STUDY ON DISTRIBUTION OF HEAVY METALS BETWEEN WATER AND SEDIMENT OF ULHAS RIVER FLOWING ALONG DOMBIVLI CITY NEAR MUMBAI

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Abstract : Ulhas river is highly polluted due to the addition of industrial wastes along Dombivli city located near Mumbai in Maharashtra state in India. Dombivli city has two industrial areas. MIDC Phase 1 and MIDC Phase 2. Industrial wastes from both these phases are added to Ulhas river waters. Hence the area of Ulhas river along Dombivli city to study the load of pollution due to addition of toxic heavy metals in river water was selected for the study. Distribution ratio of heavy metals Hg^{2+} , Cd^{2+} , Pb^{2+} , Cu^{2+} , Co^{2+} , Zn^{2+} , Cr^{6+} , Ni^{2+} and Fe^{2+} was studied for each season (Rainy, Winter and Summer) for the years 2012 and 2013. Results revealed that distribution ratio of heavy metals Pb^{2+} , Cu^{2+} , Fe^{2+} , Co^{2+} and Cr^{6+} increased in year 2013 as compared to the distribution ratio in year 2012. Distribution ratios were found to be maximum in Ni²⁺ in Year 2012 and 2013 and Minimum in Hg^{2+} in Year 2012 and 2013.

IndexTerms - water pollution; sediments; toxic heavy metals; distribution coefficient Kd; Ulhas River; industrial pollution; Dombivli; Mumbai.

I. INTRODUCTION

In today's world, we do not have any option to replace the usage of metals in our daily chores. It would not be an exaggeration to extend the statement that almost all the activities done by mankind involves the usage of metals and cannot be totally avoided. Metals have numerous applications. Metals have been used since ages by mankind. In the mid of nineteenth century, the industrialization of metals took a definite form and significantly grew in the industrial sector. Subsequent to the industrial revolution, the usage of metals also increased to a great extent. This in turn led to the increase in the generation of effluent waste, however with a greater proportion as compared to the waste treatment methods. The generated wastes were released to the natural bodies most often with no treatment. The major reason for this treatment bypassing activity was the incurring of additional expenditure required for the treatment of wastes. Though the major industries would treat the generated wastes, however the small scale industry would naturally not be inclined to treat the waste since their profit margins were very seldom. Though it is legally mandatory & binding on the part of industries to strictly adhere to the guidelines of the Central Pollution Control Board (CPCB) the environmental situation is far away from control. Many evidences related to industrial wastes mismanagement are available in India ^{1.6}. Thus, there has been a tremendous increase in pollution of water, soil and air. Most of the activities in the industries involve reactions and mixing of solid and liquid chemicals which in turn requires the equipment cleanings. Also in many of the reactions, mother liquor is also generated which is a waste. All these activities require cleaning with water, thus generating a large volume of waste water. This waste water is alleged dumped to the natural water bodies. Addition of large volume of contaminated and untreated waste water results to an increase in the concentration of pollutants including the heavy metals. These heavy metals are toxic to the life in water. Also the heavy metals settle to the bottom of the water system and pollute the sediments. Thus there is a significant impact on the quality of water and underlying sediments. In most of the stretches of almost all rivers are polluted by one or the other industry ⁷⁻¹³. After entering the aquatic environment these toxic heavy metals are adsorbed onto particulate matter. They may also form free metal ions and soluble complexes which are readily available for uptake by biological organisms ¹⁴. The metals associated with particulate material are also available for biological uptake ¹⁵, and are deposited in estuarine sediments ¹⁶. The toxic heavy metals in the sediments are taken up by the surrounding aquatic plants life. The toxic heavy metals in water and the aquatic water plants are taken up by the aquatic life in the water system. All the three i.e. aquatic water plants, fishes and the polluted water are used by mankind in their daily chores. Once deposited, binding by sulfides and/or iron hydroxides immobilises trace metals until a change in redox or pH occurs ¹⁷⁻¹⁸. The sediment present on the surface of bottom and to be specific the fine fraction accumulates the trace heavy metals and will provide a means to evaluate the accumulation of heavy metals over a long term period ^{17,19}. Thus they reach mankind and have severe ill effects on their health. In recent years, food chain contamination by heavy metals is a burning issue due to their potential accumulation in biosystems through contaminated water, soil and air ²⁰. In Maharashtra, near the Thane district, the Ulhas River is one such water body which has been subjected to water pollution from various industries during the course of its travel. The starting point is Sahyadri hills located near Khandala. The river further propagates down through industrial or MIDC areas of various cities such as Kalyan, Badlapur, Ambernath, Ulhasnagar and Ambivli and Dombivli from where it finally meets the Arabian Sea. The Ulhas River flows along Dombivli city. The Dombivli City is located near Mumbai in the Maharashtra State of India. Dombivli has been a part of the rapid industrialization. It has two industrial belts Phase I and Phase II. There are various industries in these two phases namely chemicals, pharmaceutical, textile, agrochemical, dyes manufacturing, engineering, metallurgical and paint manufacturing industries ranging from small scale to large scale located in these industrial areas. The industrial waste effluent is added to the waters of Ulhas River which flows along the city and add heavy pollution in the surrounding area ²¹⁻²⁹. In today's scenario, the study of accumulation of heavy metals in sediments and water is of utmost importance in order to understand the heavy metal sources in a better way ³⁰. Extensive studies were carried out over the years, on pollution in Ulhas river. As compared to other pollutants, toxic heavy metals in the environment are of serious concern due to their non-bio-degradable nature, accumulative properties and long biological half lives. These toxic metals not only pollute the creek waters but also pose a threat to the aquatic biota ^{31,32}. The increase in residue levels of heavy metal content in water, sediments and biota, will result in decreased productivity ²⁰ and increase in health risk in case of human beings ³³⁻³⁸. During the past few years, attempts were made to develop strategies directed towards more integrated approach in coastal environments ³⁹. Previous data on water pollution along creeks ^{20, 40-46} points out to the need of systematic and regular monitoring of pollution level for further improvement in the industrial waste water treatment methods. Hence in the present investigation we have studied the pollution load of trace and toxic heavy metals in water and sediment of Ulhas River along Dombivli City. Salient results of the investigations are presented and discussed in this paper.

