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Heavy metal contamination of Ganga river at Varanasi in relation to atmospheric deposition

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Abstract: We investigated the mid stream water quality of Ganga river as influenced by aerially - driven heavy metals at Varanasi, India. Twelve sampling stations were selected along a 20 km long stretch of the river. Mid stream sub-surface water samples collected at fortnightly intervals from all the sites were acid digested and analyzed for Cd, Cr, Cu, Ni, Pb and Zn. The data revealed that the mid-stream water of river Ganga at Varanasi is invariably contaminated by heavy metals. Highest concentrations of Cd, Cr, Cu, Ni and Pb were recorded during winter and that of Zn during summer season. The overall concentration of heavy metals in water showed the trend : Zn > Ni > Cr > Pb > Cu > Cd. Concentrations of all the heavy metals were high in down - stream sampling stations. Correlation analysis showed that heavy metal concentration in mid-stream water had significant positive relationship with rate of atmospheric deposition at respective sites. Although the concentrations of these metals in water remained below the permissible limits of Indian standards for drinking water, levels of Cd, Ni and Pb at three stations, were above the internationally recommended (WHO) maximum admissible concentrations (MAC). These observations suggest that use of such water for drinking may lead to potential health risk in long-run. The study has further relevance in understanding the atmosphere - water interaction in polluted environment and for management of water bodies even those situated away from direct anthropogenic discharge.

Resumen: Investigamos la calidad del agua de la corriente media del río Ganges en relación con los metales pesados transportados por aire en Varanasi, India. Se seleccionaron doce estaciones de muestreo sobre una sección del río de 20 km de largo. Las muestras de agua sub-superficial del centro de la corriente recolectadas quincenalmente en todos los sitios fueron sometidas a digestión ácida para analizar su contenido de Cd, Cr, Cu, Ni, Pb y Zn. Los datos revelaron que el agua de la corriente media del río Ganges en Varanasi está invariablemente contaminada por metales pesados. Las concentraciones más altas de Cd, Cr, Cu, Ni y Pb fueron registradas durante el invierno y la de Zn durante el verano. Las concentraciones generales de metales pesados en el agua mostraron la tendencia Zn > Ni > Cr > Pb > Cu > Cd. Las concentraciones de todos los metales pesados fueron altas en las estaciones de muestreo ubicadas corriente abajo. Un análisis de correlación mostró que la concentración de metales pesados en el agua de la corriente media tuvo una relación positiva significativa con la tasa de deposición atmosférica en los sitios respectivos. Si bien las concentraciones de estos metales en el agua permanecieron debajo de los límites permisibles de los estándares para el agua potable en la India, los niveles de Cd, Ni y Pb en tres estaciones superaron las concentraciones máximas admisibles (CMA) recomendadas internacionalmente (OMS). Estas observaciones sugieren que el uso de esta agua para consumo humano puede conducir a riesgos potenciales de salud a largo plazo. El estudio tiene además relevancia para el entendimiento de la interacción atmósfera - agua en un ambiente contaminado y para el manejo de cuerpos de agua, incluso de aquellos que se sitúan lejos de descargas antropogénicas directas.

Resumo: Investigámos a influência dos metais pesados transportados pelo ar na qualidade do caudal da água do rio Ganga em Varanasi, Índia. Doze estações amostra foram seleccionadas

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ao longo de 20 km do percurso do rio. As amostras de água da sub-superfície da corrente do rio foram colhidas quinzenalmente de todas as estações, foram digeridas em meio ácido e analisadas em relação ao Cd, Cr, Cu, Ni, Pb e Zn. Os dados revelaram que as águas da corrente do rio Ganga, em Varanasi, estavam invariavelmente contaminadas por metais pesados. As concentrações mais elevadas de Cd, Cr, Cu, Ni e Pb foram registadas durante o inverno enquanto no verão era o Zn. A concentração global em metais pesados na água mostrou a seguinte tendência: Zn > Ni > Cr > Pb > Cu > Cd. As concentrações de todos os metais pesados eram mais elevadas nas estações amostra mais a jusante. A análise de correlação mostrou que a concentração em metais pesados na corrente apresentava uma relação positiva com a taxa de deposição atmosférica nas respectivas estações. Embora as concentrações neste metais pesados na água se encontre abaixo dos limites permissíveis pelos standards indianos para a água de beber, os níveis de Cd, Ni e Pb em três estações, encontravam-se acima dos níveis internacionalmente recomendados (OMS) para as concentrações máximas admissíveis (CMA). Estas observações sugerem que o uso desta água para beber pode conduzir a longo prazo a potencias riscos para a saúde. O estudo tem, ainda, relevância para a compreensão da inter-acção atmosfera - água em ambientes poluídos e para a gestão das massas de água, mesmo daquelas distantes das descargas antropogénicas directas.

