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Assessment of Heavy Metals in Surface Water of Adirampattinam Seashore, Thanjavur District, Tamil Nadu, India

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Abstract

Heavy metal analysis is the most promising tool to eliminate the unwanted pollution from the atmosphere. Particularly in seawater there are number of heavy metals are present. Assessment of this analysis will be useful to detect the unwanted heavy metals and it is the reason to find the new removal techniques. In surface water of Adirampattinam seashore area the heavy metals, ^{26}Fe, ^{82}Pb, ^{30}Zn, ^{25}Mn, ^{29}Cu, ^{48}Cd, ^{33}As and ^{23}V in water have been detected by Atomic Absorption Spectroscopy (AAS) technique. It is recorded that the analysis of water has been achieved for one year of December 2016 to December 2016 when the water flow is lower and higher respectively. AAS measurements give data collected from 2 stations of Adirmpattinam, Thanjavur District, India, area in equal intervals of three months for four times. Water samples were collected year from December 2016 to December 2016. For each metallic element, AAS analysis technique used a particular wavelength and type of flame. Besides, high and low values of each heavy cation in water are recorded, indicating that the highest value is observed for As (3.250 mg/L) (Station A2) and lowest value is for Cd (0.015 mg/L) in December 2016 (Station A1).

Keywords: Heavy metals; Atomic absorption spectroscopy; Permissible limit

Introduction

Metallic elements of periodic "trace elements" are called as Heavy metals. Heavy metals have important part in the recent world decades within the framework of environmental pollution. Pollution is an undesirable change in the physical, chemical or biological characteristics of air, water and soil that may harmfully affect life or create a potential health hazard to any living organism.

Xueying Li, Haihui Zhou et al. reported that chitosan/Sulfydryl-functionalized graphene oxide composite (CS/GO-SH) was synthesized via covalent modification and electrostatic self-assembly. He analysed that the CS/GO-SH was with multifunctional groups such as AOH, ACOOH, ASH and ANH_2. The resulting CS/GO-SH was used as an adsorbent material for removal of Cu (II), Pb (II) and Cd (II) in single- and multi-metal ions systems [1].

Limei Cui et al. reported that resin loaded magnetic b-cyclodextrin bead and graphene oxide sheet (MCD-GO-R) was synthesized successfully and found to be an excellent adsorbent for Hg(II) removal. The maximum adsorption capacity was 88.43 mg g^{-1} at 323 K and pH 7.1. The as-prepared adsorbent could perform well in a wide pH range from 4.0 to 10.0. It was found that the contaminant adsorption was accomplished mainly via chelation or ion exchange and come to equilibrium in only 30 min [2].

Xia Liu In this study, phosphate-functionalized graphene oxide (PGO) was prepared by grafting triethyl phosphate onto the surface of GO using Arbuzov reaction. The application of the PGO to remove U(VI) from aqueous solution was investigated with a maximum adsorption capacity of 251.7 mg/g at pH ¼ 4.0 ± 0.1 and T ¼ 303 K. Experimental results gave a better removal efficiency toward U(VI) on PGO surface than other heavy metal ions at acidic solution, indicating the selective extraction of U(VI) from environmental pollutants [3].

Mansoor Khan et al. annalysed that Magnetic allylamine modified grapheneoxide-poly (vinylacetate-co-divinylbenzene) (MGO-DVB-VA) was synthesized and used for magnetic solid phase extraction of Pb(II), Cd(II), Cu(II), Ni(II) and Co(II) prior to their determination by flame atomic absorption spectroscopy. The preconcentration factor (PF) is 40 for all the metals and the limits of detection for Pb, Cd, Cu, Ni and Co are in the range of 0.37-2.39 mgL^{-1} and relative standard deviation below 3.1%. In optimum conditions, the recovery percent of analytes by using magnetic allylamine modified graphene oxide-poly(vinyl acetate-co-divinylbenzene) was compared with magnetic grapheneoxide. When we used the magnetic graphene oxide as adsorbent, the recovery percents of Ni(II), Cd(II) and Co(II) ions were found as 86%, 90%, and 85%, respectively [4].