II. EXPERIMENTAL

Materials and Methods

Area of study

The study was conducted out along Ulhas River where the effluents released from industries of Dombivli MIDC Industrial belt Phase I and Phase II are discharged. Suitable location/s for sampling of sediments were identified which are as given below:

Sampling Point S1: Before the discharge of effluent (D1) from Dombivli MIDC Phase I.

Sampling Point S2: After the discharge of effluent from (D1) Dombivli MIDC Phase 1.

Sampling Point S3: After the discharge of effluent from (D2) Dombivli MIDC Phase II.

Sampling Point S4: After the discharge of effluent from (D2) Dombivli MIDC Phase II.

Sampling locations are as shown below in (Figure 1).



Figure 1 : Effluent discharge and sampling locations in Ulhas River along Dombivli City.

Climatic conditions

Dombivli city has a tropical climate. The mean annual temperature is about 24.3° C to 32.9° C. Months of April and May are the hottest and most dry part of the year. In this period, the temperature rises to 38.0° C. The Humidity present in atmosphere is between 58 to 84%. The average southwest monsoon rainfall ranges from about 1850 mm to 2000 mm. Average annual rainfall in Dombivli region is from 1286 to 1233 mm⁴⁷.

Sample planning, collection and preservation

The study on pollution status along the Ulhas River was carried out for two years i.e. 2012 and 2013. Water and Sediment sampling was performed every week along identified locations of the Ulhas River along Dombivli city. This was done for all the three seasons – summer, rainy and winter for a period of twenty four months.

