air-particulate matter of Calcutta city were determined during January–February 1984. X-ray fluorescence (XRF) was applied for determination of metal concentration and gas chromatography–mass spectrometry (GC–MS) for PAH determination. Both PAH and metal concentrations found are present at higher concentration when compared with cities in Europe and USA.

KEY WORDS: PAH's, air particulates, XRF, heavy metals, GC–MS.

INTRODUCTION

The air in Calcutta is considerably more polluted during winter. The urban atmosphere is then covered by a thick smog, causing eye irritation especially in the morning and the evening. Visibility then sharply declines within the range of only a few metres. Few data are available about the possible pollutants in the air of the city. World

Health Organisation (WHO) on its global programme on air quality monitoring, measured the sulfur dioxide and total suspended particulate (TSP) matter of Calcutta air and claimed that TSP of Calcutta air is the highest in the world.[1]

Calcutta within its corporation area 104 km^2 has a population of about 4.5 million. Thus as high as about 45 000 perople per km^2. The major causes of air pollution of the city are emission from motor cars, burning of coal for domestic cooking, innumerable small factories, very frequent traffic jams and small area of road compared to city area (6 %). Doubtlessly some of them are major sources[2-4] of polyaromatic hydrocarbons and heavy metals. Other than the higher toxicity of PAH, several of these compounds are known to be potential carcinogens.[5] As Calcutta has high TSP values, chronic pulmonary diseases can be aggravated.[6-10] Other than PAH, over recent years a great deal of research has also been done on the quantification of human exposure to toxic metals. The literature records show that children living in cities have high concentration of toxic metals in their blood.[11,12] Considering all these pollution sources of Calcutta city it was expected that heavy metals and PAH would be high in Calcutta air especially during winter. A study was undertaken during January–February 1984 at five localities. The aim of this study was to examine the concentration of PAH and metals. In our procedure for collection of metals from city air, we used a low volume sampler and the air was sucked at the speed of inhalation, i.e. about 5 l/min^{-1} and at about 1.5 m above the ground. On each filter, about 1 m^3 of air was sampled. The concentration of the metal on the filter was determined by X-ray fluorescence (XRF). For determination of the PAH we employed a high volume sampler at about 400 l/min^{-1} at 2.5 to 3.0 m above the ground and gas chromatography–mass spectrometry (GC–MS) was applied. This work is the first preliminary study within Calcutta. It will surely provide an indication of the level of such pollutants in city air during winter.

EXPERIMENTAL

Sampling sites

Air particulates were collected at five different localities in Calcutta with an attempt to achieve maximum coverage of the city.

1. *Gariahat* This is at the southern part of Calcutta city. It is a posh shopping area with very busy traffic and high density of population during working hours. Innumerable small shops also flourish on footpaths. Samples were collected at the crossing of four main roads.

2. *Park Street* This is a commercial centre with high traffic density. Samples were collected at the crossing of two wide roads. Opposite to the sampling location there is a very big playground.

3. *Esplanade* Samples were collected at the junction of four wide main roads. It is both a commercial as well as a shopping centre and is situated almost at the centre of Calcutta. It also possesses very high traffic density.

4. *B.B.D.-Bag* It is at the heart of the city, being the main office area. In week days, the traffic density is highest during 8 a.m. to 6 p.m. It is also almost at the centre of the city. Samples were collected on an island at the crossing of six roads.

5. *Shyambazar* It is similar to Gariahat, but with even more dense population and high traffic density. The cars move very slowly here. Samples were collected at the junction of five roads. It is situated at the northern part of Calcutta.

For XRF analysis particles were collected on polycarbonate membrane filter (Nuclepore Corp., California) with 0.45 micropore size, 47 mm diameter. The filter holder is from Schleicher & Schnoll, Dassel, West Germany. Approximately 1 m^3 of air was sampled on each filter during peak traffic hours of the day—8 a.m.–12 noon and 4–8 p.m.

For PAH study, air particulates were collected on glass fibre filter (Staplex, Brooklyn, New York), 8 × 10 in size, with pore size 0.3 μm. Samples were collected for 8 hours, i.e. 8 a.m.–12 noon and 4–8 p.m. After sampling, the polycarbonate membrane filters for metal analysis were kept in small plastic boxes and filters for PAH analysis were packed in aluminium foil. The filters were analysed 2 months after sampling. The total number of filters analysed are as follows: Gariahat 21, Park Street 18, Shyambazar 19, Esplanade 20 and BBD-Bag 22.

