A study on trace elemental contamination and correlation of surface and sub-surface water system in parts of central – east India

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ABSTRACT

This paper presents the trace metal contamination of the Chouky-Rajnandgaon- Durg, a Central - East Indian region. The study area was divided into three different zones. Zone A is the mineralised zone; zone B is the catchments of river Shivnath and the zone C is Kotri-Indrawati catchments. The results give the elemental composition of the surface water indicating high levels of manganese and iron and noticeable presence of copper and zinc while the cadmium, chromium and lead were below the detection limit. The statistical analysis shows wide variability in geochemical sources and chemical interactions, which takes place in natural water system explainable due to negative skewness. The Kotri- Indra- wati catchments and the mineralized zones are enriched with harmful trace elements whereas the catchments of river Shivnath are more enriched with alkaline and alkaline earth metals. The surface water is highly enriched with manganese and iron as compare to seawater.
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INTRODUCTION

Metals are found throughout the earth, in rocks, soils and sediments primarily trapped in some stable form. Yet, through natural processes such as weathering and erosion, small amounts of metals are removed from bedrock and are allowed to circulate in water and air. This is essential because many biochemical processes require a given amount of many of these metals. So, even if a trace metal is toxic at high concentrations, it may be needed in small quantities to maintain life. Therefore, the biogeochemical cycle which exists ensures that the distribution of any given metal within an ecosystem be held rela-

tively constant over time. Quantities of most trace metals introduced into the environment by anthropogenic sources now far outweigh natural sources. Mining of ores, in addition to smelting and other purification practices, result in the metal being released from its stable forms and into the environment. This has produced a situation in which the natural biogeochemical cycle has been overwhelmed.

The same CRD region is contaminated by higher arsenic level in surface water[3] which can be categorised exceeding the ecological risk benchmarks. This paper has targeted the other trace elements to see if their concentration is also elevated in the studied region. The aim of this comparison is

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aimed at finding if these values exceed the Ecological Risk Screening values. The ERS (Ecological Risk Screening) is the conservative assessment to evaluate the need to conduct a detailed baseline ecological risk assessment and if an ERA is needed, to focus the scope of the assessment. The overall goal of this study is to determine the significance of contamination of the surface water system of the CRD region.

METHODS AND MATERIALS

The sampling locations of this study are presented in the Figure 1(A and B).The trace elements zinc, iron, manganese, cadmium, chromium, lead and copper were analysed by atomic absorption spectrophotometer and aluminium was analysed by eriochrome cyanine–R method by UV-Visible spectroscopy and other parameters were analysed by different analytical methods.

RESULT AND DISCUSSION

The mean results of surface water analysis for three consecutive years i.e. 1999-2001 are presented in TABLES 1 to 3. The results present the level of trace metal contamination of the three zones. Briefly the zones are:

Zone-A: The mineralised zone at the divide of Shivnath-Kotri river catchments; Zone-B: Shivnath catchments downstream of these locations e.g. the Shivnath river system passing through the Chowki-Rajnandgaon-Durg is considered as B region and Zone-C: Kotri-Indrawati catchments and the general mineralised area e.g. Dongargarh, Manpur, Mohla etc. was considered as C region.

For the purpose, the sampling was conducted at least once a month in all three seasons. Surface water analysis for trace elements carry a special significance as any deterioration in its quality is likely to affect the aquatic life as characterised by ecological risk assessment but is even more important
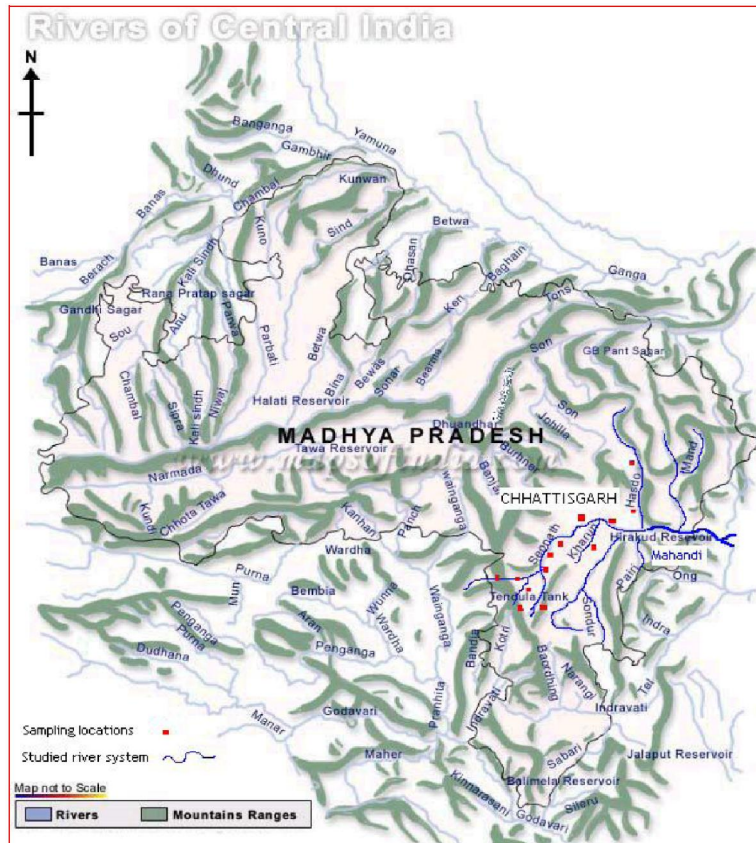