For water samples, the samples were collected in polythene bottles. These bottles were initially thoroughly cleaned with hydrochloric acid. Then potable water followed by distilled water was used for by washing. Additionally, bottles were again rinsed with the water sample to be collected. Water sampled from different stations was filtered through Whatman No. 41 (pore size 0.45 μ m) filter paper to remove suspended particles. Filtrate was preserved in polythene bottles. To prevent metal precipitation, 2 mL nitric acid was added to filtrate. Water sample was concentrated to ten folds on a water bath, followed by digestion with nitric acid digestion. When the water sample was evaporated to near dryness, it was dissolved in 2% nitric acid followed by filtration and further dilution to 50 mL with distilled water ^{48,49}.

Sediment collected for each season was mixed separately to get gross sample of each season. Sediment was collected by hand-pushing plastic core tubes (7 cm in diameter) into the sediment. Sediments where air dried for eight days and grounded using agate mortar. In order to remove stones, plant roots and to get uniform particle size the sediment was sieved through 0.5 mm sieve, packed in polythene bag and kept in a dry place. 2 g of each sample was digested on a sand bath for 2 hours in 250 ml beaker with 8 ml of aqua regia followed by evaporation to near dryness. The samples where then dissolved with 10 ml of 2% nitric acid and filtered through Whatman No. 1 filter paper followed by dilution with deionised water to give final volumes depending on the suspected level of the metals ⁴⁸. The sediment samples were subjected to nitric acid digestion using the microwave-assisted technique at 30 bar pressure and 700 watts power ^{50,51}.

The samples subjected to prior treatment were analyzed for majority of toxic metals such as – chromium (Cr), zinc (Zn), cadmium (Cd), cobalt (Co), copper (Cu), nickel (Ni), iron (Fe), lead (Pb) and mercury (Hg) by Flame Atomic Absorption Spectrophotometer. For estimation

of d mercury (Hg), hydride generation coupled with an atomic fluorescence detector and cold-vapor techniques were used respectively ⁵³. The techniques and methods followed for analysis and interpretation were according to the standard procedures ^{48,49,52-58}.

General Procedure

Quality Assurance

All instruments were used for analysis were calibrated. Chemicals and reagents were used for analysis were of Analytical Grade. Reagent blanks were used during analyses to correct the interference of reagent impurities and other environmental contaminations. To ensure the removal of all cleaning reagents traces from the laboratory apparatus, they were soaked in nitric acid before analysis followed by thorough rinsing with tap water followed by de-ionised distilled water. Distilled de-ionised water was used to wash the glassware used for analysis.

Detection Method

Heavy metal analysis by AAS technique

The analysis for the majority of the toxic heavy metals like Copper (Cu^{2+}), Mercury (Hg^{2+}), Cadmium (Cd^{2+}), Cobalt (Co^{2+}), Lead (Pb^{2+}), Zinc (Zn^{2+}), Nickel (Ni^{2+}), Chromium (Cr^{6+}) and Iron (Fe^{2+}) in water samples was done by Perkin Elmer Analyst 200 Flame Atomic Absorption Spectrophotometer (2003 model). Mercury estimation was done by hydride generation coupled with an atomic fluorescence detector and cold-vapor techniques ⁶⁰. The standard solutions were prepared by using Analytical Reagent grade chemicals in acidified metal free deionised water. Calibration curves were prepared separately for all the metals, after running different concentrations of standard solutions. To correct for reagent impurities and other sources of errors from environment reagent blank was used during analysis and subtracted from samples. For each determination, average value of three replicates was taken.

III. RESULTS AND DISCUSSION

A number of metals are normally present in relatively low concentrations, usually less than a few mg/L, in aquatic environment and are called trace metals. They are called heavy metals because in their metallic form, their densities are greater than 4 g/cc. Intake of trace heavy metals by plants have been shown to create definite health hazards.