Xiang bing Zhu et al. reported that Multi-walled carbon nanotubes (MCNTs) were dispersed in graphene oxide (GO) colloids to be further functionalized with diethylenetriamine (DETA), resulting in GO–MCNTs–DETA nanocomposites for the solid-phase extraction and analysis of Cr(III), Fe(III), Pb(II), and Mn(II) ions at the trace levels in waste water. Inductively coupled plasma optical emission spectrometry (ICP-OES) indicates that this new solid phase sorbent could facilitate the maximum static adsorption capacities of 5.4, 13.8, 6.6 and 9.5 mg g^{-1} for Cr(III), Fe(III), Pb(II), and Mn(II) ions, respectively, showing the adsorption capacity upto 95% within about 30 min [5].

Xiangxue Wang et al. in their studies reported that, Graphene oxides (GOs) have attracted intense multidisciplinary study because of

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their special physicochemical properties and their possible applications. The GOs are applied as adsorbents to remove Pb(II) ions from aqueous solutions and the sorption of Pb(II) is strongly dependent on pH and the sorption of Pb(II) is mainly attributed to the formation of strong surface complexes [6].

Chunyan Wang et al. his study proposes a new approach to prepare polyimide (PI)-based nanocomposites containing amino groups functionalized graphene oxide (GO-NH_2) via in situ polymerization and thermal imidization.GO-NH_2 nanosheets which exhibit excellent dispersibility and compatibility with the PI matrix were successfully obtained through the chemical reaction between graphene oxide (GO) and p-phenylenediamine (PDA) [7].

Ping Tan et al. reported, highly ordered layered graphene oxide (GO) membranes with larger interlayer spacing were prepared by induced directional flow and were used as adsorbents for the removal of Cu^{2+}, Cd^{2+} and Ni^{2+} from aqueous solutions.. The results indicated that the adsorption of Cu^{2+}, Cd^{2+} and Ni^{2+} onto GO membranes was greatly influenced by the pH and weakly affected by the ionic strength. The maximum adsorption capacities of the GO membranes for Cu^{2+}, Cd^{2+} and Ni^{2+} were approximately 72.6, 83.8 and 62.3 mg/g, respectively [8].

Ayman A Gouda et al. in his studies explained that a new, sensitive and simple solid phase extraction (SPE), separation and pre concentration method of some heavy metal ions, Cd(II), Cu(II), Ni(II), Pb(II) and Zn(II) at trace levels using multiwalled carbon nanotubes (MWCNTs) impregnated with 2-(2-benzothiazolylazo)orcinol (BTAO) from food and water samples were investigated. The metals retained on the nanotubes at pH 7.0 were eluted by 5.0 mL HNO_3 (2.0 mol L^{-1}). The detection limits (LODs) for Cd(II), Cu(II), Ni(II), Pb(II) and Zn(II) were found at 0.70, 1.2, 0.80, 2.6 and 2.2 lg L^{-1}, respectively [9].

Dungang Gu et al. reported that in his study, the adsorption behaviors of a range of metals onto multi-layered graphene oxide (GO) as a function of pH and metal:GO ratio were measured. The GO exhibited an affinity for the metals in the following order: Pb(II) > Cu(II) > > Cd(II)≈Ni(II)≈Zn(II) > Sr(II)≈Ca(II). A four discrete-site non-electrostatic surface complexation model was used to account for the observed adsorption behaviors. The predicted extent of Nd adsorption onto GO is within 5% of the measured extent of Nd adsorption in an EDTA-free solution, and the predicted Nd adsorption behavior with increasing EDTA concentration is also in reasonable agreement with the measurements [10].

Metals such as Arsenic, Lead, Cadmium, Mercury and Selenium are highly toxic even in minor amounts. In this paper, the heavy cations like iron (^{26}Fe), lead (^{82}Pb), zinc (^{30}Zn), manganese (^{25}Mn), copper (^{29}Cu), cadmium (^{48}Cd), arsenic (^{33}As) and vanadium (^{23}V) are detected and their amounts (mg/L) in surface area seashore area water are measured in the periods of December 2016 to December 2016. These periods are selected because the surface water, Adirampattinam, Tamil Nadu Thanjavur District, flow is low in April and high in December. Up to our knowledge, no work on heavy metals detection in surface water of Adirampattinam has been previously reported [11].