Key words: Aerial catchment, atmospheric deposition, contamination, Ganga river, heavy metals, pollution.

Introduction

Atmospheric depositions of heavy metals have dramatically altered the biogeochemical cycles of these elements (Azimi *et al.* 2003; Friedland & Miller 1999). The impact of long - distance atmospheric transport of pollutant aerosols on terrestrial environment is well documented, but the impact of aerially driven pollutant aerosols on inland water bodies has received attention only recently (Pandey & Pandey 2009a; Thornton & Dise 1998). In India, the data so far available on these lines have been mainly confined to acidic depositions (Kumar *et al.* 2001). Less attention has been paid towards atmospheric deposition - linked heavy metal contamination of freshwater bodies, especially rivers. During recent years, anthropogenic activities have dramatically increased the atmospheric deposition of pollutant elements even in areas far away from direct human influence (Pandey & Pandey 2005, 2009a). Although the atmospheric deposition of pollutant aerosols is of global concern, this problem is rapidly increasing in developing countries including India due to continued population pressure coupled with accelerated urban - industrial growth and lack of efficient control measures (Borbely - Kiss *et al.* 1999; Pandey & Pandey 2009b; Singh & Agrawal 2005).

Heavy metals are known to have serious health implications including carcinogenesis induced tumor promotion (Schwartz 1994). The growing consciousness about the health risks associated with environmental chemicals has brought a major shift in global concern towards prevention of heavy metal accumulation in soil, water and vegetables. Atmospherically driven heavy metals have been shown to significantly contaminate soil and vegetables causing a serious risk to human health when plant based foodstuffs are consumed (Pandey & Pandey 2009b, 2009c; Voutsa *et al.* 1996). Dietary intake of trace elements depends also on irrigational water use. There may not always be a strong relationship between the concentrations of trace elements in soil and plants (Siegel 2002), but there always exists a strong relationship between their concentrations in irrigational water and plants (Ahmad & Goni 2009; Sharma *et al.* 2006). Thus, the deposition of heavy metals in water bodies can doubly increase the human intake through food chain as well as through drinking water. Most of the surface discharge sources contaminate soil and water bodies under limited spatial range, aerial emissions being prone to long range transport, contaminate wider range of ecosystems especially down wind to emission sources. Furthermore, unlike surface discharge, where stream flow restricts mid stream contamination, atmospheric deposition directly adds contaminants on water surfaces.

The present study was an attempt to investigate the relationship between the concentrations of heavy metals in mid stream Ganga water and their input through atmospheric deposition at Varanasi. The River Ganga rises in the Gangotri glacier in the Himalaya Mountains at an elevation of 7138 m above mean sea level. It enters the plains at Hardwar and meanders over a distance of above 2290 km across UP, Bihar and West Bengal, and receives a wide array of chemical constituents including toxicants from a variety of natural and anthropogenic sources (Kar *et al.* 2008). Earlier studies have indicated urban sewage and industrial effluents to be the main factors responsible for deteriorating the Ganga water quality (Jain 2002; Singh & Singh 2007). Recent studies conducted in India have indicated long range atmospheric transport and deposition of trace elements and their possible role in contaminating terrestrial environment including agroecosystems (Pandey & Pandey 2009b, 2009c; Sharma *et al.* 2008; Singh & Agrawal 2005). However, there is dearth of studies explicitly addressing the heavy metal contamination of fresh water resources, linked to atmospheric deposition. The present study is an attempt to investigate the magnitude of contamination of Ganga water at Varanasi in relation to atmospheric deposition of six important heavy metals.

