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Development of New Trends in Removing Heavy Metals from Yamuna River in India

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A B S T R A C T

Heavy metal concentrations in river water and sediments depend on not only industrial and domestic waste inputs but also on the geochemical composition of the area. The study reveals that there is a considerable variation in the concentration of heavy metals in water and sediments samples. These variations may be due to the change in the volume of industrial and sewage waste being added to river at different sampling stations. The concentrations of heavy metals in sediments were found considerably higher than those measured in river water. The metal pair ratios clearly reflects maximum enrichment of all studied metals (Cd, Cr, Cu, Mn, Ni, Pb and Zn) at in Yamuna river water site S-4 followed by site S-5 for Cd, Cr, Cu and Zn, while site S-6 for Mn, Ni and Pb. It is unpolluted to moderately pollute with Pb. Activated carbon is complex materials, which cannot be characterized by chemical analysis or structural formula. Every carbon is unique with its physical and chemical characteristics, a direct result of its parent material, any pretreatment used and the temperature and nature of the activation whether it be chemical, physical or combination of both. Information on chemical structure was obtained by recording the infrared spectrum of the carbons in potassium bromide and Nujol mull in the range of 500 - 4000 cm^{-1} using a Perkin- Elmer spectrophotometer. The mercury porosimetries have been carried out with a Quantachrome model Autoscan - 60 porosimeter. The mercury density was determined as usual, by carrying out the mercury porosimetery experiments.

Keywords: Water, Heavy Metals, Yamuna river, Ganga river, Pollution

A R T I C L E I N F O

CONTENTS

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1. Introduction

Water is fundamental resource is of such importance because no living organism can survive without water (Kupchella and Hyland, 1993). Therefore, there is a demand for clean, unpolluted water in substantial supply. As a result a prerequisite of sustainable development, therefore, must be to ensure uncontaminated streams, rivers, lakes and oceans (IIED, 2002). The geographical area of India is 3,287,590 km^2 with its coastline of about 7500 km. The climate of India varies from tropical monsoon in south to temperate in north. Its terrain has upland plain (Deccan Plateau) in south, flat to rolling plain along the Ganges, deserts in west, and Himalayas in north. India is enviably endowed in respect of water resources.

The country is literally criss-crossed with rivers and blessed with high precipitation mainly due to the southwest monsoon, which accounts for 75 % of the annual rainfall. There are thirteen major river basins (area more than 20,000 km^2) in the country, which occupy 82.4 % of total drainage basins, contribute 85 % of total surface flow and house 80 % of the country's population. Major river basins are Brahmaputra, Ganga (including Yamuna Sub Basin), Indus (including Satluj and Beas Sub Basin), Godavari, Krishna, Mahanadi, Narmada, Kauvery, Brahmini (including Baitarni Sub Basin), Tapi, Mahi, Pennar and Sabarmati. The classification of river basin based on catchment area is given in Table 1. There are few desert rivers, which flow for some distance and get lost in deserts. There are some complete arid areas where evaporation equals rainfall and hence no surface-flow. The medium and minor river basins are mainly in coastal area (Bhardwaj, 2005).

Table1. Classification of River Basin in India

River Basin	**Catchment Area – km^2 (%)**	**No. of Basin**
Major	> 20,000 (82.4)	13
Medium	2,000 - 20,000 (8)	48
Minor	< 2,000 (9.6)	52

Source: Bhardwaj (2005).

Heavy metal distribution and speciation/fractionation

Heavy metals are a special group of contaminants of water reservoirs. They are of high ecological significance since they are not removed from water as a result of self-purification, but accumulate in reservoirs and enter into the food chain. The elevation of metal levels in a reservoir is shown mainly by an increase in their concentrations in the bottom sediment. Their occurrence in the environment results primarily from anthropogenic activities, though natural processes that may enrich waters with trace metals also play a noticeable role (Forstner and Wittman, 1979; Nriagu, 1989). Metals are introduced into the aquatic system as a result of weathering of soil and rocks, volcanic eruptions and from a variety of human activities involving mining, processing and use of metals and/or substances containing metal contaminants bodies (Pardo et al., 1990; Boughriet et al., 1992; Yu et al., 2001; Klavins et al., 2000). River sediments are basic components of our environment as they provide nutrients for living organisms and serves as sink for deleterious chemical species. Riverine sediments play an important role as pollutants and they reflect the history of the river pollution (Jain, 2004). Sediments act as both carriers and sinks for contaminants in aquatic environments. Trace elements, especially the so-called 'heavy metals', are among the most common environmental pollutants and their occurrence in waters and biota indicate the presence of natural or anthropogenic sources. Numerous studies have demonstrated that the concentrations of heavy metals in suspended and bed sediments can be sensitive indicators of contaminants in hydrological systems (Salomons and Forstner, 1980; Luoma, 1990). The presence of heavy metals in sediments is affected by the particle size and composition of the sediments (Foster and Hunt, 1975; Throne and Nickless, 1981; Sakai et al., 1986).