The present study was conducted to assess the risk to human health by heavy metals (Cd, Cu, Pb, Zn, Ni and Cr) through the intake of locally grown vegetables, cereal crops and milk from this polluted water irrigated site. This surface water of Adirampattinam seashore is connected with backwater of Sea. So the waste waters were polluted and it will flow through the surface of Adirampattinam landside. Surface water is not directly contaminated due to wastewater irrigation, but is an important route of transfer of heavy metals from grass to animals. Heavy metal concentrations were several fold higher in all the collected samples from wastewater irrigated site compared to clean water irrigated ones. Cd, Pb and Ni concentrations were above the 'safe' limits of Indian and WHO/FAO standards in all the vegetables and cereals, but within the permissible limits in surface water. The higher values of metal pollution index and health risk index indicated heavy metal contamination in the wastewater irrigated site that presented a significant threat of negative impact on human health. Rice and wheat grains contained less heavy metal as compared to the vegetables, but health risk was greater due to higher contribution of cereals in the diet. The study suggests that surface water of seashore converted as wastewater irrigation led to accumulation of heavy metals in seashore area stuff causing potential health risks to consumers.

Experimental Section

Atomic Absorption Spectroscopy is used to assess the heavy metals of surface water area. The elements arsenic, cadmium, copper, iron, lead, manganese, molybdenum, vanadium and zinc have been analysed. Flame spectroscopy is an analytical technique used for the qualitative and quantitative determination of the element in a sample. In this method, samples are introduced in the form of a homogenous liquid, into a flame where thermal and chemical reactions create 'free' atoms capable of absorbing, emitting or fluorescing at a characteristic wavelength.

In atomic absorption spectroscopy majority of free atoms in the commonly used flames are in the ground state. A light source emitting a narrow spectral line of the characteristic frequency is used to excite the free atoms in to the flame. The decrease in energy of the light is then measured. The absorbance is proportional to the concentration of free atoms in the flame, given by Lambert-Beer's Law.

$$Ab = Log\frac{I_0}{I_t} = kCl \quad (1)$$

Where, *Ab* is the absorbance, I_o is the intensity of incident radiation emitted by the light source, I_t is the intensity of transmitted radiation (amount not absorbed), C is the concentration of sample (free atoms), k is a constant (molar extinction coefficient) can be determined experimentally and l is the path length. This is the most common method where interference effects are known to be absent. Usually at least three standards and a blank are used to cover the range of 0.1 to 0.8 absorbance. The blank solution is used to calibrate the instrument. The standards are then analysed with the lowest concentration first, and the blank run between standards, to ensure that the baseline (zero point) has not changed. Samples are then analysed and their absorbance recorded. A graph of absorbance versus concentration is plotted. The calibration can be performed in the concentration mode in which case the concentration of the sample is read off directly. In atomic absorption spectroscopy the wavelength (nm) and flame type for different heavy metals are listed in Table 1 [10,11].

Results and Discussion

Sample collection

The heavy metals analysis was performed for surface water samples (mainly for two stations A1 and A2) collected. Water samples were examined to detect and to estimate the concentration of heavy metals. Water samples were collected four times in a year from January 2016 to December 2016. For each metallic element, AAS analysis technique used a special wavelength (nm) within the 190-320 nm range and type of flame as indicated in Table 1. Heavy metallic cation amounts, in mg/L, are detected in surface water stretch as listed below (Tables 2a-2h)

	January 2016	April 2016	August 2016	December 2016	January 2016	April 2016	August 2016	December 2016
Month and Year	A1	A1	A1	A1	A2	A2	A2	A2
Iron (Fe)	0.022	0.224	0.162	0.270	0.044	0.052	0.190	0.032
Lead (Pb)	0.037	0.183	0.064	0.172	0.032	0.023	0.192	0.047
Zinc (Zn)	0.031	0.258	0.021	0.210	0.021	0.028	0.210	0.031
Manganese(Mn)	0.045	0.211	0.043	0.250	0.031	0.017	0.220	0.045
Copper (Cu)	0.563	0.512	0.506	0.499	0.496	0.486	0.399	0.463
Cadmium (Cd)	0.026	0.022	0.023	0.012	0.021	0.031	0.022	0.030
Arsenic (As)	0.009	0.008	0.008	0.0065	0.0054	0.0081	0.0055	0.081
Vanadium (V)	0.056	0.065	0.071	0.045	0.056	0.045	0.055	0.056
Range	0.045-0.022	0.258-0.211	0.187-0.021	0.250-0.172	0.044-0.021	0.052-0.017	0.258-0.211	0.187-0.021

Table 1: The element, wavelength and type of flame.