Materials and methods

The study was performed for two years at selected sites along a 20 km long stretch of Ganga river at Varanasi from March, 2006 to February, 2008. The study area lies between 25° 18´ N latitude and 83° 1´ E longitude, in the midst of eastern Gangetic plain at 76.19 meter above msl. The climate of the region is tropical monsoonal. The year is divisible into a hot and dry summer (March - June), a humid rainy season (July - October) and a cold winter season (November - February). The ambient mean temperature was lowest in December (9.9 to 26.1 °C) and highest in June (27.8 to 40.9 °C). The rainy months remained warm and wet, with humidity reaching close to saturation. The day length is recorded as longest in June (about 14 hours) and shortest in December (about 10 hours). Wind direction shifts predominantly westerly and south - westerly in October through April and easterly and north - westerly in remaining months.

Twelve sampling stations were selected along a twenty kilometer long stretch of Ganga river and 3 sites were selected at each station. Description of different sampling sites is presented in Table 1. The bulk atmospheric deposition was collected using bulk samplers which combine the precipitation and dry fallout in the same container. These collection systems were maintained at a height of 2 m to avoid collection of re - suspended soil particulates and were devised to avoid bird nesting as described in Pandey & Pandey (2009b). The bulk collectors were replaced by clean ones at fortnightly intervals. As soon as the samples were brought to the laboratory, they were acidified with HNO_3 (Merck), filtered and stored in dark at ambient temperature before analysis.

Ternary acid digestion procedure was followed for extraction of heavy metals in atmospheric particulates (Allen *et al.* 1986). A 0.5 g of dried sample was digested with 15 ml of HNO_3, H_2SO_4 and $HClO_4$ in 5 : 1 : 1 ratio at 80 °C until a transparent solution was obtained. The solution was filtered through Whatman No. 42 filter paper and diluted to 50 ml with double distilled water. Water collectors were rinsed with nitric acid and mid stream water samples were collected from all the 12 sampling stations of river Ganga at a depth of 10 - 15 cm at fortnightly intervals. All water samples were brought to the laboratory and preserved with HNO_3. For the analysis of heavy metals, water samples (50 ml) were digested with 10 ml of concentrated HNO_3 at 80 °C until the solution became transparent (APHA 1987). The solution was filtered through Whatman No. 42 filter paper and diluted to 50 ml with double distilled water.

The concentrations of heavy metals in the filtrate of particulates and water samples were determined using Atomic Absorption Spectrophotometer (Perkin Elmer model 2130, USA), fitted with a specific lamp of particular metal using appropriate drift blank. The chemicals used were Merck analytical grade. Quality control measures were taken to assess contamination and reliability of data. Blank and drifts standards (Sisco Research Laboratory Pvt. Ltd., India) were run after five readings to calibrate the instrument.

Significant spatial and temporal variations were assessed using Analysis of Variance (ANO VA) following appropriate transformations whenever required. Correlation coefficients (r) were computed to test the linearity between heavy metal concentrations in water and their atmospheric depositions. Standard errors of means (SE) and coefficient of variation (CV) across time were computed for expressing data variability. The statistical analyses were done using SPSS Programme for micro - computers.

Table 1. Description of sampling sites.

S. N.	Sampling sites	Site code	Description
1.	Adalpura	Adlpr	2 km upstream to Sheetla Mata temple at Adalpura
2.	Bypass upstream	Bypus	2 km upstream to bypass (highway) bridge
3.	Bypass downstream	Bypds	Downstream bypass (highway) bridge
4.	Samne Ghat	Smnght	Near panton bridge connecting to Ramnagar
5.	Ravidas Park	Rvdpk	1 km upstream to Assi Ghat
6.	Assi Ghat	Asght	Near historical Assi Ghat
7.	Dandi Ghat	Dnght	1 km downstream to Assi Ghat
8.	Dashaswamedh Ghat	Dsmdght	Near historical Dashaswamedh Ghat
9.	Manikarnika Ghat	Mnkght	Near religious Manikarnika Ghat
10.	Lal Ghat	Laght	Near dye Industry
11.	Raj Ghat upstream	Rjghtus	Upstream to Malviya bridge
12.	Raj Ghat downstream	Rjghtds	2 km downstream to Malviya bridge

Results and discussion

The data on bulk atmospheric deposition (g ha^{-1} y^{-1}) of six heavy metals are presented in Table 2. Since inter - annual variations were less marked, the data presented here represent means of two consecutive years of study. Among the heavy metals studied, atmospheric deposition was highest for Zn (152.70 - 447.50 g ha^{-1} y^{-1}) and lowest for Cd (0.78 - 18.65 g ha^{-1} y^{-1}). Atmospheric deposition of Pb (3.6 - 107.34 g ha^{-1} y^{-1}) also remained high. On spatial scale, atmospheric deposition appeared higher at source oriented sites in comparison to non - source oriented locations, the value being lowest at Adlpr (site 1) and highest at Rjghtds (site 12). Variations between the sites for atmospheric deposition of heavy metals were significant (Table 2). The data collected in this study are comparable to those observed in earlier studies conducted at source and non - source oriented locations at Son-

Table 2. Atmospheric deposition of heavy metals at selected river sites of Varanasi. Values are mean ± 1SE (n = 48).