The heavy metals can be either adsorbed onto sediments or accumulated in benthic organism, sometimes to toxic levels. Therefore, the mobility, bioavailability and subsequent toxicity of metals have been a major research area. (Ankley et al., 1996; Singh, 2001; Sharma et al., 1999; Davies et al., 1991; Gonzalez et al., 2000; Srivastava et al., 1994). Heavy metals are distributed between the aqueous phase and the suspended sediments during their transport. Riverine suspended load and sediments have important function of buffering heavy metal concentrations particularly by adsorption or precipitation (Forstner and Muller, 1973). More than 97% of the mass transport of heavy metals to the oceans is associated with river sediments (Jain and Sharma, 2001).

Most riverine studies dealing with metals associated with suspended matter or bottom sediments concern total metal concentration. Although the use of total concentration as a criterion to assess the potential effects of sediment concentration implies that all forms of a given metal have an equal impact on the environment; such an assumption is clearly untenable (Tessier et al., 1979). A study of different analytical extraction methods for non-detrital heavy metals in aquatic sediments has been conducted in Canada (Tessier et al., 1980), England (Jardo and Nickless, 1989; Neal et al., 1996), Egypt (Elsokkary and Muller, 1990), Lebanon (Korfali and Davies, 2000), Spain (Rauret, 1998; Pardo et al., 1990), North Greece (Fytianos and Lourantou, 2004) and Turkey (Akcay et al., 2003). Only a few studies have been reported on the speciation of metals in Indian rivers (Jha et al., 1990; Singh et al., 2000; Jain, 2004).

Chang et al. (1998), studied the distribution of heavy metals (including Cr, Co, Zn, Ni, Pb, Cu and Cd) using sequential extraction procedure (SEP) in bottom sediments from heavily polluted section of YenshuiRiver, located in the southern Taiwan. They found that distribution of heavy metals in depth profile of sediment was not identical among different sites. Levels of Zn, Cr, Cu and Ni were higher than other metals, and within the ranges of 30 - 200, 8 - 160, 5 - 130, and 10 - 100 mg/kg, respectively. They also found that the major binding forms of Zn, Cr and Cu in sediment were 'bound to carbonates', 'bound to Fe oxides' and 'bound to organic matter', respectively. And, the percentages of different heavy metal binding forms were not significantly varied in depth profile. Singh and Hasnain (1999), collected water and bed sediments sample from the Damodar River and its tributaries and analysed to study elemental chemistry and suspended load characteristics of the river basin. Dauvalter and Rognerud (2001), studied the heavy metals (Ni, Cu, Co, Zn, Cd, Pb, Hg) pollution in the sediments of the PasvikRiver. On the basis of sediment investigations at 27 stations of the watershed, background concentrations of the heavy metals, vertical distribution of heavy metals in sediments, heavy metal concentrations in surface sediments, contamination degree, and risk index were determined. The atmospheric emissions of Ni, Cu, Co, Zn, Cd and Hg from the smelters and waste waters from tailing dams and mines of the Pechenganickel company was likely to be the main sources of increasing concentrations observed in recent sediments of the lower river reaches. Lead showed a different pattern from the other heavy metals increasing Pb concentrations in the upper sediment layers towards the Norwegian side.

The distribution of trace metals (Cu, Zn, Fe, Mn, Cd, Cr, Pb, and Ni) in water, suspended and bed sediments of the RiverHindon, a highly polluted river in the western Uttar Pradesh (India) was studied by Jain and Sharma (2001). They observed that the heavy metal concentrations in water were depend largely on the amount of flowing water and was negatively correlated with flow. Sediment analysis indicates that the large amount of heavy metals was associated with organic matter, the fine-grained sediment fraction and Fe-Mn oxides. A high positive correlation of most of the metal ions in sediments with Fe, Mn and organic matter indicate that these constituents play a major role in metal ion transport.

Lower metal concentrations in bed sediments during post-monsoon season established that monsoon had a slight effect on status of metals in sediments by causing renewal and mobilization of metals from the sediments. Kuang-Chung et al. (2001), studied the spatial distribution of acid volatile sulfides (AVS), simultaneously extracted metals (SEM), and other binding phases of heavy metals in anoxic sediments of the Ell-Renriver. By comparing the spatial distributions of SEM/AVS ratio with various binding phases in extremely anoxic sediments (redox potential was between-115 and -208 mV), both organic matter and carbonates could be considered to be the main additional binding phases of SEM other than AVS. In addition, AVS appeared to have the priority to bind with SEM. By comparing the binding phases of heavy metals before and after AVS extraction, they found that Fe-oxides could also be considered to be the main additional binding phase associated with Zn in slightly anoxic sediments (redox potential was between -50 mV and -130 mV), while organic matter with Cu being the next.

Characterization and spatial distribution of heavy metals in sediment from Cedar and Ortega rivers sub basin was studied by Ouyang et al. (2002). They investigated the characteristics and spatial distribution of heavy metals, including Pb, Cu, Zn and Cd, from sediments in the sub basin using field measurements and three-dimensional kriging estimates. Sediment samples collected from three sampling depth intervals (i.e., 0 - 0.10, 0.11- 0.56 and 0.57 - 1.88 m) in 58 locations showed that concentrations of Pb ranged 4.47 - 420.0, Cu 2.30 - 107.0, Zn 9.75 - 2,050.0, and Cd 0.07 - 3.83 mg/kg dry weight, respectively. Kriging estimates showed that Pb, Cu and Cd concentrations decreased significantly from the sediment depth of 0.10 to 1.5 m, whereas Zn concentrations were still enriched at 1.5 m. It further revealed that the Cedar River area was a potential source area since it was more contaminated than the rest of the sub basin. Comparison of aluminum (Al) - normalized metal concentrations indicated that most of the metals within the top two intervals (0 - 0.56 m) had concentrations exceeding the background levels by factors of 2 - 10.