Analysis of PAH on glass fibre filter

The particulate samples were extracted continuously with benzene and methanol consecutively for a period of 8 hours. The procedure is adequate for PAH with molecular weight under 300 a.m.u.[13,14] The aliphatic HCs present on particles were removed from the extract prior to the analysis of PAH by GC–MS in the single ion monitoring mode to increase detection efficiency for PAH-molecular ions. The group separation described by Grimmer *et al.*[15] based on the partition of aliphatic and aromatic HCs between cyclohexane/dimethylformamide (DMF)–water, was reinvestigated for quantitative yield on a test mixture of PAH and was proven to be unsatisfactory. The lower PAH can remain in the first cyclohexane layer with yields of 10–20%. Obviously these are not recovered in the back extraction step from dimethylformamide–water 1:1, in which some higher PAH remains with a yield of 8–15%. The following modifications yield a superior recovery of PAH: The first partition was performed between *n*-hexane and DMF–water (9:1) twice and the upper layer washed with water (98% recovery of aliphatics in *n*-hexane, no PAH left over), the second partition was performed by back extraction of DMF–water (1:1) with cyclohexane and benzene consecutively (97% recovery of all PAH). Fine soot particles entrained in the Soxhlet–methanol extraction are mostly discarded with the DMF–water layer, but occasionally some may persist in the organic layer.

To the samples *p*-terphenyl and triphenylbenzene are added as internal standards. The use of capillary column GC is necessary to separate isomeric PAH-pairs with expected different reactivity. Further clean-up of the PAH fraction is not required, since single ion monitoring of the molecular ions combined with narrow retention time windows will eliminate most interferences[16] or overlap with other isomeric trace PAH compounds. A fused silica 25 m capillary column coated with Sil 5 (i.d.: 0.32 nm, film thickness 0.2 μm) was programmed from 140–250 °C at the rate of 40 min^{-1} to elute the PAH from phenanthrene up to dibenzanthracenes. This column was directly inserted into the transfer line to the ion source of a Finnigan GC–MS coupled to a model 6000 Data System provided with mass fragmentography software. Switching of the ion masses was performed at specific time intervals by reading the new parameters from paper tape under operator control.

Analysis of metal ions on Nuclepore filter

The X-ray fluorescence measurements were performed with a Kevexray 810 system. A water cooled tungsten anode X-ray tube, powered by a Siemens Crystalloflex II high voltage generator, was used as a primary radiation source. Excitation of the samples was done in a secondary target geometry using the fluorescent K-radiation of a molybdenum target.[17] Characteristic X-rays in the samples were analysed with a Si(Li)-semiconductor detector. After signal processing the spectral data was accumulated in a 1024 channel multichannel analyser (Tracor, Northern, MS 701). Spectra were stored on a magnetic tape unit and transferred to a VAX II/780 computer of off-line processing.

The loaded Nuclepore filters were mounted by pressing them between two Teflon rings in order to obtain a smooth and flat surface. No further preparation was necessary to analyse the filters. The samples were irradiated for 3000 s under vacuum at a X-ray tube potential of 40 kV and with a current of 40 mA.

Thin standards, prepared by evaporating elements of simple compounds onto a 4 μm thick Mylor foil were used to calibrate the instrument.[18] Spectrum analysis was done using the non-linear least squares fitting algorithm AXIL.[19] The overall accuracy and precision of the X-ray fluorescence analysis procedure is better than 10%.[19] The elements S, Cl, K, Ca, Ti, Mn, Fe, Cu, Zn, Sr, Pb, V, Cr and As, were found and quantitatively analysed in samples. All values were corrected for blank contribution of the filter substrate. Values for bromine are not given because of the high bromine content of the Nuclepore filter.

RESULTS AND DISCUSSION

Table 1 presents the concentration of PAH in the five locations. The highest concentrations were obtained in Shyambazar. Its concentration is almost twice as high as in the other localities. The concentration in BBD-Bag and Esplanade and so also, those in Park Street and Gariahat are comparable to each other. Despite the preliminary character of the study it can be derived that the concentration of PAH in Calcutta during winter is higher than in many other major cities. Table 2 gives a comparison of winter concentration PAH in Calcutta with Wilrijk, Belgium,[20,21] Paris,

Table 1 Concentration of PAH in five localities of Calcutta (ng/m^3)