Sampling station	Atmospheric deposition (g ha^{-1} y^{-1})					
	Cd	Cr	Cu	Ni	Pb	Zn
Adlpr	0.78 ± 0.05	0.48 ± 0.03	2.25 ± 0.16	0.31 ± 0.02	3.60 ± 0.24	152.70 ± 11.45
Bypus	11.78 ± 0.76	5.20 ± 0.35	36.90 ± 2.66	4.20 ± 0.31	59.50 ± 4.12	332.70 ± 2.50
Bypds	16.25 ± 1.02	6.11 ± 0.38	47.85 ± 3.10	5.31 ± 0.29	84.95 ± 6.30	369.48 ± 2.69
Smnght	13.27 ± 0.67	6.00 ± 0.33	42.66 ± 2.92	5.05 ± 0.33	78.24 ± 5.22	337.15 ± 2.94
Rvdpk	12.00 ± 0.68	6.25 ± 0.35	57.66 ± 4.55	4.15 ± 0.28	75.50 ± 6.32	298.1 ± 16.52
Asght	13.65 ± 0.84	6.10 ± 0.47	44.85 ± 3.35	5.10 ± 0.31	76.95 ± 4.69	345.20 ± 3.15
Dnght	10.50 ± 0.72	5.33 ± 0.26	63.56 ± 4.95	5.55 ± 0.31	77.96 ± 6.51	316.3 ± 18.61
Dsmdght	12.92 ± 0.93	5.89 ± 0.38	45.18 ± 2.96	4.96 ± 0.27	81.56 ± 5.38	319.40 ± 2.66
Mnkght	10.17 ± 0.67	4.50 ± 0.23	61.43 ± 4.27	5.26 ± 0.41	84.56 ± 6.20	317.1 ± 30.10
Laght	10.07 ± 0.62	6.00 ± 0.35	67.16 ± 4.57	5.80 ± 0.35	85.33 ± 6.41	318.45 ± 26.05
Rjghtus	18.65 ± 1.26	6.27 ± 0.44	57.33 ± 3.76	5.78 ± 0.32	107.34 ± 7.85	447.50 ± 3.18
Rjghtds	18.26 ± 1.39	6.28 ± 0.39	57.68 ± 4.15	5.90 ± 0.37	106.90 ± 9.26	440.61 ± 3.24
F ratio	67.35*	31.11*	34.15*	59.24*	107.26*	34.61*

*P < 0.001

bhadra (Singh & Agrawal 2005), Varanasi (Sharma *et al.* 2008) and Udaipur (Pandey & Pandey 2009b).

Concentrations of heavy metals in mid stream water showed almost similar spatial trends (Figs. 1 and 2) as observed for atmospheric deposition.