Element	Wavelength (nm)	Type of Flame
Iron (Fe)	248.3	Air-Acetylene
Lead (Pb)	283.3	Air-Acetylene
Zinc (Zn)	213.9	Air-Acetylene
Manganese(Mn)	279.5	Air-Acetylene
Copper (Cu)	324.7	Air-Acetylene
Cadmium (Cd)	228.8	Air-Acetylene
Arsenic (As)	193.7	Argon-Hydrogen
Vanadium (V)	318.4	Nitrous Oxide-Acetylene

Table 2a: Arsenic is converted to its hydride and aspirated into argon-hydrogen flame.

Month and Year	A1	A2
January 2016	0.255	0.215
May 2016	0.319	0.233
August 2016	0.315	0.199
December 2016	0.310	0.258
Range	0.319-0310	0.258-0.199

Table 2b: Lead in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

Month and Year	A1	A2
January 2016	0.128	0.096
April 2016	0.134	0.112
August 2016	0.139	0.124
December 2016	0.130	0.121
Range	0.139-0.128	0.124-0.096

Table 2c: Zinc in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

Month and Year	A1	A2
January 2016	0.119	0.120
April 2016	0.125	0.129
August 2016	0.099	0.101
December 2016	0.121	0.127
Range	0.099-0.125	0.101-0.129

Table 2d: Manganese in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

Month and Year	A1	A2
January 2016	0.129	0.054
April 2016	0.121	0.059
August 2016	0.124	0.072
December 2016	0.126	0.081
Range	0.129-0.121	0.081-0.054

Table 2e: Copper in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

Month and Year	A1	A2
January 2016	0.019	0.027
April 2016	0.022	0.031
August 2016	0.032	0.037
December 2016	0.025	0.025
Range	0.022-0.032	0.037-0.025

Table 2f: Cadmium in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

Month and Year	A1	A2
January 2016	2.012	1.041
April 2016	2.780	1.820
August 2016	2.580	0.994
December 2016	2.824	1.210
Range	2.824-2.012	1.820-0.994

Table 2g: Arsenic in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

Month and Year	A1	A2
January 2016	1.210	0.942
April 2016	0.924	1.410
August 2016	1.281	1.962
December 2016	1.108	0.941
Range	1.210-0.924	1.410-0.941

Table 2h: Vanadium in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

and explained by the bar chart of Figures 1a-1g. The Figure 2 displays the high (left) and low (right) value of metallic concentration in water for each heavy metal [12].

Iron (Fe): The high value of iron 0.258 mg/L is observed at A1 and A2 in May 2016 and the low value is 0.017 mg/L at A1 and A2 in August 2016. The maximum permissible limit is 0.3 mg/L. It is observed that the values are slightly higher than in summer. The high value in summer may be due to the industrial waste water exposed to the water system and high concentration of industrial effluent and domestic sewages. The concentration of metals in the surface of a river may increase according to the effluent discharged from nearby industries [13].

Lead (Pb): The high value of lead 0.319 mg/L is observed at A1 and A2 stations Surface water in equal four months in one year at two stations stretch. The maximum permissible limit of lead is 0.1 mg/L. In our present study A1 and A2 show higher value than the permissible limit. The lead pollution on the surface water may be the result of discharge of effluents, old plumbing, house hold sewages, agricultural runoff containing phosphatic fertilizers and human and animal excreta. Presence of lead in phosphatic fertilizers has been revealed [6]. Low