Concentrations of heavy metals in water and sediments

Heavy trace and toxic metals are not necessarily fixed by the sediments permanently, but may be recycled via biological and chemical agents both within the sedimentary compartment as well as in the water column. Behaviour of these metals in the coastal marine sediments is largely related to their capacity for complexation with organic matter in truly dissolved, colloidal, macro particulate phases. If the trace metals persist for a longer time in the environment, it may lead to the possibility of their environmental transformation and may be converted into compounds which are more toxic in nature. Concentration of heavy metals by physical, chemical and biological processes is represented by a number generally known as 'concentration factor'. This concentration of heavy metals in water and sediment is represented by specific term known as distribution coefficient (Kd).

Distribution coefficient (Kd)

Major repository and a potential source of trace metals are the Coastal marine sediments. Sediments act as sinks for many inorganic as well as organic pollutants which are transported through the water column from various sources. As a result of their particle reactivity, trace metals accumulate in the sediments. Thus they persist in the environment long enough after the removal of primary source. The ratio of concentration of an element in sediment (dry weight in g/Kg) to the concentration of element in the water (g/L) under equilibrium conditions is the Distribution coefficient (Kd). The *Kd* values (L/Kg) of different industrially important trace and toxic heavy metals in the Ulhas River along Dombivli City were determined. The seasonal variation in *Kd* values (L/Kg) of metals for water and sediments are presented in and Tables 1 and Figures 3-7. A wide range in *Kd* values were observed for different metals. The *Kd* values (L/Kg) for different metals varied from between 0 to 2207.3 in summer, rainy and winter seasons of 2012. The *Kd* values (L/Kg) for different metals varied from between 0 to 2207.3 in summer, rainy and winter seasons of 2012. The distribution ratio (Kd) of heavy metals Pb^{2+} , Cu^{2+} , Fe^{2+} , Co^{2+} and Cr^{6+} increased in year 2013 as compared to the distribution ratio in year 2012. The distribution ratios were found to be maximum in Ni²⁺ in Year 2012 and 2013 and Minimum in Hg^{2+} in Year 2012 and 2013. The variation in *Kd* values of different metals may be due to the solubility of different metals in water (less soluble the metal, higher the *Kd* value) & contribution of industrial source at a specific site.

Samples of water and sediment which were collected at the sampling points from the Ulhas River were analyzed for heavy metal content in each season i.e. Winter, Rainy and Summer Season of Year 2012 and 2013. The ratio of season wise average values of concentration of heavy metal in sediment to the average values of concentration of heavy metal in water i.e. Distribution Ratio (Kd) was determined and is presented in Table 1.

	Distribution Ratio (Kd)								
	Year 2012			Year 2013					
Heavy Metals	Winter	Rainy	Summer	Winter	Rainy	Summer			
Hg^{2+}	6.0	0	3.6	7.7	0	2.9			
\mathbf{Cd}^{2+}	8.8	5.5	1.3	39.7	5.6	1.0			
Pb ²⁺	72.1	53.1	133.2	92.1	51.3	186.7			
Cu ²⁺	485.9	497	250.5	566.1	400.8	277.7			
Co ²⁺	486.4	116.7	107.5	567.9	170.6	153.3			

Table 1: Distribution coefficient (Kd) of heavy metals in each season of year 2012 and 2013.

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www.jetir.org (ISSN-2349-5162)

Zn^{2+}	540.4	709.6	422.4	596.2	1141.7	411.3
Cr ⁶⁺	1717.2	144.3	741.3	976.2	123	577.3
Ni ²⁺	1241.5	217.5	534.8	2777.1	157	313.3
Fe ²⁺	2085.4	2137.8	2207.3	2075.9	1673.5	2249.7

Mercury (Hg²⁺):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Mercury (Hg^{2+}) were 6, 0, 3.6 and 7.7, ND, 2.9 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 0 in rainy season and maximum Distribution coefficient (Kd) was 6.0 in winter. In 2013, the minimum Distribution coefficient (Kd) was 0 in rainy season and maximum Distribution coefficient (Kd) was 7.7 in winter.