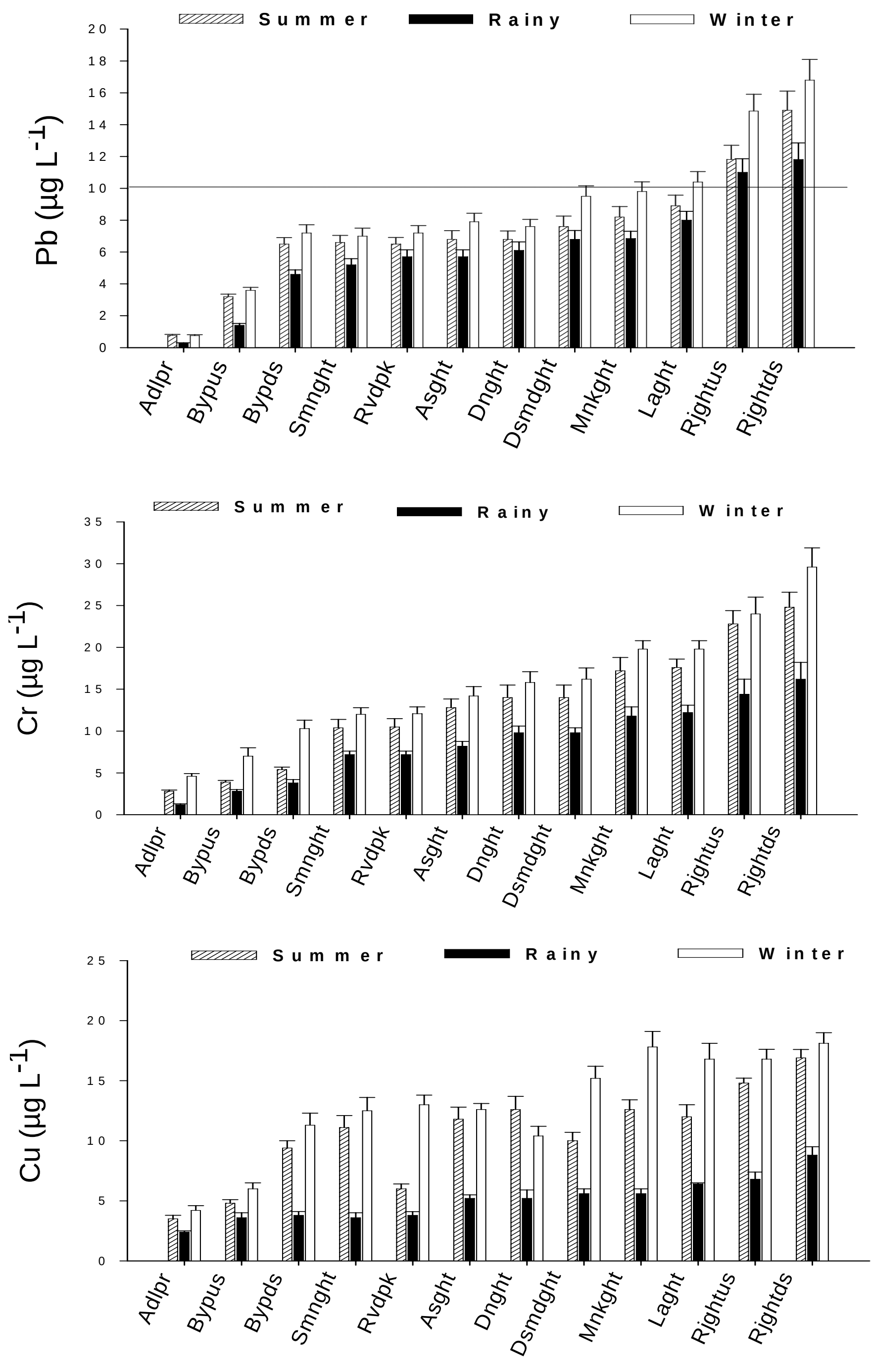

Fig. 1. Mid stream water concentrations of heavy metals in Ganga river at Varanasi. Values are mean (n = 48) ± 1SE. Values not appeared in the figure are below detectable limits. Horizontal line indicates internationally recommended maximum admissible concentration.