Figure1(a) : Map indicating the studied river systems and sampling locations along the Mahanadi River System. The most intensively studied river was Shivnath River
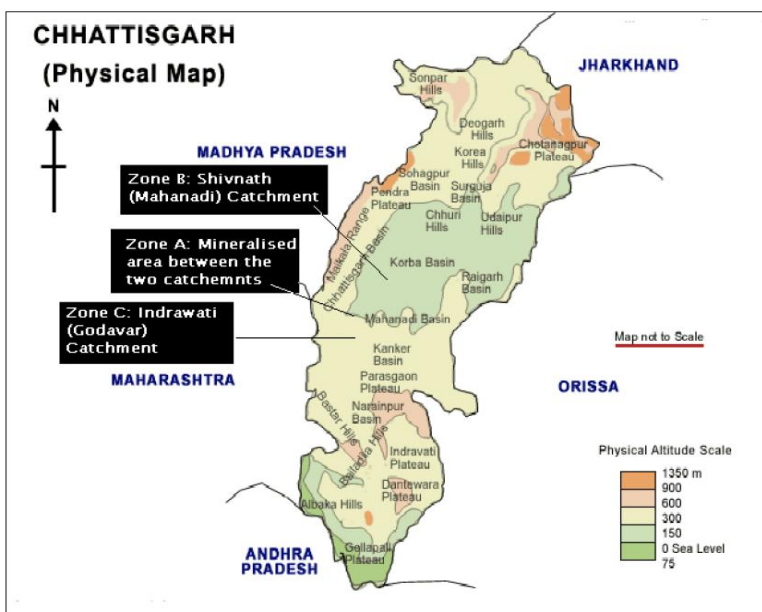
Figure1(b) : Graphical representation of the three zones as devised in this study

TABLE 1 : Trace metal contamination of Surface water in Zone A Yearly mean (µg/L) in three consecutive years

Trace Metals	1999	2000	2001
Al	260	810	745
Fe	BDL	2640	2050
Mn	80	1340	BDL
Cu	100	69	BDL
Zn	BDL	84	BDL
Cd	BDL	BDL	BDL
Cr	BDL	BDL	BDL
Pb	BDL	BDL	BDL
Ca	44600	80737	135450
Mg	129200	97342	49050
Na	26600	46820	22450
K	3670	5543	4320

TABLE 2 : Trace metal contamination of Surface water in Zone B Yearly mean (µg/L) in three consecutive years

TRACE METALS	1999	2000	2001
Al	114	BDL	1000
Fe	0	2375	1525
Mn	75	1720	BDL
Cu	BDL	BDL	BDL
Zn	BDL	20	BDL
Cd	BDL	BDL	BDL
Cr	BDL	BDL	BDL
Pb	BDL	BDL	BDL
Ca	84100	99650	64750
Mg	36520	78300	41900
Na	45000	51266	62000
K	2010	8800	7600

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TABLE 3 : Trace metal contamination of Surface water in Zone C Yearly mean (µg/L) in three consecutive years

TRACE METALS	1999	2000	2001
Al	120	BDL	900
Fe	350	1745	896
Mn	BDL	1720	560
Cu	BDL	BDL	BDL
Zn	BDL	ND	ND
Cd	BDL	BDL	BDL
Cr	BDL	BDL	BDL
Pb	BDL	BDL	BDL
Ca	99210	101995	88066
Mg	56200	109480	37633
Na	55000	62030	37566
K	11000	10440	4866

for the reason that a large number of people are directly dependent on it. In CRD (Chowki–Rajnandgaon-Durg) region all major towns and cities draw their drinking water from the Shivnath River only and hence, any deterioration in its water quality has a potential to cause problems in the various stages of purification, supply and consumption.