Cadmium (Cd²⁺):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Cadmium (Cd^{2+}) were 8.8, 5.5, 1.3 and 39.7, 5.6, 1.0 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 0 in rainy season and maximum Distribution coefficient (Kd) was 6.0 in winter. In 2013, the minimum Distribution coefficient (Kd) was 0 in rainy season and maximum Distribution coefficient (Kd) was 7.7 in winter.

Lead (Pb^{2+}):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Lead (Pb^{2+}) were 72.1, 53.1, 133.2 and 92.1, 51.3, 186.7 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 53.1 in rainy season and maximum Distribution coefficient (Kd) was 133.2 in Summer. In 2013, the minimum Distribution coefficient (Kd) was 51.3 in rainy season and maximum Distribution coefficient (Kd) was 186.7 in Summer.

Copper (Cu²⁺):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Copper (Cu^{2+}) were 485.9, 497.0, 250.5 and 566.1, 400.8, 277.7 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 250.5 in summer season and maximum Distribution coefficient (Kd) was 497.0 in Rainy Season. In 2013, the minimum Distribution coefficient (Kd) was 566.1 in Winter.

Cobalt (Co²⁺):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Cobalt (Co^{2+}) were 486.4, 116.7, 107.5 and 567.9, 170.6, 153.3 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 107.5 in summer season and maximum Distribution coefficient (Kd) was 486.4 in Winter Season. In 2013, the minimum Distribution coefficient (Kd) was 567.9 in Winter.

Zinc (Zn^{2+}) :

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Zinc (Zn^{2+}) were 540.4, 709.6, 422.4 and 596.2, 1141.7, 411.3 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 422.4 in summer season and maximum Distribution coefficient (Kd) was 709.6 in Rainy Season. In 2013, the minimum Distribution coefficient (Kd) was 411.3 in Summer season and maximum Distribution coefficient (Kd) was 1141.7 in Rainy.

Chromium (Cr⁶⁺):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Chromium (Cr^{6+}) were 1717.2, 144.3, 741.3 and 976.2, 123.0, 577.3 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 144.3 in Rainy season and maximum Distribution coefficient (Kd) was 1717.2 in Winter Season. In 2013, the minimum Distribution coefficient (Kd) was 123.0 in Rainy season and maximum Distribution coefficient (Kd) was 976.2 in Winter.

Nickel (Ni²⁺):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of Nickel (Ni^{2+}) were 1241.5, 217.5, 534.8 and 2777.1, 157.0, 313.3 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 217.5 in Rainy season and maximum Distribution coefficient (Kd) was 1241.5 in Winter Season. In 2013, the minimum Distribution coefficient (Kd) was 157.0 in Summer season and maximum Distribution coefficient (Kd) was 2777.1 in Winter.

Iron (Fe^{2+}):

From the above data in Table 1, it is observed that Distribution coefficient (Kd) values of

Iron (Fe^{2+}) were 2085.4, 2137.8, 2207.3 and 2075.9, 1673.5, 2249.7 respectively for the Winter, Rainy and Summer season of 2012 and 2013 respectively. In 2012, the minimum Distribution coefficient (Kd) was 2085.4 in Winter season and maximum Distribution coefficient (Kd) was 2207.3 in Summer Season. In 2013, the minimum Distribution coefficient (Kd) was 1673.5 in Rainy season and maximum Distribution coefficient (Kd) was 2249.7 in Summer.

The ratio of yearly average values of concentration of heavy metal in sediment to the yearly average values of concentration of heavy metal in water i.e. Distribution Ratio (Kd) was determined and is presented in Figure 2-5.

Distribution coefficient (Kd) of yearly average values of Mercury (Hg²⁺) and Lead (Pb²⁺) in year 2012 and 2013 are shown in Figure 2.



Figure 2: Distribution Coefficient (Kd) yearly average values of Mercury (Hg²⁺) and Lead (Pb²⁺) in year 2012 and 2013.