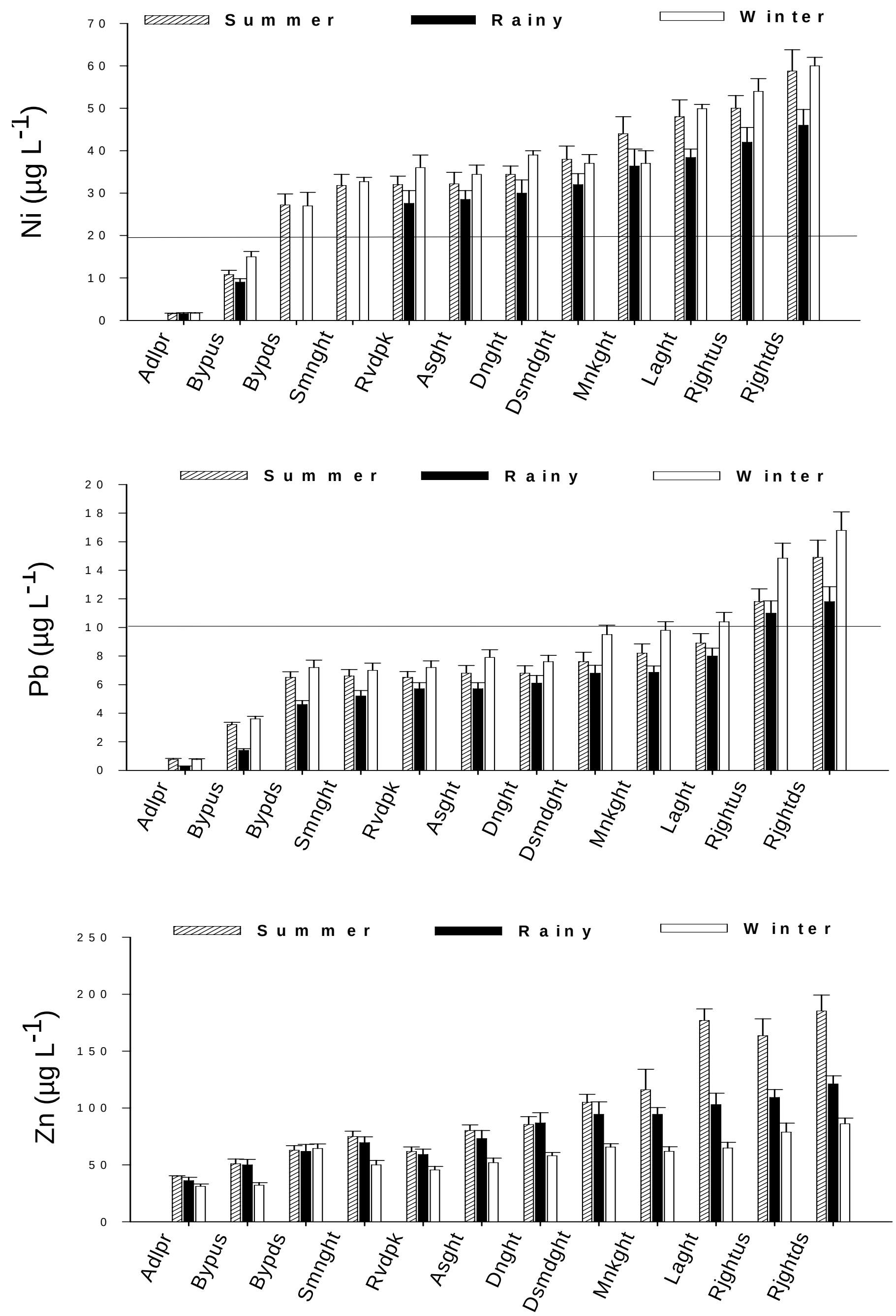

Fig. 2. Mid stream water concentrations of heavy metals in Ganga river at Varanasi. Values are mean (n = 48) ± 1SE. Values not appeared in the figure are below detectable limits. Horizontal lines indicate internationally recommended maximum admissible concentration.

With few exceptions, corresponding to atmospheric deposition, concentrations of all six metals increased almost consistently down stream from site 1 (Adlpr) to site 12 (Rjghtds). Mid stream water concentration was recorded maximum for Zn (31.2 - 185.2 µg L^{-1}) and minimum for Cd (ND - 8.4 µg L^{-1}). Concentration of Ni (1.6 - 60.8 µg L^{-1}) also appeared relatively higher (Fig. 2). Similar observations have

Table 3. F ratio obtained from two - way analysis of variance (ANOVA) for heavy metal concentrations in Ganga water.

Source of variation	Cd	Cr	Cu	Ni	Pb	Zn
Site (s)	57.30**	23.67**	18.52**	26.50**	71.33**	16.90**
Season (s)	48.21**	16.40**	11.90**	17.22**	42.80**	4.60*
S × s	32.08**	7.64**	95.00**	9.68**	33.65**	3.38*

*P < 0.01; ** P < 0.001

been made by Eder & Offiong (2002) for Lower Cross River Basin, Southeastern Nigeria. With respect to season, except for Zn, metal concentrations in water were recorded highest during winter followed by summer and rainy seasons. Analysis of variance indicated significant effects of site, season and their interaction for all heavy metals (Table 3).