DATA ANALYSIS

A thorough analysis of the data was carried out on all results. The elemental composition of surface water shows high levels of manganese (Mn) and iron (Fe) and noticeable presence of copper (Cu), and Zinc (Zn). The values of cadmium (Cd), chromium (Cr), and lead (Pb) were below the detection limit. The skewness calculations in a data set characterise the degree of asymmetry of a distribution around its mean. Positive skewness indicates a distribution with an asymmetric tail extending toward values that are more positive. Negative skewness indicates a distribution with an asymmetric tail extending toward values that are more negative. In the data set generated in this study a marginal negative skewness has been noted which is explainable due to the wide variability in geo-chemical sources and chemical interactions which take place in natural water system.

The most mineralised zone, the zone of Shivnath-Kotri-River catchments shows a maximum level of aluminium at 810 µG/L, median of 745 µG/L and

mean of 605 µG/L. As far as asymmetry in the data set is concerned, this study has shown a wide variability, which is mainly the result of the meteorological effects, and further of the widely changeable geological pattern in the studied area. Standard deviation calculations on the data set show a moderate dispersion of the values around the mean, which symbolises equilibrium levels of these elements in the environment. Iron is the most abundant trace element at the divide of Shivnath-Kotri River catchments with a high standard deviation and sample variance. Manganese in this Zone is appreciable with a positive skewness symbolising a positive asymmetry in its distribution TABLE 4.

Zone B which is the catchments of River Shivnath downstream of these location presents a similar picture with slightly elevated levels of iron and manganese but lower levels of copper and zinc. This is plausible as being the downstream area of mineralised zone this area receives greater input of iron and manganese, which can travel longer distance compared to the copper, and zinc which are likely to be scavenged faster from the water column in view of they being micronutrient at lower concentrations TABLE 5.

Zone C which is in opposite direction to the Zone B and which is influenced by the partial runoffs from Zone A shows contaminations of Fe and Mn similar to the Zone A and also elevated levels of Cu and Zn. The asymmetry in the data set is low which means a normal distribution of the values and hence a more

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TABLE 4 : Statistical analysis of the data from Zone A in three different years

Trace metals	Mean	Standard error	Median	Standard deviation	Sample variance	Skewness	Range	Minimum
Al	605	173.52	745	300.54	90325	-1.64	550	260
Fe	1563.33	800.01	2050	1385.65	1920033	-1.39	2640	0
Mn	473.33	433.95	80	751.62	564933.3	1.71	1340	0
Cu	56.33	29.55	69	51.19	2620.33	-1.05	100	0
Zn	28	28	0	48.49	2352	1.73	84	0
Cd	BDL	0	0	0	0	NA	0	0
Cr	BDL	0	0	0	0	NA	0	0
Pb	BDL	0	0	0	0	NA	0	0
Ca	86929	26408.25	80737	45740.42	2.09E+09	0.59	90850	44600
Mg	91864	23298.87	97342	40354.83	1.63E+09	-0.59	80150	49050
Na	31956.67	7527.61	26600	13038.2	1.7E+08	1.54	24370	22450
K	4511	549.06	4320	950.99	904393	0.87	1873	3670

TABLE 5 : Statistical analysis of the data from Zone B in three different year

Trace metals	Mean	Standard Error	Median	Standard Deviation	Sample Variance	Skewness	Range	Minimum	Maximum
Al	371.33	316.05	114	547.42	299665.3	1.65	1000	BDL	1000
Fe	1300	694.77	1525	1203.38	1448125	-0.82	2375	BDL	2375
Mn	598.33	561.25	75	972.11	945008.3	1.72	1720	BDL	1720
Cu	BDL	0	0	0	0	0	0	BDL	BDL
Zn	6.67	6.67	0	11.55	133.33	1.73	20	BDL	20
Cd	BDL	0	0	0	0	0	0	BDL	BDL
Cr	BDL	0	0	0	0	0	0	BDL	BDL
Pb	BDL	0	0	0	0	0	0	BDL	BDL
Ca	82833.33	10094.65	84100	17484.45	3.06E+08	-0.32	34900	64750	99650
Mg	52240	13122.23	41900	22728.37	5.17E+08	1.62	41780	36520	78300
Na	52755.33	4963.65	51266	8597.30	73913585	0.76	17000	45000	62000
K	6136.67	2092.21	7600	3623.82	13132033	-1.52	6790	2010	8800