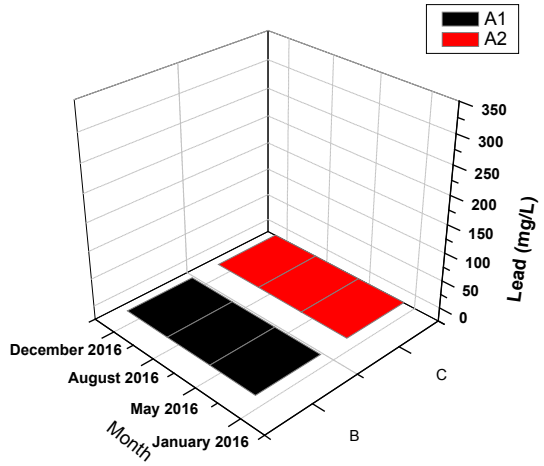

Figure 1a: Lead in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

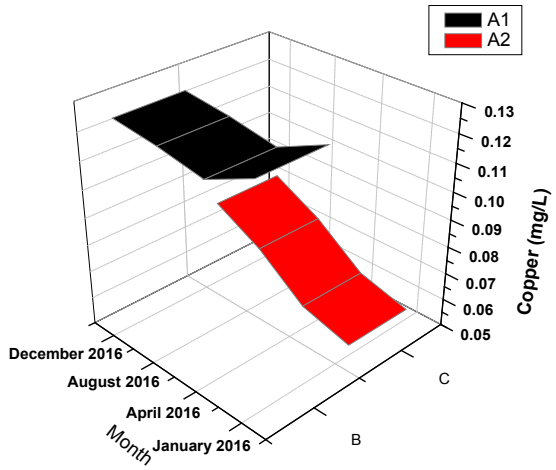

Figure 1d: Copper in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

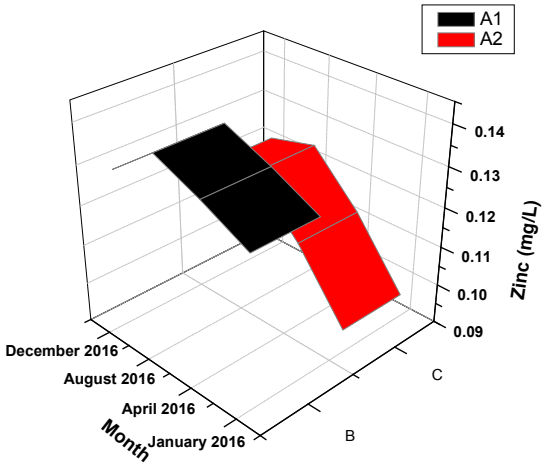

Figure 1b: Zinc in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

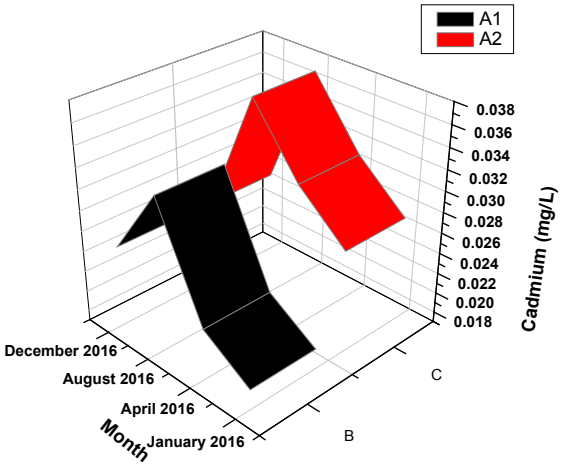

Figure 1e: Cadmium in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

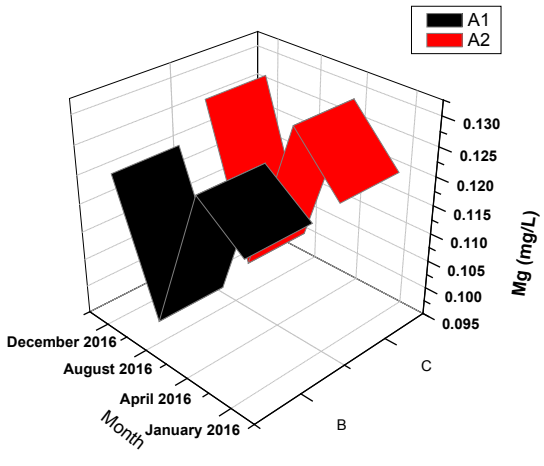

Figure 1c: Manganese in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

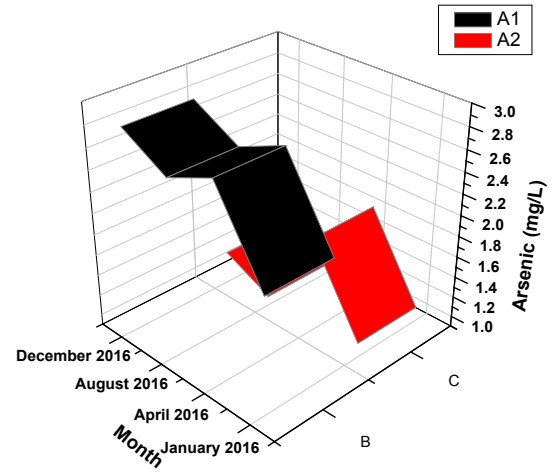

Figure 1f: Arsenic in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

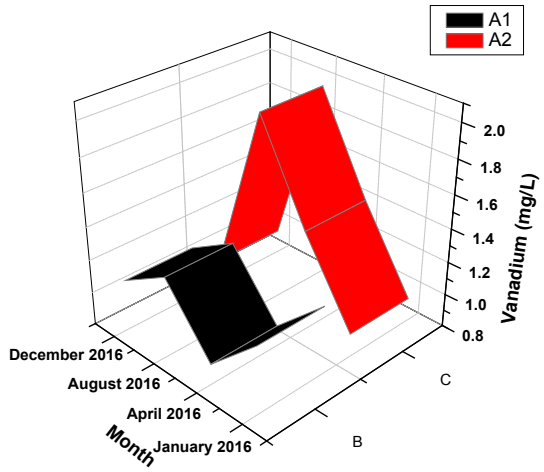

Figure 1g: Vanadium in mg/L of surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

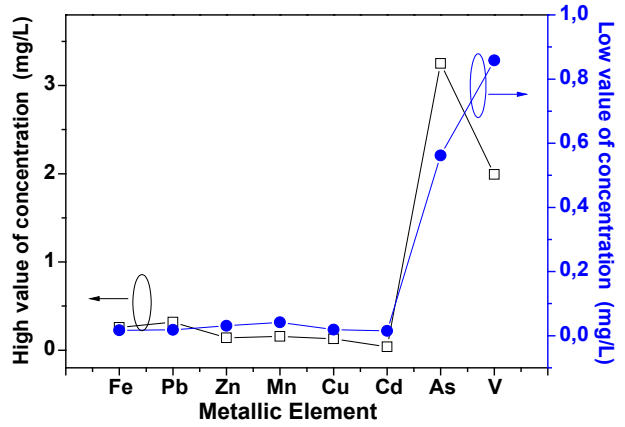

Figure 2: High (left) and low (right) value of metallic concentration in water for each heavy metal surface water of Adirampattinam seashore area stretch at two stations A1 and A2.

values at particular stations may be due to the complex formation with organic materials during the passage through the soil [14].

Zinc (Zn): The high value of Zinc 0.139 mg/L is noticed at A1 during May 2016 and a low value of 0.031 is observed at A2 in the month of August 2016. The minimum permissible limit is 5 mg/L. In our present study, the value of Zinc is below than the permissible limit in all the stations. The value of zinc in summer is higher than in monsoon. The high value of zinc may be due to the agricultural runoff, domestic waste discharge and the industrial effluents [15]. Zinc occurs at concentrations of approximately 250 mg/kg in dry solids of faces [16]. Since the public use the river bed as open toilet, it may be one another reason for higher value. Zinc is toxic and may cause vomiting, renal damage and cramps. Its presence above the permissible limit is unacceptable in water used for drinking purpose [17].