2. Materials and Methods

Study area:

The Yamuna River, one of the major tributaries of the Ganga river system in northern India has been selected for this study. It has been acclaimed as a holy river in Indian mythology and various pilgrimage centers e.g. Yamunotri (Uttaranchal), Paonta Sahib (Himachal Pradesh), Mathura, Vrindavan, Bateshwar & Allahabad (all in Uttar Pradesh) are located at the banks of this river. Large urban centers e.g. Yamuna Nagar, Sonepat, Delhi, the political nucleus of India, GautamBudh Nagar, Faridabad, Mathura, Agra and Etawah are also established on its banks. Large industrial centers have also been developed either on banks or in its basin.The total length of Yamuna River from origin at SaptrishiKund to its confluence with Ganga at Allahabad is 1376 km traversing through five states.

The main stream of river originates from the Yamunotri glacier (Saptrishi Kund) near Bander punch peaks (38^0 59' N $78^0$27'E) in the Mussoorie range of the lower Himalayas at an elevation of about 6320 meter above mean sea level in Uttarkashi district of Uttaranchal. The head waters of Yamuna river are formed by several melt streams, the chief of then gushing out of the morainic smooth at an altitude of 3250 m, 8 km North West of Yamunotri, hot springs at the latitude 31^0 2'12" N and longitude 78^0 26' 10". Arising from the source, the river flows through series of curves and rapids for about 120 km to emerge into Indo-Gangetic plains at Dak Patthar in Uttaranchal. Downstream of Hathnikund the river regains water from ground water accrual and contributions of feeding canals and small tributaries etc. From Hathnikund the river sluggishly meanders and reaches Delhi at Palla after travelling a distance of about 224 km. In the present study, total ten sites (Figure 3.1), namely Bagpat (Site-1), Sonipat (Site-2), Pallo (Site-3), Nizamuddin bridge (Site-4), Palwal (Site-5), Agra canal (Site-6), Agra upstream (Site-7), Agra downstream (Site-8), Bateshwar (Site-9) and etowh (Site-10) were selected to assess the impact of localized domestic and industrial effluents and the heavy metal concentrations and their speciation forms in sediments of the yamuna river.

The first two sites (1-3) are located in the area of relatively low river pollution. Other four sites (3-6) are located in the region of high river pollution as there are a number of wastewater drains and two highly polluted tributaries emptying into the river in this stretch. The last four sites (7-10) are in the downstream region of moderate pollution as the river considerably recovers in the course.

Sampling and analysis of river water

Grab water samples were collected from 30 cm below the water surface at three points (1/4, 1/2, 3/4) across the river width at all the 10 sites during the study period (March, 04; June, 04; Sept., 04; Dec., 2011; March, 05; June, 05; Sept., 05 and Dec., 2012). Water samples were collected in high quality polyethylene bottles previously soaked and washed with 10 % nitric acid and double distilled water (Duncan and Harrison, 1981). The samples were transported to laboratory in ice-box under low temperature conditions (APHA, 1998). The river water temperature was measured at the time of sampling using Mercury Thermometer and pH was measured in the laboratory using glass electrode pH meter (Metrohm, Model No. 740) (Singh et al., 2005). For the 'total metal' estimation river water samples were acidified in the field with concentrated HNO_3 (5 ml/l of water sample, to reduce the pH of the sample, pH > 2.0). The total metal content was determined by digesting 200 ml of sample with a mixture of concentrated HNO_3 and $HClO_4$ acid (10 ml + 2 ml) (Singh et al., 2005). The digested samples were filtered through Whatman filter No. 42 and finally volume were made 10 ml with 0.1 N HNO_3 and analyzed for heavy metals using ICP-AES (Model Labtam-8440).

Sampling and analysis of river sediments

The bed sediments samples were collected from shallow water about 20 m from the bank at three points (1/4, 1/2, 3/4) across the river width at all the 10 sites during the study period ((March, 04; June, 04; Sept., 04; Dec., 2011; March, 05; June, 05; Sept., 05 and Dec., 2012), using grab sampler and kept in polythene bags. The samples were air dried for removal of water contents associated with the sediments. The dried samples were then ground with pestle mortar, homogenized, sieved to below 100 mesh size, sealed in clean polythene bags and stored in refrigerator for characterization and speciation (Sakai et al., 1986).

Statistical analysis:

All statistical analysis [mean, maximum, minimum, standard deviation and Pearson correlation coefficients (r)] were performed by using Microsoft Excel and SPSS-10 for Windows.