From Figure 2, it is average yearly observed that the yearly average Distribution Coefficient (Kd) of Mercury (Hg^{2+}) has decreased from 6.5 in year 2012 to 5.5 in Year 2013. The yearly average Distribution Coefficient (Kd) of Lead (Pb^{2+}) has increased from 5.1 in year 2012 to 15.3 in Year 2013

Distribution coefficient (Kd) of yearly average values of Copper (Cu^{2+}), Iron (Fe²⁺) and Cobalt (Co^{2+}) in year 2012 and 2013 are shown in Figure 3.



Figure 3: Distribution Coefficient (Kd) yearly average values of Copper (Cu^{2+}) , Iron (Fe^{2+}) and Cobalt (Co^{2+}) in year 2012 and 2013. From Figure 3, it is average yearly observed that the yearly average Distribution Coefficient (Kd) of Copper (Cu^{2+}) has increased slightly from 75.6 in year 2012 to 83.5 in Year 2013.

The yearly average Distribution Coefficient (Kd) of Iron (Fe²⁺) has increased from 243.3 in year 2012 to 351.8 in Year 2013. The yearly average Distribution Coefficient (Kd) of Cobalt (Co^{2+}) has increased from 399.2 in year 2012 to 448.6 in Year 2013.

Distribution coefficient (Kd) of yearly average values of Cadmium (Cd^{2+}), Zinc (Zn^{2+}) and Chromium (Cr^{6+}) in year 2012 and 2013 are shown in Table 2 and the variation is represented in Figure 4.



Figure 4: Distribution Coefficient (Kd) yearly average values of Cadmium (Cd²⁺), Zinc (Zn²⁺) and Chromium (Cr⁶⁺) in year 2012 and 2013.

From Figure 4, it is average yearly observed that the yearly average Distribution Coefficient (Kd) of Cadmium (Cd^{2+}) has decreased from 432.9 in year 2012 to 377.1 in Year 2013.

The yearly average Distribution Coefficient (Kd) of Zinc (Zn^{2+}) has increased from 523.5 in year 2012 to 499.3 in Year 2013.

The yearly average Distribution Coefficient (Kd) of Chromium (Cr⁶⁺) has increased from 554.0 in year 2012 to 692.6 in Year 2013.

Distribution coefficient (Kd) of yearly average values of Nickel (Ni²⁺) in year 2012 and 2013 are shown in Table 2 and the variation is represented in Figure 5.



Figure 5: Distribution Coefficient (Kd) yearly average values of Nickel (Ni²⁺) in year 2012 and 2013.

From Figure 5, it is average yearly observed that the yearly average Distribution Coefficient (Kd) of Nickel (Ni²⁺) has decreased from 2136.7 in year 2012 to 1972.8 in Year 2013.

IV. CONCLUSION

The regulatory authority responsible for enforcing and controlling the Environmental Norms in India is CPCB i.e. Central Pollution Control Board. CPCB regulates the enforcement, monitoring and effective implementation of the pollution control norms under Water Prevention and Control of Pollution Act 1974. Hence for ensuring the effectiveness of implementation of the water quality norms to a desired level, monitoring at regular intervals is very important. Due to this periodic monitoring, trends of water quality could be known and measures to strategically prioritize the control efforts and actions can be planned. For taking these measures, accurate data indicating the exact problems, location, seriousness and contributing factors, is required. This information will help to determine and plan cost efficient and long term solutions related to water quality improvement. The above study on pollution due to trace and toxic heavy metals in water and subsequently

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in sediments of Ulhas River along the Dombivli City near Mumbai will be useful in evaluating effectiveness of pollution control measures already is existence; rational and adequate planning & prioritization of pollution control strategies; assessing the fitness of water for different uses; assessing assimilative capacity of a water body thereby reducing cost on pollution control and evaluating water quality trend over a period of time. The present study on distribution coefficient of metals will help to provide a means for evaluating the long term accumulation of heavy metal contaminants.

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