The range of concentrations observed during the present study was below the permissible limits of Indian standard for drinking water (BIS 1991). Furthermore, the levels of Cr (1.2 - 29.6 μg L^{-1}), Cu (2.4 - 18.1 μg L^{-1}) and Zn (31.2 - 185.2 μg L^{-1}) in mid stream water of all the twelve locations were well below their internationally recommended (WHO) maximum admissible concentrations (MAC) of 50, 1000 and 5000 μg L^{-1}, respectively (WHO 1996). However, concentrations of Cd and Ni at 11th and 10th locations, respectively were higher than their respective MAC of 3.0 and 20 μg L^{-1}. Similarly, 22 % of the total water samples showed Pb concentration greater than MAC value (10 μg L^{-1}). The Indian standards (BIS or CPCB standards) show high deviations from WHO standards for environmental quality including drinking water (Bharti 2007). Heavy metals are highly toxic and hence such contaminations have serious human health associated risks.

Correlation analyses indicated significant ($p < 0.01$) relationships between mean concentrations of Cd, Cr, Cu, Ni, Pb and Zn in water and their respective rates of atmospheric depositions (Table 4). This indicates that the atmospheric deposition is the main contributory variable for elevated concentrations of heavy metals in the mid stream water. Similar observations have been made by Thornton & Dise (1998) in a number of streams in English Lake District of Cumbria. The total aerial metal input which was computed as Cd + Cr + Cu + Ni + Pb + Zn, ranged from 160.12 g ha^{-1} y^{-1} (Site 1, Adlpr) to 643.35 g ha^{-1} y^{-1} (Site 12, Rjghtds). A corresponding trend in total metal concentration in water (35.10 - 318.10 μg L^{-1}) is indicative of the role of aerially driven input of heavy metals in mid stream water contamination. Temporal variability measured in terms of coefficient of variation remained higher (cv = 0.122 - 0.336) for mid stream heavy metal concentration in comparison to atmospheric deposition (cv = 0.115 - 0.326). However, the differences between mean cv values of these two functions were not significant. This further supports the view that the atmospheric deposition is the main contributor to mid stream water contamination.

The main sources of these elements into the atmosphere are urban - industrial emissions in adjoining areas. Vehicular emission is the main contributor to urban particulates (Clarke *et al.* 1996). Re-suspension of the land - deposited particulates could further substantiate atmospheric loading and deposition of heavy metals (Pandey & Pandey 2009b). Predominant south westerly wind has further raised the atmospheric loading and transport of heavy metals towards down wind locations. These inputs are in addition to the urban flushing and surface run off being added to the river. Since stream flow restricts cross stream lateral mixing of surface - fed materials, atmospheric deposition can be considered as the major factor responsible for mid stream heavy metal contamination of Ganga river. A comparison of Adlpr site with the rest of the sampling locations shows that more than 86 % of mid stream water samples of the latter registered about 1.5 to 36 fold increase in heavy metal concentrations under the influence of anthropogenic emission sources. Except for Adlpr (site 1), the remaining 11 locations fall into medium to high concentration zones (Eder & Offiong 2002). Significant variations between site and down stream rising levels of heavy metals indicated the cumulative effect of stream flow and atmospheric deposition.

The results presented in this study clearly indicate that Ganga river at Varanasi receives sizably high atmospheric inputs of Cd, Cr, Cu, Pb and Zn. Mid stream water concentrations of metals showed significant correlations with their atmospheric depositions. The study suggests that the aerial

Table 4. Correlation coefficient (r) values for heavy metal concentration in water and atmospheric deposition.

Metals in water	Metals in atmospheric deposition					
	Cd	Cr	Cu	Ni	Pb	Zn
Cd	0.543**					
Cr	0.361	0.480**				
Cu	0.425*	0.342	0.512**			
Ni	0.187	0.410*	0.361	0.461*		
Pb	0.436*	0.409*	0.415*	0.392	0.567**	
Zn	0.162	0.269	0.318	0.408*	0.236	0.428*

*P < 0.05; **P < 0.01

catchment can sizably contribute to surface water contamination of potentially toxic metals. Our observations form the first report on atmospheric deposition - linked mid stream water contamination of Ganga river. Although the concentrations of these metals in water were below the permissible limits of Indian standards, more than 80 % of water samples of 11 out of twelve stations considered in this study, showed Cd and Ni levels above the internationally recommended maximum admissible concentrations. This invites serious attention from human health perspective since Ganga water is also used for drinking by a large population.

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