Compounds (relative carc.)[a]	*MW/notation*	*Gariahat*	*Park Street*	*Esplanade*	*BBD-Bag*	*Shyambazar*
Benz(a)anthracene (+ +)	228 α	19.9	30.2	44.4	50.2	65.6
Chrysene (+)	228 β					
Triphenylene	228 γ	22.4	23.7	40.3	36.8	59.7
3,4,benzacridine (+)	229	1.06	1.17	1.97	1.68	4.76
Benzanthrone (+)	230	9.11	13.78	20.2	12.98	23.8
Benzo(b)fluoranthene (+ +)	252 α	108.8	111.7	123.6	127.6	200
Benzo(j)fluoranthene (+ +)	252 β	35.1	22.4	32.9	32.7	66.1
Benzo(e)pyrene (+)	252 γ	48.5	52.4	85.3	83.1	182
Benzo(a)pyrene (+ + +)	252 δ	30.2	43.2	55.3	55.5	120.2
o-phenylene pyrene (+)	276 α	26.5	36.9	56.2	50.5	72.4
Dibenz anthracene (+ + +)	278 α	8.59	12.1	17.3	15.1	40.9
Non-carcinogen						
Phenanthrene	178 α	13.2	11.2	15.6	18.1	21.4
Anthracene	178 β	2.61	2.51	2.85	4.25	5.5
Benzoquinoline	179 α	1.08	0.84	1.09	0.88	1.80
Phenanthridine	179 β	0.60	0.98	0.55	0.69	1.83
Fluoranthene	202 α	11.02	10.62	12.55	16.5	26.1
Pyrene	202 β	19.9	24.2	24.2	39.5	43.6
Benzo(a)fluorene	216 α	3.66	3.50	1.84	5.69	6.32
Benzo(b)fluorene	216 β	2.51	2.71	4.36	5.11	8.01
Benzo(c)fluorene	216 γ	1.30	1.80	2.94	2.78	6.21
Methyl pyrene	216 δ	1.38	1.75	2.58	2.30	4.96
Methyl fluoranthenes	216 ε	1.29	1.34	1.93	2.18	4.98
Perylene	252 ε	4.45	5.29	8.35	6.98	7.65
Benzo(ghi)perylene	276 β	33.6	57.8	83.1	77.4	141
Picene	278 β	6.1	6.75	11.85	7.35	29.50
1,2,5,6 and 1,2,7,8 dibenzacridine	279 $\alpha+\beta$	8.12	7.27	13.16	9.36	7.50

[a] Santodonato *et al.*[30]

Table 2 Comparison of PAH concentration of Calcutta with Paris (France), Wilrijk (Belgium) and New Jersey (USA). Concentration in ng/m^3

Compounds (*relative carc.*)	*Mt*	*Calcutta*[a] (*India*)	*Mt*	*Wilrijk*[b] (*Belgium*)	*Mt*	*Paris*[c] (*France*)	*Mt*	*New Jersey*[d] (*USA*)
Benz(a)anthracene (+ +)	228 α	19.9 – 65.6			228 α	4.8 –25.0	228 α	0.56–0.88
Chrysene (+)	228 $\beta+\gamma$	22.4 – 59.7	228 $\beta+\gamma$	26–48	228 $\beta+\gamma$	8.9 –29	228 $\beta+\gamma$	0.62–2.42
3,4,benzacridine (+)	229	1.06– 4.76	229	1.8– 3.9	229	n.a.	229	n.a.
Benzathrone (+)	230	9.11– 23.8	230	n.a.	230	n.a.	230	n.a.
Benzo(b)fluoranthene (+ +)	252 α	108.8 –200.0	252 $\alpha+\beta$	26–66	252 α	15.7 –36.0	252 α	0.78–1.09
Benzo(j)fluoranthene (+ +)	252 β	22.4 – 66.1			252 β	5.5 –13.0	252 β	0.70–1.04
Benzo(e)pyrene (+)	252 γ	48.5 –182	252 $\gamma+\delta$	17–44	252 γ	8.8 –27.0	252 γ	1.09–1.83
Benzo(a)pyrene (+ + +)	252 δ	30.2 –120.2			252 δ	4.6 –15.0	252 δ	0.69–1.06
o-phenylene pyrene (+)	276 α	26.5 – 92.4	276 α	3.9–12.0	—	—	—	—
Dibenz anthracene (+ + +)	278 α	8.59– 40.9	278 α	2.2– 7.4	278 α	0.69– 2.8	278 α	0.19–0.75
Non-carcinogen								
Phenanthrene	178 α	11.2 – 21.4	178 $\alpha+\beta$	7.7–18.0	—	—	—	—
Anthracene	178 β	2.51– 5.5						
Benzoquinoline	179 α	0.84– 1.80	179 $\alpha+\beta$	1.1– 1.7	—	—	—	—
Phenanthridine	179 β	0.55– 1.83						
Fluoranthene	202 α	10.02– 26.1	202 α	16.0–18.0	—	—	—	—
Pyrene	202 β	19.9 – 43.6	202 β	9.1–15.0	202 β	6.7 –10.4	202 β	1.79–2.77
Benzo(a)fluorene	216 α	1.8 – 6.32	216 α	3.5– 6.6	—	—	—	—
Benzo(b)fluorene	216 β	2.51– 8.01	216 $\beta+\gamma$	3.4– 6.2	—	—	—	—
Benzo(c)fluorene	216 γ	1.30– 6.21		—	—	—	—	—
Methyl pyrene	216 δ	1.38– 4.96	216 $\delta+\varepsilon$	1.3– 2.9	—	—	—	—
Methyl fluoranthenes	216 ε	1.29– 4.98		—	—	—	—	—
Perylene	252 ε	7.65– 8.35	252 ε	1.3– 3.0	252 ε	0.88– 4.4	—	—
Benzo(ghi)perylene	276 β	33.6 –141.0	276 β	8.5–23	276 β	11.5 –31.0	276 β	1.13–1.44
Picene	278 β	2.95– 11.85	278 β	2.1– 7.6	278 β	1.5 – 4.4	—	—
1,2,5,6 and 1,2,7,8 dibenzacridine	279 $\alpha+\beta$	7.27– 13.16	279 $\alpha+\beta$	0.3– 2.1	—	—	—	—

