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Human health risk assessment from exposure of heavy metals in river water samples of Jonk River, Kasdol area, Chhattisgarh India

Swati Sarwa[1*], Dr. Piyush Thakur[2], Dr Shweta Chodey[3]
[1, 2] School of Biological and Chemical Sciences, Mats University, Raipur, Chhattisgarh, India
[3] Department of Basic Sciences, Govt. Engineering College, Raipur, Chhattisgarh, India

Abstract
Contamination in river water can directly pose significant health risks through oral ingestion, particle inhalation, and dermal contact. Here, we studied the contamination level of heavy metals in water samples to assess the carcinogenic and non-carcinogenic doses through multi-pathway exposures for health hazard point of view. The concentration of metals viz. zinc, iron, lead, cadmium, copper, aluminum, nickel, arsenic, mercury and manganese were measured in the collected water samples of Kasdol area, Raipur C.G., India by using atomic absorption spectrometry (AAS). The average contents of the metals studied were as a heavy metals respectively. The contamination factor, pollution load index, and index of geo-accumulation were also calculated to assess the quality of river water. Carcinogenic risk also has been calculated for heavy metals carcinogenic doses for different exposure routes follow the order doses for ingestion (CDI_{ing-ca}) > > doses for inhalation (CDI_{inh-ca}) > > doses for dermal absorption ($CDI_{dermal-ca}$). Our results showed that the average values of Mn, Cd, Ni, Zn in the river water were within the acceptable limits, whereas the average values of Fe, Zn, Cu, Cd, Al, As, Ni and Hg in the water samples were superior than USEPA (1999) rule. The hazard index for both children and adults is less than unity, indicating no non-carcinogenic risk for both children and adults.

Keywords: water contamination, risk assessment, carcinogenic doses, hazard index, pollution load index

1. Introduction

The contamination of river water by heavy metals is a substantial environmental problem worldwide (Alloway 1995) [1] due to the abundance of sources and their environmental persistence (Lim *et al*. 2008) [15]. The biotoxic effects of many heavy metals in human biochemistry are of great concern. Heavy metals are omnipresent in the environment, as a result of both natural processes and anthropogenic activities (Wilson and Pyatt 2007) [39] and is estimated that the contribution of metals from anthropogenic sources in river water is higher than the contribution from natural ones (Nriagu and Pacyna 1988) [22]. The natural sources are primarily rocks, minerals and atmospheric deposition, while anthropogenic sources mainly include direct or indirect emissions of trace elements from human activities, such as mining, smelting, the burning of fossil fuels, waste incineration and disposal, sewage irrigation, the use of leaded gasoline and motor traffic, production and extensive use of chemical fertilizers and pesticide, daily activities, aerosolized trace elements, etc. Due to the disturbance and acceleration of natures slowly occurring geochemical cycle of metals by man, most river water of rural and urban environments may accumulate one or more of the heavy metals above defined background values high enough to cause risks to human health, plants, animals, ecosystems, or other media. The role of heavy and trace elements in the water system is increasingly becoming an issue of global concern at private as well as government levels, especially as water constitutes a crucial component of rural and urban environments (United States Department of Agriculture 2001). The water environment is interfacial, heterogeneous and changing and the basic characteristics of water contamination are different from those of air and water, such as concealment and hysteresis, accumulation and irreversibility (Zhou and Song 2000) [42]. Ten elements copper, cadmium, nickel, iron, zinc, magnesium, lead, arsenic, aluminum, mercury, chlorine, boron, are essential nutrients required in trace amounts for plant growth as well as human and animal health. In water ecosystems, the toxicity and mobility of these metals depend on various factors like total specific chemical form, metal binding state, concentration of metals and properties. The water is regarded as the ultimate sink for metals discharged in to the environment (Hooda 2010) [8]. Extreme levels of many metals in water can reduce water fertility and may result in poor quality of agricultural products and ultimately can endanger human, animal, and ecosystem health effects such as cancer, reproductive, development and neurological disorders, cardiovascular, kidney and renal problems, lung damage, contact dermatitis and brittle hair and hair loss.

