Chromium pollution assessment of water in the Hindon River, India: Impact of industrial effluents

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Abstract
The objective of the study is to reveal the seasonal variations in the Hindon River water pollution with respect to chromium contamination. The study was carried out from December 2014 to January 2017 by selecting twelve sampling stations of River Hindon. To get the extend of trace and ultratrace chromium contamination, water samples were collected from twelve different sites along the course of the River and its tributaries for summer and winter seasons. Higher concentrations of chromium (0.096mg/L) in summer and (0.088) in winter at Mohannagar (Ghaziabad) were recorded, thus indicating very high pollution of chromium at this sampling station. The concentration of chromium was determined using Atomic Absorption Spectrophotometer (AAS). In the past decades the increased use of chromium in several anthropogenic activities and consequent contamination of soil and water have become an increasing concern. Cr exists in several oxidation states but the most stable and common forms are Cr(0), Cr(III) and Cr(VI) species. Cr (VI) is a notorious environmental pollutant because it is a strong oxidant and much more toxic than Cr (III) and also carcinogenic.

Keywords- River pollution, Chromium, Hindon River (India), assessment, water pollution

Introduction
The Hindon River, historically known as the Harnandi River, has been a major source of water to the highly populated and predominantly rural population of Western Uttar Pradesh (India). The river was once considered to be so clean that its water was believed to cure the Kaali Khansi (bad cough). However, now the water quality of the River Hindon has been drastically deteriorated due to discharge of industrial as well as domestic sewage and due to application of chemical fertilizers and pesticides in agriculture. The heavy discharge of industrial effluents directly into the Hindon River at places has caused intolerable burden on the river’s natural ability to assimilate pollutants (Janhit Foundation 2007). The aquatic media such as lakes, Rivers, ponds, stream and coastlines are national wealth for any nation. Pollution is unavoidable harmful by product of industrial development (Khanna et al., 2014). The main sources of pollution in River include Municipal and Industrial (sugar, pulp and paper, Distilleries etc.) wastes flowing from Saharanpur, Muzaffarnagar and Ghaziabad areas. In India it is estimated that more than 8642X10⁷ m³ of wastewater is generated per annum from 212 class I cities and 241 class II towns. Only 23% of wastewater is being treated mostly at primary level prior to disposal and 77% untreated water is discharged on land (Bhutiani and Ahamad 2018; Rajeswaramma ,2002).The water quality gets further deteriorated due to confluence of River Kali and River Krishni as these rivers also receive toxic effluents from their surrounding industries and agricultural operations. The river is highly influenced of pollution due to discharge of industrial effluents, run off of surface water and Municipal sewage. A number of studies regarding pollution aspects of River Hindon and its tributaries have been carried out by different workers (Singhal et al. 1987, Khare 1994, Ruhela et al., 2017, Lokesh 1996, Jain and Ram 1997a, Jain and Sharma 2001, 2006, Jain et al., 2002, 2004a, 2005, Sharma 2001, Sharma et al., 2009a, Bhutiani et al., 2017, Sharma et al., 2010). In the present paper estimation of chromium at various sampling points
of river Hindon and characterization with respect to different point Sources contributing the pollution of chromium are discussed in this paper. The river originates from Upper Shivaliks (Lower Himalayas) and flows through six major districts, viz., Saharanpur, Muzaffarnagar, Meerut, Baghat, Ghaziabad and Gautambudh Nagar covering the distance of about 200 km before joining the river Yamuna at Tilwara village in Gautambudh Nagar. It serves as the major source of drinking water to the highly populated and predominantly rural population of Western Uttar Pradesh (India). It commands the area of about 5000 sq km of nearby towns of it catchment area of its flow region benefitting many villages and towns. In highly populated and predominantly rural catchment areas, the river is considered as main source for drinking, agriculture and industrial source of water. The recent rapid urban growth and industrial development along the route of river has raised many issues and conflicts due to pollution of river. Ghaziabad is a fastest growing industrial city in UP manufacturing industries of diesel engines, electroplating, bicycles, picture tubes, tapestries, glassware, pottery, vegetable oil, paint and varnish, heavy chains, automobile pistons and rings, steel, pharmaceuticals, liquor, etc. About 300 industrial units are located in Ghaziabad alone of which 60 units are situated along the Hindon river and around its catchment area and its two main tributaries, the Kali (West) and Krishni rivers. The Hindon receives hundreds of liters of urban wastewater and sewage water per day in Ghaziabad catchment area. In this way, the rapid industrialization and rapidly growing urbanization of Ghaziabad is the major reason and further threat to Hindon river pollution. In this study the efforts are made to evaluate the level of chromium metal pollution in water of River Hindon. The main goal of this study is to assess the impact of urban and industrial activities on chromium contamination in Hindon River.