Heavy metal adsorption/removal

Heavy metals in water have been a major preoccupation for many years because of their toxicity towards aquatic life, human beings and environment. As they do not degrade biologically as organic pollutants, their presence in industrial effluents or drinking water is a public health problem due to their absorption and therefore, possible accumulation in organisms. These toxic metals are released into the environment by different ways (coal combustion, sewage wastewaters, automobiles emissions, mining activities and the utilization of fossil fuels). These metals have a harmful effect on human physiology and other biological systems when they are found above the tolerance levels. Conventional methods for removal are chemical precipitation, chemical oxidation, chemical reduction, ion exchange, filtration, electrochemical treatment and evaporation. All these procedure present significant disadvantages, such as for instance incomplete removal, high-energy requirements, and production of toxic sludge or waste products also requiring disposal. These methods are often very expensive. Alternative methods for heavy metal removal were developed in the last decade (Mouflih et al., 2005). Adsorption with the selection of a suitable adsorbent can be a low-cost and effective technique for the removal of heavy metals from wastewater. (Huang et al., 1986; Huang and Corapcioglu, 1987; Huang and Rhoads, 1989; Huang and Morehart, 1990). These suggested adsorbents are activated carbon, alumina, silica and ferric oxide, which generally have high metal adsorption capacity, but are expensive and difficult to be separated from the wastewater after use. Activated carbon is a highly effective adsorbent that is widely used for water and gaseous emissions purification. This is due to their extended specific surface area between 500 and 2000 m^2/g, their high pore volume and the presence of surface functional groups, especially oxygen groups (Bansal et al., 1988). The removal of heavy metal, one of the major water pollutants, through adsorption process has been proved to be successfully by many researchers. Natural materials that are available in large quantities or certain waste products from industrial operations (Netzer et al., 1974; Hamadi et al., 2001; Youssef et al., 2004) and agricultural by-products (Kadirvelu et al., 2001; Ranganathan, 2000), may have potential as inexpensive adsorbents. Generally, adsorbents can be assumed as low cost if they require little processing, are abundant in nature, or are a by-product or waste material from another industry (Babel and Kurniawan, 2003).

3. Results and Discussion

Characteristics of river water

The characteristics of the river water during the study period are presented in the Table 2. The river water temperature during the study period ranged between 18.75-29.67°C. The minimum temperature was observed during the winter at site S-6 (18.75°C), and maximum during the summer at site S-2 (29.67°C). The river water pH was observed alkaline throughout the study period ranged between 8.08 – 8.62. The minimum pH was observed at site S-5, and maximum at site S-10.

Characteristics of river sediments

The physicochemical characteristics of the river bed sediments are presented in Table 3. The sediments of river Yamuna are characterized with alkaline pH between 7.30 and 8.24. The minimum pH was observed at site S-4 while maximum at site S-6. The organic matter content varied between 0.35 and 14.26 % at site S-1 and S-4 respectively.

Heavy metals in the Yamuna river system

The overall minimum, maximum, mean and standard deviation values of the heavy metals viz., Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn in Yamuna river water and bed sediments at different sampling sites during the study period are given in Table 4 and 5. Concentrations of different heavy metals in river water and bed sediments measured at three points (1/4, 1/2, 3,4) across the river width at each site. Iron normalized (M/Fe) heavy metal concentrations at different sampling sites in river sediments are presented in Table 6.

Cadmium (Cd) in river water

The overall minimum, maximum, mean and standard deviation values of Cd in Yamuna river water at different sampling sites during the study period are given in Table 4. Cadmium was detected only in 9.2 % river water samples. The overall values of Cd concentrations ranged between ND - 0.011 mg/l, and the highest mean value of Cd concentration was 0.001 ± 0.002 mg/l, found at site S-3, S-4 and S-10 during the study period. Concentrations of Cd measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 4.Spatial variations of Cd concentration in Yamuna river water.The highest mean value of Cd concentration was 0.007 mg/l, found at site S-4 in the month of Sept., 04. Cadmium was not detected in most of the river water samples. Cadmium concentration was found well below the BIS guideline value (0.01 mg/l) prescribed by BIS (1982) for Class-A, Inland surface waters, in all river water samples throughout the study.

Cd in river sediments

The overall minimum, maximum, mean and standard deviation values of Cd in Yamuna river sediment at different sampling sites during the study period are given in Table 5. Cadmium was detected in most of the sediment samples (99.1 %). The overall values of Cd concentration ranged between ND - 12.74 µg/g, and the highest mean value of Cd concentration was 5.01 ± 3.21 µg/g, found at site S-4 during the study period. Concentrations of Cd measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 5.Spatial variations of Cd concentration in Yamuna river sediments.The highest mean value of Cd concentration was 8.38 µg/g, found at site S-4 in the month of Dec.,04.

Chromium (Cr) in river water

The overall minimum, maximum, mean and standard deviation values of Cr in Yamuna river water at different sampling sites during the study period are given in Table 4. Chromium was detected in 44.6 % river water samples. The overall values of Cr concentrations ranged between ND - 0.194 mg/l, and the highest mean value of Cr concentration was 0.014 ± 0.029 mg/l, found at site S-9 during the study period. Concentrations of Cr measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 4.Spatial variations of Cr concentration in Yamuna river water. The highest mean value of Cr concentration was 0.087 mg/l, found at site S-10 in the month of Dec. 04.

Cr in river sediments

The overall minimum, maximum, mean and standard deviation values of Cr in Yamuna river sediment at different sampling sites during the study period are given in Table 5. Chromium was detected in all sediment samples. The overall values of Cr concentration ranged between 0.26 – 40.16 µg/g, and the highest mean value of Cr concentration was 17.66 ± 9.20 µg/g, found at site S-4 during the study period. Concentrations of Cr measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 5. Spatial variations of Cr concentration in Yamuna river sediments. The highest mean value of Cr concentration was 23.46 µg/g, found at site S-5 in the month of Sept.,04.