Heavy metals can exert their toxicity via dermal, inhalation and ingestion pathways from water influence human health with severe consequences. The direct ingestion is the most important pathway for human exposure to water contamination among all three pathways. Exposure of children to heavy metal was accepted as highest risk group because children have a higher adsorption rate of heavy metal between 39 to 270 mg kg^{-1} due to their accidental ingestion of water particles may lodge in the lungs and there is a chance that contamination may be absorbed into the bloodstream. Inhalation is not a serious source of exposure as compared to ingestion, but may be relevant to those exposed repeatedly over a long time period. Due to urbanization in Kasdol area water is increasingly laced with natural weathering process, atmospheric deposition and anthropogenic pollutants from municipal sewers, domestic waste water discharge, burning of fossil fuels and toxic effluents discharge from sewage treatment plants and agricultural fields that are treated with fertilizers and pesticides, respectively.

Trace elements in water cannot be degraded by microorganisms but they can accumulate and be absorbed by crops, enter the human body through direct cannot or the food chain and harm human health (Qin *et al*. 2014) [24]. The accumulation of heavy metals in water profile and plants is of increasing concern because of the potential human health risks. Moreover, the selected few locations come under the hilly terrain and the rest of the locations are situated on the outer plains of the studied regions. The lands of hilly areas are agricultural lands, so the water samples of that region may be contaminated with heavy metals due to industrialization and urbanization. Depending upon the sources like use of pesticides, fertilizers, discharge from industries waste, and other human activities, ten heavy metals were selected zinc, lead, copper, cadmium, aluminum, nickel, arsenic, magnesium, iron, mercury to analyze in the water samples of the studied areas.
The objective of the present study area to assess the concentration of heavy metals in river water samples and to assess carcinogenic and non-carcinogenic human health risks of water contaminated by heavy metals through multi pathway exposures. Many geochemical approaches, such as contamination factor (CF), pollution load index (PLI) and index of geo accumulation are also calculated for evolution of anthropogenic influence on water samples. Moreover no study area for the contamination of water in this region has been done earlier. So, this study can also be helpful to establish a research baseline and the results can be used as the baseline for further bio remediation actions in this study area.

2. Material and Methodology

The study area

An observed was conducted to identify the concentration of heavy metals in the water samples of the Kasdol area. The total geographical area of the Kasdol area Baloda Bazaar district longitude 21.6569 N° and 82.1592 E° is the outer plains. Most of the hilly area is at medium high altitude and is not residential. The concentration of heavy metals in the water samples of the Kasdol area Baloda Bazaar district has been already measured by our research groups to measure the level of heavy metals in the river water samples. The concentration of heavy metals was found to be very high in groundwater rather than stored water due to human activities and industrialization and was found that all values were well within the limit suggested by (WHO 2007) [38] and (European Union Commission 1998). So to check out the presence of the contents of heavy metals in water sample if present through the discharge of contaminated water sources in the water samples of the studied area, the present study was conducted. This study has been carried out in Kasdol area, Baloda Bazaar district of Chhattisgarh, India (Fig.1), which is the central state Baloda Bazaar is geographically located at latitude 21.6569 north and 82.1592 east. The area lies in the foothills of sub mountainous region. Physiographical, the district is occupied by two major units extending throughout the district. The unit the extends in the central part is the hilly tract of formations and the unit extending is the central part are a plain tract, the outer plain. In Baloda Bazaar district, three sample sites are chosen i.e., site-1 Rajandevri, site-2 Golajhar, & site-3 Chandan, and surface water samples of Jonk river were collected and tasted during monsoon, winter and summer seasons. These rivers act as major drainage lines in the area and enter outer plain part of the district. Nine water samples were collected at a depth of 15 cm from the top surface from village of the Kasdol area, Baloda Bazaar district, Chhattisgarh, India. The water samples were collected in pathology bottles and named properly. The collected water samples were then air dired, grounded and homogenized with a mortar and pestle and sieved through 300 µm mesh (Kaur *et al*. 2018) [11]. The measurement of trace element concentration in river water is generally a combination of a digestion procedure for dissolution of elements and subsequent measurement of the dissolved elements. For determination of heavy metals, the prepared water samples were digested using AAS method consisting of digesting water samples. The digested samples were cooled and filtered through whatman filter paper no. 1. The filtrates were analyzed by atomic absorption spectrophotometer (AAS) (Rahman *et al*. 2012; Sun *et al*. 2010) [25, 29]. The concentration of iron, zinc, lead, aluminum, mercury, nickel, copper, cadmium, arsenic, and magnesium were measured by air acetylene flame.