Chromium (Cr) is the 17th most abundant element in the Earth’s crust (Avudainayagam et al., 2003). It occurs naturally as chromite (FeCr₂O₄) in ultramafic and serpentine rocks or complexed through its forms like crocoite (PbCrO₄), bentorite (Ca₆(Cr,Al)₂(SO₄)₃), and tarapacaite (K₂CrO₄), vaquelinite (CuPb₂CrO₄P(OH)₄), among others (Babula et al., 2008). Chromium is widely used in industry as plating, alloying, tanning of animal hides, inhibition of water corrosion, textile dyes, mordents, pigments, ceramics glazes, refractory bricks, and pressure-treated lumber (Avudainayagam et al., 2003). Due to this wide anthropogenic use of chromium, the consequent environmental contaminations have increased and have become an increasing concern since last several years (Zayed and Terry 2003). Chromium exists in several oxidation states, but the most stable and common forms are Cr (0), the trivalent Cr (III), and the hexavalent Cr (VI) species. Cr (0) is the metallic form, produced in industry and is a solid with high fusion point usually used for the manufacturing of steel and other alloys. Cr (VI) in the forms of chromate (CrO₄²⁻) dichromate (CrO₇²⁻), and CrO₃ are considered to be the most toxic forms of chromium. The Cr toxicity in plants depends on its valence state. Cr (VI) is being highly mobile and toxic as well while Cr (III) is less mobile and less toxic.

**Chromium in Water**

Chromium may enter the natural waters by weathering of Cr-containing rocks, direct discharge from industrial operations, leaching of soils, among others. A detailed review on the critical assessment of Cr in the environment has been published by (Kimbrough et al., 1999, Kotas and Stasicka 2000). The leather industry is the major source of high influx of Cr into water bodies and becomes source of entering in biosphere. This source alone accounts for over 40% of its industrial use (Barnhart 1997). In India, about 2000-32,000 tons of elemental Cr annually escapes into the environment from tanning industries. The recommended limits for Cr concentration in water are 8 μg L⁻¹ for Cr(III) and 1 μg L⁻¹ for Cr(VI). In the effluents in the vicinity of Cr industries the levels of Cr normally ranges from 2 to 5 g L⁻¹ (Chandra et al., 1997).

**Sources of Chromium**

Chromium is a naturally occurring element in rocks, soil, plants, animals, and volcanic dust and gases (ATSDR 1998). Environmental contamination due to Cr become a major issue primarily if there are high concentrations of chromium in soil and water bodies from the industrial and agricultural activities (Chanda and Parmar, 2003, Schiavon et al., 2008). Chromium contaminated soil and water ultimately destroys the crop and impart serious health hazards in human beings by entering through food chain (Vernay et
al., 2007). Cr (VI) species are released into the environment from a wide range of industries as discussed earlier and in addition due to timber processing, pulp and paper production and oxidative dyeing, tobacco smoke, and upon leaching from improper sanitary landfills (ATSDR 1998, Zayed and Terry 2003). In addition, Cr also enters water bodies upon natural leaching from topsoil and rocks. Cr(III) is used in leather tanning, nuclear and high temperature research for the treatment of cooling tower water, and in magnetic tapes, safety matches, photographic chemicals, drilling of mud and toner used in photocopy machines (ATSDR 1998). Various materials used to inhibit corrosion and cleaning water also enhance Cr (VI) content in potable waters (Sperling et al., 1992). Cr released into air upon coal combustion and emissions from Cr-based automotive catalytic convertors finally ends up in soils, (ATSDR 1998).