Copper (Cu) in river water

Copper was detected in 60.5 % river water samples. The overall values of Cu concentrations ranged between ND – 0.101 mg/l, and the highest mean value of Cu concentration was 0.008 ± 0.021 mg/l, found at site S-3 during the study period. Concentrations of Cu measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 4 (a-b). Spatial variations of Cu concentration in Yamuna river water.The highest mean value of Cu concentration was 0.041 mg/l, found at site S-4 in the month of Jun., 04.

Cu in river sediments

The overall minimum, maximum, mean and standard deviation values of Cu in Yamuna river sediment at different sampling sites during the study period are given in Table 5. Copper was detected in most of the sediment samples (88.9 %). The overall values of Cu concentration ranged between ND–158.40 µg/g, and the highest mean value of Cu concentration was 22.46 ± 15.40 µg/g, found at site S-4 during the study period. Concentrations of Cu measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 5. Spatial variations of Cu concentration in Yamuna river sediments.

Iron (Fe) in river water

Iron was detected in most of the river water samples (78.3 %). The overall values of Fe concentrations ranged between ND – 5.956 mg/l, and the highest mean value of Fe concentration was 0.965 ± 1.763 mg/l, found at site S-6 during the study period. Concentrations of Fe measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 4.Spatial variations of Fe concentration in Yamuna river water. The highest mean value of Fe concentration was 4.961 mg/l, found at site S-6 in the month of Dec

Iron (Fe) in river sediments: Iron was detected in all the sediment samples. The overall values of Fe concentration ranged between 1724.20 – 15788.10 µg/g, and the highest mean value of Fe concentration was 7142.06 ± 4507.06 µg/g, found at site S-4 during the study period. Concentrations of Fe measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 5. Spatial variations of Fe concentration in Yamuna river sediments. The highest mean value of Fe concentration was 13974.77 µg/g, found at site S-4 in the month of Mar., 04.

Manganese (Mn) in river water

Manganese was detected in most of the river water samples (68.3 %). The overall values of Mn concentrations ranged between ND – 0.649 mg/l, and the highest mean value of Cd concentration was 0.054 ± 0.121 mg/l, found at site S-1 during the study period. Concentrations of Mn measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 4. Spatial variations of Mn concentration in Yamuna river water. The highest mean value of Mn concentration was 0.248 mg/l, found at site S-6 in the month of Jun., 04.

Manganese (Mn) in river sediments

Manganese was detected in all the sediment samples. The overall values of Mn concentration ranged between 37.18 – 641.40 µg/g, and the highest mean value of Mn concentration was 258.98 ± 130.27 µg/g, found at site S-4 during the study period. Concentrations of Mn measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 5. Spatial variations of Mn concentration in Yamuna river sediments. The highest mean value of Mn concentration was 450.27 µg/g, found at site S-4 in the month of Mar., 04.

Nickel (Ni) in river water

The overall minimum, maximum, mean and standard deviation values of Ni in Yamuna river water at different sampling sites during the study period are given in Table 4 Nickel was detected in most of the river water samples (71.3 %). The overall values of Ni concentrations ranged between ND - 0.089 mg/l, and the highest mean value of Ni concentration was 0.010 ± 0.019 mg/l, found at site S-10 during the study period.

Nickel (Ni) in river sediments

The overall minimum, maximum, mean and standard deviation values of Ni in Yamuna river sediment at different sampling sites during the study period are given in Table 5. Nickel was detected in all sediment samples. The overall values of Ni concentration ranged between 1.04 – 62.04 µg/g, and the highest mean value of Ni concentration was 24.56 ± 13.12 µg/g, found at site S-4 during the study period. Concentrations of Ni measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 5.

Lead (Pb) in river water

The overall minimum, maximum, mean and standard deviation values of Pb in Yamuna river water at different sampling sites during the study period are given in Table 4. Lead was detected in most of the river water samples (73.3 %). The overall values of Pb concentrations ranged between ND – 0.820 mg/l, and the highest mean value of Pb concentration was 0.044 ± 0.166 mg/l, found at site S-3 during the study period. Concentrations of Pb measured at three points (1/4, 1/2, 3/4) across the river width at each site .Spatial variations of Pb concentration in Yamuna river water. The highest mean value of Pb concentration was 0.273 mg/l, found at site S-3 in the month of Dec., 04. Lead concentration was found well below the BIS guideline value (0.1 mg/l) prescribed by BIS (1982) for Class-A, Inland surface waters, in most of the river water samples except at site S-3 (0.273 mg/l), S-7 (0.124 mg/l), S-8 (0.223 mg/l), and S-9 (0.224 mg/l) in the month of Dec., 04.

Lead (Pb) in river sediments

Lead was detected in most of the sediment samples (98.7 %). The overall values of Pb concentration ranged between ND–125.70 µg/g, and the highest mean value of Pb concentration was 57.35 ± 37.91 µg/g, found at site S-4 during the study period. Concentrations of Pb measured at three points (1/4, 1/2, 3/4) across the river width at each site are given in Table 5 (a-b). Spatial variations of Pb concentration in Yamuna river sediments. The highest mean value of Pb concentration was 108.97 µg/g, found at site S-4 in the month of Mar., 04 and 05.