Fig 1: Kasdol area in Baloda Bazar District of Chhattisgarh, India.

Analytical procedure
For quality control, blanks and duplicates (samples) were analyzed after every eight sample analyses to assess the instrumental stability. All solutions were prepared using double distilled water. The calibration curves were obtained for the metals using their standard solutions to establish standard curves before metal analysis, and correlation coefficients of the obtained standard curve were greater than 0.99. The precision of analytical procedures was expressed as relative standard deviation (RSD), which ranged from 5 to 10% and was calculated from the standard deviation divided by the mean value (Table 1).

Statically analysis
Analysis of variance was used to determine whether the concentrations of metals varied sighificantly with value less than 0.05 ($p<0.05$) considered statistically significant (Zar 1999) [40]. The minimum, lower quartile, median, upper quartile and the maximum were calculated by Tukey's resistant five letters summary statistics (Tukey 1977) [31] and visualized by box whisker plots. Shapiro wilks tests were used to determine the normality of data and was considered statistically significant if p value was greater the 0.05. All the statistical tests were carried out by using statistical software SPSS 16.0. The cluster analysis and principal component analysis (PCA) were performed to identify the relationships between the heavy metals concentration in the water samples (Facchinelli *et al*. 2001) [6].

Theoretical methods
Three contamination indices, namely the contamination factor (CF), the pollution load index (PLI), and index of geo accumulation (I_{geo}), were calculated to evaluate the degree of contamination in water samples.

Contamination factor
The level of contamination by heavy metal in water samples is expressed in terms of a CF calculated as:

$$CF = C_m\,(\text{Sample}) / C_m\,(\text{Background}) \quad (1)$$

Where C_m (sample) is the concentration of a given metal in water sample and C_m (background) is the value of the metal equals to the world surface rock average given by Martin and Meybeck (1979) [17]. The following terminology is used to describe the contamination factor: $CF<1$ (low contamination); $1\leq CF<3$ (moderate contamination); $3\leq CF<6$ (considerable contamination); $CF>6$ (very high contamination), respectively.

Pollution load index
PLI is used to evaluate the pollution level of heavy metals and has been evaluated following the method proposed by Tomlinson *et al*. (1980) [30]. It provides adequate information about the significance of the measured concentration of metals on the intrinsic water features. This parameter is expressed as:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots\ldots CF_n)^{1/n} \quad (2)$$

Where n is the number of metals. PLI represents comparative means to assess a site quality, where a value of $PLI<1$ denotes perfection or no pollution; $PLI=1$ presents that only baseline levels of pollutants are presented and $PLI>1$ would indicate deterioration of site quality (Tomlinson *et al*. 1980) [30].

Index of Geo accumulation
Index of geo accumulation (I_{geo}) was calculated to enable the qualitative assessment of water contamination by heavy metals and calculated by using formula given by muller 1969 [18]. It is used to quantify the degree of anthropogenic contamination (Sahu and Bhosale 1991) [28].

$$I_{geo} = \text{Log}_2\,[C_m\,(\text{sample})/1.5 \times C_m\,(\text{background})] \quad (3)$$

Where the factor 1.5 is introduced to include possible variation of the background values due to lithogenic effect (Rogan *et al*. 2010) [27]. Muller (1981) [19] proposed seven grades or classes of the geo accumulation index. These grades are as follows: $I_{geo}\leq 0$, uncontaminated (grade 0); $0<I_{geo}\leq 1$, uncontaminated to moderately contaminated (grade 1); $1<I_{geo}\leq 2$, moderately contaminated (grade 2); $2<I_{geo}\leq 3$, moderately to heavily contaminated (grade 3); $3<I_{geo}\leq 4$, heavily contaminated (grade 4); $4<I_{geo}\leq 5$, heavily to extremely contaminated (grade 5); $I_{geo}>5$, extremely contaminated (grade 6).