Sources of Pollution of River Hindon

The Hindon river originates in the lower Himalayas allocation named Purka Tanka village, situated in the upper east area of Saharanpur district. The main sources of pollution of river includes the Municipal Waste of the Saharanpur, Muzaffarnagar, Baghpāt, Ghaziabad and Gautambudh Nagar, districts and industrial effluents of sugar, pulp, paper, distilleries and other miscellaneous industries through tributaries as well as direct discharge from industries. In summer, the river dries up right from its origin to Saharanpur town. The effluents of Nagdev Nullah and of Star Paper Mill drain at Saharanpur together generate the flow of polluted water, discharged into river water. The Municipal Wastewater generated from the Saharanpur city is discharged to the Hindon river through Dhamola nullah. The industrial effluents from the Cooperative Distillery and Municipal Wastewater from Budhana town also join the river in this stretch. The river Kali, carrying industrial effluents of Shubham paper mill. It carries Municipal Wastewater and effluents of industries located in the Muzaffarnagar city. The Krishni River tributary has its confluence with the Hindon river at Barnawa village in Baghpāt district, also carrying substantial industrial effluents from four sugar mills, one dairy plant, one distillery and three paper mills located at Sikka. The Hindon receives the industrial effluents of one more sugar mill, i.e. Kinauni owned by Bajaj group. The lower region of Hindon river catchmen further receives heavy load of Municipal effluents of Ghaziabād district, through the three sewer drains. Next to enter the Hindon River at Greater Noida is the Municipal effluents flow diverted in through part of diverted canal at Mohannagar and falls into the Yamuna upstream of the Okhla barrage. Further at downstream a joins the Yamuna at Tilwara village in Gautambudh Nagar. The location of main source of pollution has also been indicated Figure 1.

Material and Methods

Study Area

The Hindon river is one of the important rivers in Western Uttar Pradesh (India), having a basin area of about 7083 km². The catchment area is a part of the Indo-Gangetic Plain, composed of Pleistocene and subrecent alluvium and it lies between latitude 28° 30’ to 30° 15’ N and longitude 77° 20’ to 77° 50’. The Hindon river covers a distance of about 200 km before joining the Yamuna River downstream near Delhi. The Ganga and Yamuna rivers are East and West bound to respectively to Hindon river. A general plan of sampling locations with respect to different outfalls of Municipal and industrial effluents in the Hindon river is shown in Figure 2. Twelve stations (HR1 to HR12) in the river were selected in order to obtain general information on the longitudinal and seasonal variation of chromium contamination along its course covering a total distance of 200 km.

Preparation of Samples and Analysis

Hindon river water samples were collected from twelve locations from sites of the river catchment area in both Summer and Winter seasons for a period of two years. Samples were collected in May, representing Summer season, and in January representing Winter season. At each station, three samples were collected from 1/3, 1/2 and 2/3 width of the river along the transect and mixed together to obtain a composite sample. All the samples were collected from the upper 15 cm of the water surface and stored in steam cleaned polyethylene bottles fitted with screw caps. The collected samples were filtered using Whatman No-42 filter paper and preserved in double distilled water with addition of few drops of 6N of HNO₃ and suitable aliquots.
Figure 1: Sources of Pollution of Hindon River
Chromium pollution assessment of water in the Hindon

Figure 2: The Hindon river system showing location of sampling sites
HR1-Khajnawar; HR2-Beherki; HR3-Santagarh; HR4-Nanandi; HR5-Maheshpur; HR6-Budhana; HR7-Atali; HR8-Barnawa; HR9-Daluhera; HR10-Surana; HR11-Mahannagar; HR12 Tilwara Village

were taken each time for analysis (APHA (Taylor et al., 2006). The precision of the analytical procedures, expressed as the relative standard deviation (rsd) ranged from 5 to 10% for different stations values of chromium. Precision for the
analyses of standard solutions was better than 5%. Average values of three replicate analysis were taken for each value confirmation. The chromium concentrations (mg/L) were recorded during present study.