sTABLE levels of contamination. It can be surmised that it is possible that Zone C will be even more contaminated if the study is extended in its area in the Zone C as the geographical area covered in the present study is not very large in Zone C as compared to the Zone B. Similarity in the trends of Cu and Zn in Zone A and C are a clear indication of this fact TABLE 6.

TABLE 7 presents the final statistical analysis of the combined results taking all zones and all season together. This analysis makes it very clear that the surface water in a mineralised zone and its adjoining areas possesses higher values of the trace elements. In this zone the values of dissolved alu-

minium can go to as high as 1 mg/L, iron to 2.65 mg/L, manganese to 1.72 mg/L, copper to 0.1 mg/L, and zinc to 0.08 mg/L. The occurance of lead, chromium and cadmium has not been noted in dissolved state. This zone can also be characterised by high maximum values of calcium, magnesium, sodium and potassium with maximum at 135, 129, 62 and 11 mg/L respectively. The elevated concentration of these elements is characteristic feature of metal abundance owing to the geological anomaly and weathering reactions. The important point is that in such an environment any anthropogenic interference is likely to cause extremely adverse impact on the environment as the environment is already in a stressed condi-

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TABLE 6 : Statistical analysis of the Zone C in three different year

Trace metals	Mean	Standard error	Median	Standard deviation	Sample variance	Skewness	Range	Minimum	Maximum
Al	438.78	138.61	260.00	415.84	172923.40	0.25	1000.0	0.00	1000.0
Fe	1286.78	337.61	1525.0	1012.83	1025815	-0.13	2640.0	0.00	2640.
Mn	610.56	255.03	80.00	765.10	585377.80	0.77	1720.0	0.00	1720.0
Cu	18.78	12.69	0.00	38.06	1448.44	1.80	100.00	0.00	100.00
Zn	14.86	11.86	0.00	31.39	985.14	2.37	84.00	0.00	84.00
Cd	BDL	NA	NA	NA	NA	NA	NA	NA	NA
Cr	BDL	NA	NA	NA	NA	NA	NA	NA	NA
Pb	BDL	NA	NA	NA	NA	NA	NA	NA	NA
Ca	35491.2	8495.14	35491.2	25485.43	650000000	0.06	90850.	17840.00	54180.
Mg	16243.75	11464.77	16243.75	34394.32	1180000000	0.66	92680.	8399.6	40798.
Na	45414.67	4746.89	46820.	14240.68	203000000	-0.50	39580.	22450.00	62030.
K	6472.11	1045.98	5543.0	3137.95	9846726	0.23	8990.0	2010.0	11000.

TABLE 7 : Final statistical analysis of the results of Zone A, B, C in three different Years

Trace metals	Mean	Standard error	Median	Standard deviation	Sample variance	Skewness	Range	Minimum	Maximum
Al	438.78	138.61	260.00	415.84	172923.40	0.25	1000.0	0.00	1000.0
Fe	1286.78	337.61	1525.0	1012.83	1025815	-0.13	2640.0	0.00	2640.
Mn	610.56	255.03	80.00	765.10	585377.80	0.77	1720.0	0.00	1720.0
Cu	18.78	12.69	0.00	38.06	1448.44	1.80	100.00	0.00	100.00
Zn	14.86	11.86	0.00	31.39	985.14	2.37	84.00	0.00	84.00
Cd	BDL	NA	NA	NA	NA	NA	NA	NA	NA
Cr	BDL	NA	NA	NA	NA	NA	NA	NA	NA
Pb	BDL	NA	NA	NA	NA	NA	NA	NA	NA
Ca	35491.2	8495.14	35491.2	25485.43	650000000	0.06	90850.	17840.00	54180.
Mg	16243.75	11464.77	16243.75	34394.32	1180000000	0.66	92680.	8399.6	40798.
Na	45414.67	4746.89	46820.	14240.68	203000000	-0.50	39580.	22450.00	62030.
K	6472.11	1045.98	5543.0	3137.95	9846726	0.23	8990.0	2010.0	11000.

tion[3,4].