Zinc (Zn) in river water

Zinc was detected in majority of the river water samples (69.2 %). The overall values of Zn concentrations ranged between ND - 0.366 mg/l, and the highest mean value of Zn concentration was 0.040 ± 0.088 mg/l, found at site S-1 during the study period. Concentrations of Zn measured at three points (1/4, 1/2, 3/4) across the river width at each site.

Zinc (Zn) in rivers ediments

Zinc was detected in most of the sediment samples (98.3 %). The overall values of Zn concentration ranged between ND–273.60 µg/g, and the highest mean value of Zn concentration was 85.61 ± 43.72 µg/g, found at site S-4 during the study period. Concentrations of Zn measured at three points (1/4, 1/2, 3/4) across the river width at each site. Spatial variations of Zn concentration in Yamuna river. The highest mean value of Zn concentration was 142.66 µg/g, found at site S-4 in the month of Jun., 04.

Discussion

Water temperature is important parameter in assessing the water quality as it plays a major role in the aquatic environment through influencing a variety of physicochemical and biological processes/parameters. Most chemical reactions involving dissolution of solids are accelerated by increased temperatures. The solubility of gases, on the other hand, decreases at elevated temperatures. Because biological oxidation of organics in streams and impoundments is dependent on an adequate supply of dissolved oxygen, decrease in oxygen solubility is undesirable. Temperature also affects other physical properties of water. The different temperature at different locations during the same seasons was due to the different sampling time on the same day. The hydrogen (H^+) and hydroxyl (OH^-) ions are controlling variables in aqueous systems as they influence both physico-chemical and biological processes in the aquatic environment. The equilibrium between these two ionic species is influenced by reaction with acids and bases introduced into the aqueous system. pH, an indicator of acidity is a measure of water's ability to neutralize base.

The sediment of river Yamuna is characterized with alkaline pH throughout the study period and ranged between 7.30 and 8.24. The organic matter content varied between 0.35 and 14.26 %. The organic matter was found maximum at the site S-4 and possibly originated from the discharge of sewage and plant materials from surrounding catchment areas. A higher percentage of silt in the sediment results in a very loose fabric with a very high porosity and high permeability, resulting in easy transportation of the sediment downstream (Singh et al., 2005). Many heavy metals of toxicological significance have low solubilities in the range of Eh and pH conditions found in natural waters and river sediments act as sinks for trace metals mobilized in the drainage basin (Salomons and Forstner, 1984). Bed sediment metal contents reflect the influence of catchment lithology, anthropogenic contamination and chemical reactions (precipitation, complexation, and adsorption) between the water column and the sediment particle surfaces. Transport, deposition, resuspension and solubilization of these metals in the fluvial system is dominated by hydrological processes and by the chemistry of the water column (Foster and Charles worth, 1996). The concentration of heavy metals in Yamuna river water ranged as, Cd (ND - 0.011); Cr (ND - 0.194); Cu (ND - 0.101); Fe (ND - 5.956); Mn (ND - 0.649); Ni (ND - 0.089); Pb (ND - 0.820); and Zn (ND - 0.366) mg/l, respectively. No definite and regular spatial or temporal trends could be observed for the different metals in river water. Most of the heavy metals viz., Cd, Cu, Mn, Ni, and Zn in Yamuna river water were found well below their respective BIS guideline (Inland surface water, Class A: 1982) values during study period. Cr, Fe and Pb. Cr level in Yamuna water were found exceeding their BIS guideline value (0.05 mg/l) at S-2, S-8, S-9 and S-10 in the month of June, 03, Mar., 04, and Dec., 03, respectively.

Table 2: Characteristics of sampling sites

Sites	District	[G]Water temperature îC			[G]pH
		Summer	Monsoon	Winter	
S-1	Bagpat	29.50	28.63	19.38	8.40
S-2	Sonipat	29.67	29.25	19.38	8.60
S-3	Palla	28.67	29.00	20.25	8.59
S-4	Nizamuddin bridge	29.33	29.00	20.50	8.33
S-5	Agra canal	28.33	28.75	20.13	8.08
S-6	Palwal	27.33	28.50	18.75	8.53
S-7	Agra upstream	29.00	29.63	19.75	8.54
S-8	Agra downstream	29.67	28.38	19.75	8.55
S-9	Bateshwar	28.67	27.63	19.38	8.60
S-10	Etawah	29.00	27.50	19.63	8.62

[G]Mean values of temperature and pH, measured during study period.

Table 3: Characteristics of Yamuna river sediments at different sites

Sites	pH	Sand (%)	Silt (%)	Clay (%)	Organic Matter (%)
S-1	8.10	81.07	7.40	11.53	0.345
S-2	8.13	78.30	9.90	11.80	0.707
S-3	8.14	82.40	5.87	11.73	0.862
S-4	7.30	55.17	24.53	20.30	14.257
S-5	7.90	76.43	6.13	17.50	8.068
S-6	8.24	79.40	7.73	12.87	2.207
S-7	8.10	81.27	8.77	9.97	1.396
S-8	8.17	80.10	9.00	10.90	1.345
S-9	8.04	77.40	12.17	10.43	1.431
S-10	8.11	79.00	11.25	9.75	1.534