n.a. not available.

[a] Range of PAH in five places of Calcutta during January–February 1984.

[b] Range of 3 days during winter.

[c] Range in 2 places during January 1980.

[d] Geometric mean of three urban sites during winter 1983 in New Jersey (USA).

France[22] and New Jersey, USA.[23] The concentrations of PAH in Calcutta are about a factor 5 higher than the level in Europe. Compared with New Jersey the situation is a lot worse. In winter the people living in Calcutta are practically breathing the exhaust fumes of which the carcinogenic potential is well known.[24] The ratio of BeP/BaP is substantially higher than 1, where it is close to 1 for diesel and gasoline engines.[25] This high value of BeP/BaP indicates that there should be contribution from some sources other than internal combustion engines. Although it is difficult to assign sources from PAH profile if the individual contributions are not characterized. It is possible that this additional emission is due to higher coal burning for domestic energy.

With the concentration of BaP in winter (Table 1), we will try to make some assessment about the health risk of the population. We are assuming that we have measured peak concentration and for our calculation we are taking an average of $50\,ng/m^3$. Considering an adult breathes $20\,m^3$ of polluted air a day and taking into account that particle size distribution will favour the enrichment of the submicrometer fraction, the daily intake of BaP from the airborne particle (considering about one-fifth is absorbed by the system) will be $0.2\,\mu g$ or about $75\,\mu g$ per year. For test animals the amount needed in mg to induce carcinomas within a few weeks, the same amount will enter to the people in Calcutta after 15 years (while in the case of Europe 50 years). However, one has to consider that in this calculation we have taken very preliminary data, favourable inhalation behaviour, particle size distribution etc.

Table 3 shows the concentration of the few metals (and two nonmetals). The elemental concentrations are comparable among the localities. However, Shyambazar again shows a higher concentration. The metal concentrations in Calcutta city air particulates are very high when compared with the yearly average values of many cities of the world. However, one has to realise that these values of PAH, metals and peak hours are in winter, by side of roads on the day when traffic was at a maximum.

In Table 4, we compare the average elemental concentrations of all localities of Calcutta with that of Berlin (West Germany), Brussels (Belgium), Kanpur and Bombay (India). The Berlin concentration reported in the table is the maximum value during a "smog" episode with very high particulate concentrations during

Table 3 Some metal and non-metal concentration on air particulate matter in five locations of Calcutta ($\mu g/m^3$)