Manganese (Mn): A high value of Manganese 0.156 mg/L is found at A1 in the month of May 2016 and a low value of 0.042 mg/L at A2 in the month of August 2016. The maximum permissible limit for Manganese is 0.1 mg/L. In our present study the values of manganese show less than the permissible limit at A6 station. High values of manganese are observed in monsoon compared to summer. This may be due to the flushing action of the rain [18]. Manganese is present in water as a soluble divalent ion. It is non-toxic to animals but it is objectionable and tenacious stains to laundry and plumbing fixtures. It is also not suitable for domestic purpose [19]. The high value of manganese may be due to the influence of industrial effluents and domestic sewage entering into the river system.

Copper (Cu): A high value of Copper 0.129 mg/L is observed at A1 in the month of May 2016 and a low value of 0.019 mg/L is noticed at A2 in the month of August 2016. The maximum permissible limit of copper is 1.0 mg/L. In our present study the value of copper is lower than the permissible limit in all the stations. Copper is widely distributed and is an essential element for all living system. It is also used for bioactivity of living system. But higher value of copper is considered as pollutant. The high value of copper is noticed in summer. This may be due to the sewage of domestic and agricultural inputs [20]. The low value of copper may be the result of adsorption process by the soil reduces the concentration of the heavy metals in water.

Cadmium (Cd): A high value of Cadmium 0.037 mg/L is found at A1 in May 2016 and a low value of 0.015 mg/S is found at A2 in the month of August 2016. The maximum permissible limit of Cadmium is 0.01 mg/L. The value is found to be higher than the permissible limit in all stations due to the pollution. Cadmium is considered to be hazardous metal because of its toxicity and accumulation capacity in the living system [21].

Arsenic (As): A high value of Arsenic 3.250 mg/L is found at A1 in the month of May 2016 and a low value of 0.562 mg/L is found at A2 in the month of August 2016. The maximum permissible limit of arsenic is 0.2 mg/L. The value of arsenic is found higher than the permissible limit at all stations. The high values may be due to the application of *arsenic* in the form of herbicides, insecticides and fungicides in the agricultural lands which may be thrown to the river by runoff. Like cadmium, arsenic is also biologically essential and toxic. It leads to the protein complexation with enzymes and uncoupling of phosphorylation [22]. Many arsenic compounds are water soluble and hence arsenic concentration of water can occur readily.

Vanadium (V): A high value of Vanadium 1.992 mg/L is observed at A1 in the month of May 2016 and a low value of 0.858 mg/L is observed at A2 in the month of August 2016 as shown in Figure 2. The permissible limit of vanadium is 7 mcg/L. The high value is observed in summer than monsoon. It may be due to the low flow rate of river water coupled with high temperature increases the concentration of heavy metals which are fed into the river through untreated sewage, automobile wastes, electroplating units etc. [23].

Conclusion

Water is the most important and vital molecule of our planet and covers 75% of earth surface. But it is getting polluted due to high industrial growth. The heavy metals produced by industrial activities are recurrently added to it and considered as dangerous pollutants. Increasing concentration of toxic heavy metals Pb(II), Cd(II), Cu(II), Ni(II) and Co(II), in wastewater is a severe threat for human. Heavy metal contaminated water is highly carcinogenic and poisonous at even relatively low concentrations. Lead concentration at stations A1 and A2 are low than the permissible limit. At all other stations, the values of lead are high than the permissible limit. The low value may be due to the complex formations with organic materials during the passage through the soil. The pollution is caused by discharge of effluent, old

plumbing, sewages, agricultural runoff etc. Zinc concentration is below at all stations than the permissible limit. The high value is observed in summer which may be due to the agricultural run off, domestic wastes and industrial pollution. Manganese concentration is less than the permissible limit at station A2. High values of manganese are observed in monsoon than summer which may be due to the flushing action of the rain. Copper concentration is low than the permissible limit in all the stations. A high value is observed in summer than monsoon. The high value may be due to the seepage of domestic sewage and agricultural inputs and the low value is observed due to the adsorption process by the soil. Cadmium concentration is found to be high than the permissible limit almost in all stations. The high value may be due to the usage of herbicides, insecticides and fungicides etc. Vanadium concentration is observed below than the permissible limit. The value is found to be higher in summer than in monsoon. It may be due to the low flow rate of river water in summer coupled with high temperature. The high value is due to the untreated sewage, waste from automobile workshop, electroplating units etc.

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