Results and discussion
The chromium concentrations in Hindon river are represented in figure 3. Metals in dissolved forms are generally more toxic than metals in the inert or compound form. Toxic levels of Cr metal in water may impose serious threat to aquatic species as well as human. The release of toxic metal in biologically available forms by human activity may damage or alter aquatic ecosystems. In the present investigation Cr was estimated in water of river Hindon. The higher level of Cr concentration was recorded in winter season 0.096 mg/L at Mohannagar, Ghaziabad district station HR-11 and lowest in i.e. summer season 0.009 mg/l at Khajnawar Saharanpur district at HR-1 station. Sampling stations i.e. HR-10 (Surana) and HR-11 (Mohannagar) were the most polluted sites of Hindon in respect to Cr concentration. Our studies indicate that Cr metal level was relatively higher than previously reported. (Ajmal et al., 1987). Studied and heavy metal contamination in sediments of Hindon at Mohannagar. The higher ranges of metal in water clearly indicate the increasing Cr pollution level in Hindon river (Jain et al., 2005). Indicating that metal pollution load in Hindon is increased due to mixing of water of river Kali and Krishni, which also carries Municipal and Industrial effluents, (Rafael et al., 1990, Ruhela et al., 2017). The other researchers (Modak et al., 1992 and Vertacnik et al., 1995). Also observed similar trends in similarly situated river system. Our studies clearly indicated the impact of recent Industrial and Urban sewage on Hindon water quality. Earlier reports in Indian river sediments also reported in the range 115-817 mg/kg of Cr in river Ganga (Singh et al., 2002), and 228 - 273 mg/kg in sediments of Narmada river (Jain et al., 2008). Hexavalent form of Cr is known to cause wide range of human health effects including mutagenic and carcinogenic risks (Das and Mishra 2008). Interactions between metals and organic matter in bed sediment have often been recorded (Langston 1982, Wren et al., 1983, Stephenson and Mackie 1988, Rada et al., 1989, Coquery and Welbourn 1995). In general, the concentrations of dissolved chromium are found lowest in Winter months and highest during Summer months. The detection levels for different stations No HR-1 to HR-12 Cr pollution level in Hindon river in recent years were 0.009, 0.012, 0.042, 0.037, 0.057, 0.042, 0.041, 0.038, 0.056, 0.076, 0.088 and 0.056 mg/L for Summer season and 0.021, 0.017, 0.048, 0.041, 0.065, 0.047, 0.048, 0.044, 0.068, 0.072, 0.096 and 0.063 mg/L for Winter season respectively. In Mohannagar and Sahibabad Industrial areas (nearby to HR-11 sampling station) several metal plating and lead industries are located. As per the record of Uttar Pradesh Pollution Control Board (UPPCB) about 67 galvanizing/ electroplating, 12 chemical, 115 textiles dyeing, 27 lead processing, 11 frozen metal processing and 8 paper and pulp industries are located in Ghaziabad city and in nearby areas. Possibly effluents from these industries has directly or indirectly contributed to metal enrichments in Hindon river at Ghaziabad. Another possible metal contamination source could be due to discharge of Dusna drain (an industrial drain flow near to HR-11 station) which carries industrial wastewater from Dusna industrial area. Sampling station HR-11 showed relatively higher level chromium content (Fig. 3) and the chromium for different sampling sites of the river. It can be seen from the results that the station HR-3 at Santagarh, site at river Hindon just downstream of the confluence of Nagdev and Star Paper Mill drain is the most polluted site followed by station HR-7 Atali, site at river Hindon just downstream of the confluence of Kali river having enrichment due to metal coming from waters from all sites. The higher concentration of Cr metal in the upper section of the river may be linked to combined effluents of Nagdev Nallah, Star Paper Mill drain and Dhamola Nallah joining the river Hindon. The Nagdev nallah receives Municipal wastewater of the adjoining villages and Industrial effluents from various industrial units. Further downstream, higher Concentration of these metals may be attributed to the mixing of water of river Kali and Krishni, which carry Municipal and Industrial effluents of various types of industries of Muzaffarnagar region and sugar mill effluent from Shamli. The Star Paper Mill is located near Saharanpur railway station and manufactures several varieties of paper used in writing, printing, craft wrapping and wall papers. The raw materials