Element	*Gariahat*	*Park Street*	*Shyambazar*	*Esplanade*	*B.B.D-Bag*
S	15.0–34.5	20.6–30.7	19.2–59.07	19.7–32.6	23.8–40.7
Cl	7.1–14.9	7.9–12.3	12.6–35.5	10.3–16.5	7.2–20.3
K	8.8–19.6	23.1–31.7	15.5–71.8	17.2–26.3	10.5–50.3
Ca	17.1–49.3	53.4–80.7	36.3–244.9	34.9–57.4	22.0–114.2
Ti	1.8–4.1	4.5–7.2	3.09–15.4	4.9–6.1	2.10–13.9
V	0.11–0.28	0.21–0.35	0.24–0.38	0.09–0.41	0.42–0.84
Cr	0.09–1.48	0.02–0.98	0.07–1.94	0.03–0.84	0.04–0.33
Mn	0.28–0.87	0.14–1.67	0.65–4.2	0.14–2.60	0.62–3.02
Fe	11.8–44.0	12.6–54.3	17.3–56.1	12.4–42.3	14.6–39.3
Cu	0.43–0.89	0.1–2.05	0.138–2.64	0.22–0.78	0.37–0.81
Zn	0.50–4.39	0.12–3.63	0.82–5.44	2.2–4.7	1.3–3.09
As	0.16–1.21	0.12–0.39	0.28–1.92	0.22–0.31	0.34–1.96
Sr	0.15–0.21	0.13–0.25	0.14–0.96	0.18–0.23	0.14–0.62
Ph	2.08–10.95	1.45–11.09	3.65–15.9	1.3–9.7	1.03–9.4

Table 4 Comparison of metal and non-metal concentration of Calcutte aerosol with Brussels (Belgium), Berlin (W. Germany), Kanpur and Bombay (India) (conc. in $\mu g/m^3$)

Elements	*Calcutta*	*Berlin*	*Brussels*	*Kanpur*	*Bombay*
S[a]	19.93	n.a.	n.a.	8.41[a]	n.a.
Cl	10.28	n.a.	n.a.	n.a.	n.a.
K	11.65	n.a.	n.a.	n.a.	n.a.
Ca	43.6	n.a.	n.a.	n.a.	n.a.
Ti	2.6	n.a.	n.a.	n.a.	n.a.
V	0.18	0.101	0.169	n.a.	n.a.
Cr	0.11	0.098	0.077	0.091	n.a.
Mn	0.43	0.286	0.385	n.a.	n.a.
Fe	26.4	9.660	16.174	7.10	13.97
Cu	1.12	0.932	0.130	2.60	0.55
Zn	3.04	4.340	2.326	1.21	0.97
As	0.26	0.067	n.a.	n.a.	n.a.
Sr	0.17	n.a.	n.a.	n.a.	n.a.
Pb	6.63	1.40	4.32	5.35	0.463

[a] As sulfate.
n.a., not available.

winter 1981–1982.[26] In the case of Brussels,[27] the values reported are the maximum observable daily concentration (98th percentiles) during the years 1972–1977. The comparative study shows that the average values of Calcutta during the middle of January to the middle of February are even slightly higher than the maximum concentrations of these two Western European cities. Table 4 also compares the average values in Calcutta with that of the value of the highly industrialised city of Kanpur (India) during January–February, 1973–1975.[28] Table 4 also compares with the Zn, Cu, Fe and Pb values of Bombay (India). Bombay is also known to be very polluted.

The comparative study of all these cities show that Calcutta's pollutant concentrations during the period of measurement are really high. The average TSP value found during our sampling period was 650 $\mu g/m^3$. Considering such high TSP values, the concentrations of pollutants found is not unrealistic.

The reason for such high concentrations in Calcutta during winter is not easy to explain. The factors may be: (a) the unfavourable meteorological conditions; (b) high population density; (c) more than 80% of the population use coal or kerosene for cooking, and most of the cases do not use a chimney; (d) the conditions of the motor cars within the city are not up to the expected level to cause minimum pollution; (e) the roads are narrow and the percentage of the roads in comparison to the city area is only 6; (f) pretty often traffic jams occur and cars cannot move more than an average 10–15 miles per hour within the city; (g) for about the past 10 years the construction work of the metro-rail is functioning within the city of Calcutta; (h) during winter the city air is covered with "smog" during the morning and evening.

CONCLUSION

The overall study of PAH and metals on Calcutta air particulates indicate that in winter the concentration is very high (or higher than high). It is expected that since the TSP value is the highest in the world and the meteorological conditions during winter favour the pollutants to stay above ground level. The picture is expected to be different during the summer and the monsoon season. Although this is a preliminary study, it gives an idea of pollution during winter. Round-the-year studies for a few years are needed to know the actual concentrations of these pollutants.

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