Human health risk estimation
Heavy metals may enter the human body through 3 pathways which includes direct ingestion of water, inhalation of particulates emitted from water and dermal contact of contamination water through water adhered to exposed skin. So, the carcinogenic and non-carcinogenic hazard risk (HI) for children and adults was calculated using equation (4) (8) adapted from USDOE (2011) [32] and USEPA (2010, 2011) [33]:

$$CDI_{ing\ nc}\,(\text{mg kg}^{-1}\text{day}^{-1}) = C \times IngR \times EF \times ED / BW \times AT_{nc} \times 10^{-6} \quad (4)$$

$$CDI_{inh\ nc}\,(\text{mg m}^{-3}) = C \times EF \times ET \times ED / PEF \times 24 \times AT_{nc} \quad (5)$$

$CDI_{dermal\ nc}$ (mg $kg^{-1}day^{-1}$) = $C \times SA \times AF \times ABS_d \times EF \times ED / \times BW \times AT_{nc} \times 10^6$ (6)

$HI = HQ_{ing} + HQ_{inh} + HQ_{dermal}$ (7)

$HI = CDI_{ing\ nc}/R_fD_{ing} + CDI_{inh\ nc}/RfC_{inh} + CDI_{dermal\ nc}/R_fD_{ing} \times ABS_{GI}$ (8)

The carcinogenic risk was calculated for the life time exposure, estimated as the increment probability of an individual developing cancer over a lifetime as a result of total exposure to the potential carcinogens (Cd & Cr). The carcinogenic hazard risk (TOTAL RISK) was calculated using Equation (9)-(14) adapted from USDOE (2011) [32]:

$CDI_{ing\ ca}$ (mg $kg^{-1}day^{-1}$) = $C \times IngR_{adj} \times EF / AT ca \times 106$ (9)

$IngR_{adj} = (ED \times IngR)_{child}/BW_{child} + (ED_{adult}-ED_{child}) \times IngR_{adult} / BW_{adult}$ (10)

$CDI_{inh\ ca}$ (μg m^{-3}) = $C \times EF \times ET \times ED \times 10^3/PEF \times 24 \times AT_{ca}$ (11)

$CDI_{dermal\ ca}$ (mg kg^{-1} day^{-1}) = $C \times ABS_d \times EF \times DEF_{adj} / AT_{ca} \times 10^6$ (12)

$DFS_{adj} = (ED \times SA \times AF)_{child} / BW_{child} + (ED_{adult}-ED_{child}) \times (SA \times AF)_{adult}/BW_{adult}$ (13)

$Total\ risk = risk_{ing} + risk_{inh} + risk_{dermal}$

$Total\ risk = CDI_{ing\ ca} \times CSF_{ing} + CDI_{inh\ ca} \times IUR + CDI_{dermal\ ca} \times CSF_{ing} /ABS_{GI}$ (14)

Where CDI_{ing}, CDI_{inh} and CDI_{dermal} were the chronic daily dose contacted through oral ingestion (mg $kg^{-1}day^{-1}$), inhalation (mg m^{-3} for non-cancer and µg m^{-3} for cancer), and dermal contact with water particles (mg $kg^{-1}day^{-1}$), respectively. R_fD_{ing} and R_fC_{inh} were the chronic oral reference dose and chronic inhalation reference concentration, respectively. CSF_{ing} and IUR were the chronic oral slope factor and chronic inhalation slope factor. The definitions and values of other parameters are shown in Tables 2 and 3.