Table 4. Overall minimum, maximum, mean and SD of heavy metals in Yamuna river water at different sites during March, 2011 to December, 2012

Sites		Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
		(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)	(mg/l)
S-1	Mean	-	0.007	0.005	0.757	0.054	0.005	0.008	0.040
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	ND	0.055	0.021	4.247	0.567	0.015	0.025	0.366
	SD	-	0.012	0.007	1.211	0.121	0.005	0.008	0.088
S-2	Mean	-	0.012	0.004	0.555	0.037	0.006	0.010	0.028
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	ND	0.194	0.028	3.697	0.199	0.041	0.041	0.327
	SD	-	0.039	0.008	0.942	0.061	0.009	0.010	0.072
S-3	Mean	0.001	0.005	0.008	0.395	0.016	0.006	0.044	0.018
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	0.011	0.053	0.101	2.189	0.056	0.026	0.820	0.086
	SD	0.002	0.013	0.021	0.599	0.019	0.008	0.166	0.020
S-4	Mean	0.001	0.005	0.003	0.255	0.019	0.007	0.009	0.016
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	0.010	0.064	0.019	2.089	0.194	0.044	0.043	0.096
	SD	0.003	0.015	0.004	0.453	0.041	0.011	0.013	0.023

S-5	Mean	-	0.003	0.004	0.473	0.022	0.007	0.015	0.025
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	ND	0.024	0.015	5.274	0.140	0.026	0.047	0.111
	SD	-	0.005	0.005	1.102	0.035	0.008	0.015	0.028
S-6	Mean	-	0.003	0.003	0.965	0.052	0.006	0.011	0.037
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	ND	0.039	0.014	5.956	0.649	0.020	0.030	0.334
	SD	-	0.008	0.004	1.763	0.134	0.007	0.010	0.068
S-7	Mean	-	0.011	0.004	0.153	0.023	0.007	0.026	0.022
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	ND	0.102	0.032	0.526	0.195	0.030	0.259	0.115
	SD	-	0.022	0.007	0.159	0.052	0.010	0.054	0.028
S-8	Mean	-	0.013	0.003	0.126	0.007	0.006	0.038	0.018
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	ND	0.170	0.008	0.893	0.088	0.029	0.494	0.066
	SD	-	0.036	0.003	0.204	0.018	0.008	0.101	0.020
S-9	Mean	-	0.014	0.004	0.118	0.006	0.008	0.039	0.022
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	ND	0.113	0.022	0.542	0.063	0.040	0.550	0.103
	SD	-	0.029	0.005	0.124	0.013	0.011	0.111	0.028
S-10	Mean	0.001	0.013	0.003	0.096	0.005	0.010	0.023	0.019
	Min.	ND	ND	ND	ND	ND	ND	ND	ND
	Max.	0.010	0.184	0.013	0.776	0.045	0.089	0.131	0.054
	SD	0.002	0.039	0.004	0.161	0.012	0.019	0.034	0.020

ND = Not detected.

Table 5: Overall minimum, maximum, mean and SD of heavy metals in Yamuna river **sediments** at different sites during March, 2011 to December, 2012

Sites		**Cd**	**Cr**	**Cu**	**Fe**	**Mn**	**Ni**	**Pb**	**Zn**
		(«g/g)	**(«g/g)**	**(«g/g)**	**(«g/g)**	**(«g/g)**	**(«g/g)**	**(«g/g)**	**(«g/g)**
S-1	Mean	0.85	6.42	6.62	4466.36	133.23	9.40	15.34	23.90
	Min.	0.15	1.12	ND	2008.20	66.68	1.04	ND	ND
	Max.	3.24	22.12	95.19	10708.10	279.40	24.90	53.30	102.88
	SD	0.60	5.29	19.18	2230.43	65.04	6.51	13.90	19.93
S-2	Mean	0.73	9.92	5.11	5302.40	172.99	9.97	17.65	22.82
	Min.	ND	1.88	ND	2168.20	65.90	3.12	ND	ND
	Max.	1.98	25.22	27.05	12348.10	427.40	34.24	67.50	51.50
	SD	0.62	6.25	6.30	2718.08	89.04	6.39	14.98	14.15
S-3	Mean	1.07	10.58	6.52	5958.96	187.24	13.62	25.34	41.79
	Min.	ND	0.26	ND	2848.20	65.08	4.17	2.97	8.60
	Max.	4.14	34.48	38.43	15788.10	641.40	62.04	124.50	273.60
	SD	0.94	7.36	8.88	3256.50	119.56	11.72	24.15	54.00
S-4	Mean	5.01	17.66	22.46	7142.06	258.98	24.56	57.35	85.61
	Min.	0.32	0.48	1.34	2348.20	60.48	7.61	3.83	17.42
	Max.	12.74	40.16	56.59	15428.10	551.40	52.84	125.70	219.98
	SD	3.21	9.20	15.40	4507.06	130.27	13.12	37.91	43.72
S-5	Mean	2.81	14.63	15.32	6124.06	186.82	15.96	35.14	58.83
	Min.	0.24	2.62	ND	1894.20	54.38	2.16	ND	3.88
	Max.	9.84	37.14	80.95	14108.10	350.20	39.64	103.40	184.04
	SD	2.19	9.71	17.10	3705.99	80.19	10.82	25.34	47.10
S-6	Mean	2.05	13.24	12.10	6746.23	220.65	19.79	43.30	49.50
	Min.	0.32	3.00	ND	1724.20	98.48	5.24	5.73	10.56