used in the manufacturing processes include wood, bamboo, jute sticks, straw, hemp, grass etc. The important chemicals used as raw materials by these industries are sodium sulphate, sodium hydroxide, sodium sulphide, sodium carbonate, calcium hypochlorite and magnesium bisulphate. The wastes from different processes and manufacturing units flow separately for some distance but finally join at one point and form the composite waste of pulp and paper mill before finally discharging in Hindon river. The composite effluent from the factory is discharged almost without any treatment into the river through an open canal. The canal is about 3 km in length and outfalls on the right bank of river Hindon near the village of Paragpur with a considerable force. Due to the presence of caustic soda and other alkaline mixtures, soapy and fibrous organies froth is continuously generated at the point of discharge. A characteristic smell of sulphate mercaptan hydrogen sulphide and methane is all pervading in the mixing zone and is strongly felt in the area. Due to these factors, dirty black subsoil with foul odour is also found in the nearby area and a dark brown colour is imparted to the river water, which allows easy distinction between the effluent and the river water. Hindon river is polluted due to industrial as well as discharge of sewage of urban area of Ghaziabad. However, the spatial variation in metal enrichments among sampling station indicates the point and non-point sources of heavy metals. HR-11 is located nearby (within 2–5 km territory) the Mohannagar and Sahibabad industrial area of Ghaziabad city. These are major industrial sites of Ghaziabad Manufacturers diesel engines, electroplating, bicycles, picture tubes, tapestries, glasswares, pottery, vegetable oil, paint and varnish, heavy chains, automobile pistons and rings, steel, pharmaceuticals, liquors, etc. The industrial units extracts a large volume of water from Hindon river for manufacturing processes, and also discharge considerable amount of industrial effluents into river; often with nominal or no treatment. In general, industrial effluents are the main source of Cr in water. The source of these metals in Hindon could be an industrial drain (flowing approximately 1 km far from catchment) which joins to main stream of Hindon river near Mohannagar. This industrial drain carries waste water from a distillery and beverage industries in Ghaziabad. Moreover, another industrial drain, i.e. Karehada drain carrying effluents form a textile industries also joins the main stream of Hindon at 3 km upstream of HR-11 station. The textile and distillery industries are classifies as most polluting industries in the country, and are responsible for severe degradation of surface water bodies of river system of India.

Figure 3: Experimental Levels of Chromium (mg/L) in water of different sampling sites of Hindon River for summer and winter season

Conclusion
Therefore these important studies revealed that the chromium pollution level in Hindon river has become alarming in the intent of process of Cr bioaccumulation indicating threat to human life and
Chromium pollution assessment of water in the Hindon

its serious impacts including as source of cancer like diseases. Results indicated relatively higher level of chromium in water at HR-10 (Surana) and HR-11 (Mohannagar) and different sampling stations. The river Hindon is subjected to varying degree of pollution caused by numerous untreated and or partially treated effluents discharge and Municipal and Industrial effluents. The river is highly influenced due to chromium metals pollution. Higher concentrations of chromium in river water in the upstream are largely due to the mixing of effluents from Star Paper Mill and Cooperative Distillery. The toxic pollutants from these sources will ultimately reach the ground water and enter in the food chain posing a threat to human health because of their carcinogenic nature.

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