Table 1: Measurements for system suitability of heavy metals in water samples using AAS

Sr.no.	Heavy metals	Standard Solution concentration (mg/l)	Mean absorbance	Standard deviation	Relative standard deviation
1.	Zn	0.5	0.038583	0.0027	0.5
2.	Cu	1	0.064181	0.0058	0.7
3.	Pb	0.5	0.034611	0.0026	0.6
4.	Mn	2	0.047403	0.0036	0.9
5.	Fe	5	0.233847	0.0011	0.4
6.	Al	1.0	0.0025	0.000014	0.6
7.	Hg	0.5	0.004111	0.00039	1.2
8.	Ni	5	0.050833	0.0049	0.7
9.	As	10	0.027639	0.0011	2.3
10.	Cd	0.5	0.014292	0.0005	1.7

Table 2: Definition and values of factor for health risk assessment

Factors	Definition	Values		References
		Children	Adults	
C (mg kg^{-1})	Heavy metal concentration			
AF (mg cm^{-2})	Water to skin adherence factor	0.2	0.07	(USDOE 2011) [32]
BW (kg)	Average body weight	16.2	61.8	(WHO 2006)
ED (year)	Exposure duration	6	30	(USDOE 2011) [32]
IngR (mg day^{-1})	Water ingestion rate for receptor	200	100	(USDOE 2011) [32]
SA (cm^2 $event^{-1}$)	Skin surface area available for exposure	2800	5700	(USDOE 2011) [32]
EF (d $year^{-1}$)	Exposure frequency	350		(USDOE 2011) [32]
ET (h day^{-1})	Exposure time	24		(USDOE 2011) [32]
AT_{nc} (day)	Averaging time for non-carcinogenic	ED × 365		(USDOE 2011) [32]
AT_{ca} (day)	Averaging time for carcinogenic	LT × 365		(USDOE 2011) [32]
DES_{adj} (mg year kg^{-1} day^{-1})	Water dermal contact factor age adjusted	362		Equation (13)
$IngR_{adj}$ (mg year kg^{-1} day^{-1})	Water ingestion rate age adjusted	113		Equation (10)
LT (year)	Life time	72		(WHO 2006)
PEF (m3 kg^{-1})	Water to air particulate emission factor	1.36×10^9		(USDOE 2011) [32]
ABS_d	Dermal absorption factor	0.001		(USEPA 2011) [33]

Table 3: Toxicological parameters for different heavy metals of health risk assessment

Elements	RfDing (mg kg^{-1}day^{-1})	RfCinh (mg m^{-3})	ABSGt	CSFing (mg kg^{-1} day^{-1})	IUR (μm^{-3})
Zn	0.3	0.004	1	-	-
Fe	0.7	-	1	-	-
Cu	0.004	0.0014	1	-	-
Cd	0.001	0.00001	0.25	-	0.0018
Cr	0.003	0.0001	0.013	0.005	0.012
Mn	0.14	0.00005	1	-	-
References	(USDOE 2011) [32]	(USDOE 2011) [32]	USEPA (2011) [33]	(USDOE 2011) [32]	(USDOE 2011) [32]

3. Result and discussion

Heavy metal contamination concentration in water samples

The descriptive statistics of different metals in the water samples of the studied area are given in Heavy metal concentration in water samples ranges from 0.1 to 0.22 mg/l for Zn, 0.011 to 0.059 mg/l for Cu, 0.094 to 0.01 mg/l for Pb, 0.021 to 0.059 mg/l for Mn, 0.02 to 0.3 mg/l for Fe, 0.001 to 0.007 mg/l for Al, 0.001 to 0.008 mg/l for Hg, 0.1 to 0.9 mg/l for Ni,0.1 to 0.5 mg/l for As, 0.006 to 0.02 mg/l for Cd, respectively. Since no information is available regarding the presence of trace element contents in Baloda Bazar district, the metal contents for this study were compared with the levels occurring in unpolluted water according to Kabata Pendias and Pendias (1992) [10] and the Indian Standards (Awashthi 2000) [2]. The concentration of metals in the studies water is compared with National and International guidelines for metals in water (Table 4). The mean concentration of Zn, Cu, Pb, Mn, Fe are found higher than the limit of heavy metal in water samples prescribed by USEPA (1999) [33]. This may be due to the automotive transport, chemical pesticides used in water samples and the application of numerous bio molecules, livestock, manures, composts and municipal sewage sludge. All values of Fe and Mn are found above the limit suggested by USEPA (1999) [33]. However, only the range concentration of Fe is maximum than the range suggested by Indian Standards (Awashthi 2000) [2]. Agriculture and food waste, municipal wastes, fertilizers and atmospheric deposition can result in high concentration of lead in water samples.