	Max.	5.16	31.42	93.86	13308.10	423.40	38.84	90.40	147.22
	SD	1.14	8.09	18.20	3932.24	80.19	10.51	28.64	32.93
S-7	Mean	1.20	9.93	10.87	5874.05	147.94	13.12	26.12	25.70
	Min.	0.27	2.96	ND	2888.20	98.40	2.23	0.50	0.28
	Max.	2.32	32.52	158.40	11388.10	307.40	29.04	56.10	51.93
	SD	0.71	6.73	31.89	2937.39	52.05	7.22	15.94	11.34
S-8	Mean	1.59	9.89	8.04	5790.27	139.36	14.03	28.29	34.50
	Min.	0.01	1.63	ND	2264.72	37.18	1.75	1.15	7.40
	Max.	4.16	27.64	92.01	12228.10	317.40	28.48	62.70	120.40
	SD	1.02	7.13	18.17	3280.45	69.32	8.18	16.99	25.56
S-9	Mean	1.56	8.64	9.54	5849.96	152.57	13.73	27.95	28.76
	Min.	0.14	1.66	ND	2743.78	52.42	1.96	2.40	ND
	Max.	3.24	20.12	72.79	12348.10	315.40	32.00	65.50	89.68
	SD	0.85	6.11	15.35	3639.09	70.78	9.85	19.11	18.18
S-10	Mean	1.48	8.16	8.37	5704.79	136.85	12.31	27.54	24.35
	Min.	0.21	1.02	ND	1953.96	48.28	2.46	3.00	8.60
	Max.	4.02	18.84	72.97	14228.10	363.40	34.48	75.30	49.82
	SD	0.91	6.30	15.77	3943.20	72.92	10.15	18.31	11.69

Table 6: Iron-normalized mean metal concentration [(M/Fe) x 10^3] values for Yamuna river sediments at different sites

Sites	Cd	Cr	Cu	Mn	Ni	Pb	Zn	
S-1	0.17	1.44	1.48	29.83	2.11	3.44	5.35	
S-2	0.14	1.87	0.96	32.63	1.88	3.33	4.30	
S-3	0.18	1.78	1.09	31.42	2.29	4.25	7.01	
S-4	0.70	2.47	3.14	36.26	3.44	8.03	11.99	
S-5	0.46	2.39	2.50	30.51	2.61	5.74	9.61	
S-6	0.30	1.96	1.79	32.71	2.93	6.42	7.34	
S-7	0.20	1.69	1.85	25.18	2.23	4.45	4.38	
S-8	0.27	1.71	1.39	24.07	2.42	4.88	5.96	
S-9	0.27	1.48	1.63	26.08	2.35	4.78	4.92	
S-10	0.26	1.43	1.47	23.99	2.16	4.83	4.27	

4. Conclussion

Heavy metal concentrations in river water and sediments depend on not only industrial and domestic waste inputs but also on the geochemical composition of the area. The study reveals that there is a considerable variation in the concentration of heavy metals in water and sediments samples. These variations may be due to the change in the volume of industrial and sewage waste being added to river at different sampling stations. Most of the heavy metals viz., Cd, Cu, Mn, Ni, and Zn in Yamuna river water were found well below their respective BIS guideline value prescribed for Inland surface waters, Class-A, BIS (1982) during study period. Cr, Fe and Pb. Cr level in Yamuna water were found exceeding their BIS guideline value (0.05 mg/l) at S-2, S-8, S-9 and S-10 in the month of Jun., 03; Mar., 04, and Dec., 03 respectively. Iron level frequently exceeded the guideline value (0.3 mg/l) at most of the locations.

The metal pair ratios clearly reflects maximum enrichment of all studied metals (Cd, Cr, Cu, Mn, Ni, Pb and Zn) at in Yamuna river water site S-4 followed by site S-5 for Cd, Cr, Cu and Zn, while site S-6 for Mn, Ni and Pb. In most of the cases the average total heavy metal (Cr, Cu, Fe, Mn, Ni and Zn) concentrations determined in Yamuna river sediments were found to be lower than the corresponding shale values except for Cd and Pb. The Cd concentration in the Yamuna river sediments was found to be more than 6 times higher (1.83 µg/g) than that of its shale (0.30 µg/g), while Pb concentration was 1.5 times higher (30.47 µg/g) than that of shale (20.0 µg/g). A comparison of the present study result with the other rivers reveals that the average concentration of Cd and Pb are high in the Yamuna river sediments. The results were found to be highly promising. Both equilibrium and kinetic studies were performed in batch mode because of its simplicity to determine various parameters necessary to establish the operation of fixedbed reactors. The Langmuir adsorption isotherm model as compared to the Freundlich model better fits the sorption data. Further, the nonlinear isotherm models better fit the data as compared to the linear ones. It can be concluded that the coconut shells/fibers commodity group will benefit from this research because it will add value to surplus byproducts. Carbon users would also be benefited because this material offers a viable alternative to coal based activated carbons. Thus, the studies presented here reveal that the derived low-cost activated carbons could be fruitfully employed as adsorbents for the removal of Cr(VI) and Cr (III) from water/wastewater without any sludge production.

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