Table 4: Concentration of heavy metal in river water samples of Baloda Bazar district, Chhattisgarh, India

Heavy metal	Descriptive statistics (mg/l)				Geochemical background^a	Limits suggested by various agencies		
	Min.	Max.	Mean±S.D.	G.M.		Indian standards^b	USEPA^c	World range^d
Zn	15	277	112±53	111	127	300-600	110	17-125
Cu	0.5	182	37±39	22	0.00	135-200	16	6-60
Pb	0.05	165	65±37	73	0.00	-	0.5	-
Mn	58	150	350±140	334	0.69	-	30	-
Fe	719	13202	7403±4320	7329	35900	75-150	30	-
Al	0.2	60	64±33	62	16050	5.0	-	-
Hg	0.001	25	23±98	13	0.00	0.001	-	-
Ni	21	244	92±73	54	0.00	-	25	5-120
As	0.6	0.9	31±62	63	0.00	-	-	-
Cd	0.2	0.9	0.4±0.2	0.4	0.00	3-6	0.6	0.07-1.1

Values are given in milligram per liter (mg/l) ^{a} Martin and Meybeck (1979) [17] ^{b} Indian standard (Awashthi 2000) [2] ^{c} USEPA (1999) [33] ^{d} World range (Kabata-Pendias and Pendias 1992) [10]

Health risk assessment results

Non carcinogenic doses and hazard exposure of metals in the water samples for both children and adults. The maximum exposure doses for children and adults are 0.19 and 2.4 mg kg^{-1}day^{-1}, respectively both for Fe. The chronic oral ingestion doses for children are 2-5 orders of magnitude higher than by the other two doses. While the chronic oral ingestion doses for adults are 2-4 orders of magnitude higher than by the other two doses. The inhalation non carcinogenic doses have been varied from 1.4 to 1.1 for children and for adults. The dermal non carcinogenic doses have been varied from 6.6 to 5.0 for children and from 1.2 to 9.3 for adults. The non-carcinogenic doses of the different exposure pathways indicated that $CDI_{ing\ nc}$ >> CDI_{dermal} >> $CDI_{inh\ nc}$ for children which is possible due to hand to mouth habits of children. The total doses due to the sum of ingestion, inhalation and dermal doses are varied from 1.4 to 0.18 for children and are varied from 1.4 to 2.3 for adults. Oral ingestion hazard quotient HQ_{ing} is the main route of exposure to metals in water that leads to a higher risk and followed by dermal absorption hazard quotient except for Mn, similar to other studies (Ferreira Baotusta and De Miguel 2005; Zheng *et al*. 2010; Luo *et al*. 2012) [7, 12, 16] for children.

The concentration of all studied heavy metal in water samples both for children and adults are well below the unity, hence there is no non carcinogenic hazard index for both adults and children. The hazard quotient values for ingestion HQing and dermal absorption HQdermal and total hazard index values of all analyzed metals for children are higher than for adults, indicating that children may have more potential non cancer risk than adults. The statistical parameters for carcinogenic doses of ingestion, inhalation and dermal absorption. The average highest values of oral ingestion, inhalation and dermal absorption carcinogenic doses were 1.1, 4.0 and 4.0, respectively.

4. Conclusion

As a lower dense and growing area, Kasdol area, Baloda Bazar district, Chhattisgarh, India. The order of the mean concentrations of tested heavy metals as Pb>Fe>Mn>Cu>Zn>Al>Hg>Ni>As>Cd. This may be due to the automotive transport, pesticides used in water samples and the application of numerous bio solids, livestock, manures, composts and municipal sewage sludge. The contour distribution in the water samples is uniform for Pb, Fe, Mn, Cu and Zn over the whole studied area except Fe and Pb, respectively. The values of iron and manganese in water samples for contamination factor and index of geo accumulation face low contamination. We proposed to take sample of at least 3 areas and about samples of each two months in a year. Using clustering analysis method and typical AAs method compare the standard of the heavy metals in the river water of Jonk Rive and increasing contamination of heavy metals of the river. The Baloda Bazar district is free from any kind of health risk due to carcinogenic Pb in the water samples. Hence, the studied area is safe from health hazard point of view.

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6. Refrenceces

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