If the relative contribution of the three zones to the total observed mean values is considered then the Figure 2 clearly show that Zone A and C are the major contributor to the observed final mean in case of harmful trace elements viz. Al, Mn, Cu and Zn whereas the Zone B is more enriched with alkaline and alkaline earth elements viz Na, K, Ca, and Mg.

A correlation study was conducted to see the inter-relationship of the elements analysed. This study is of particular importance to trace the origin and interactions of the measured parameters. Based on the results presented in TABLE 8 it can be seen that Fe and Mn have shown a strong positive correlation. Such a correlation is clearly expected in a region, which is geologically mineralised with iron

ore. This correlation also proves the accuracy of the analyses from geochemical signatures point of view. A positive correlation of Zn with Mn is also a proof of the correctness of data set because in geo-chemical analyses the Mn levels is taken as an indicator of the trace contamination. A significant negative correlation of calcium with copper is significant as it indicates a differing origin of the two. Negative correlation means that the higher levels of Ca will result in the lowering of Cu in the water body. Such a relationship could be expected in different mode of origin of the two elements. The origin of copper in water bodies in dissolved state can be expected in more acidic environments where the natural availability of calcium could be lower hence the inverse relationship is well expected and justified.

The forgoing data and discussions make it clear that
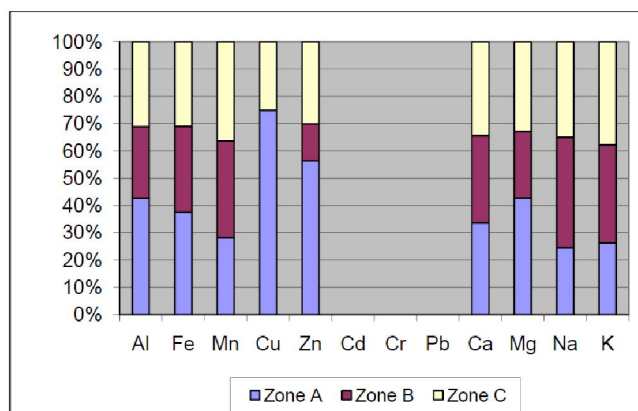
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Figure 2 : Percentage Contribution of the three zones to final mean of trace metals

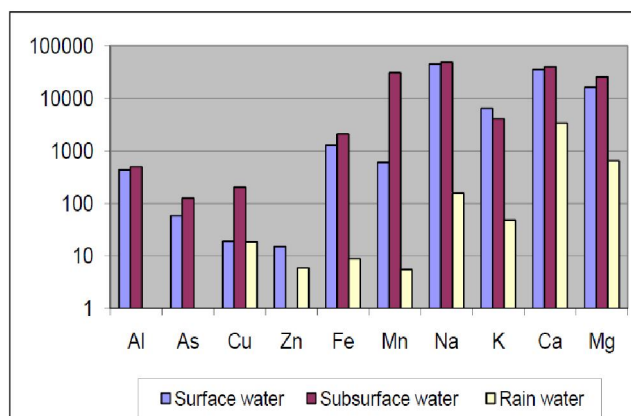

Figure 3 : Segmental differences in the elemental contaminations in studied region

TABLE 8 : Correlation matrices of the three different zones

	Al	Fe	Mn	Cu	Zn	Cd	Cr	Pb	Ca	Mg	Na	K
Al	1											
Fe	0.28	1										
Mn	-0.31	0.66	1									
Cu	0.06	-0.11	-0.01	1								
Zn	0.3	0.63	0.71	0.41	1							
Cd	0	0	0	0	0	1						
Cr	0	0	0	0	0	0	1					
Pb	0	0	0	0	0	0	0	1				
Ca	-0.09	0.44	0.17	-0.64	-0.05	0	0	0	1			
Mg	-0.38	0.12	0.47	0.74	0.39	0	0	0	-0.36	1		
Na	-0.22	0.18	0.4	-0.41	0.13	0	0	0	-0.09	-0.06	1	
K	-0.32	0.29	0.42	-0.36	0.01	0	0	0	0.24	0.16	0.72	1

the surface water of the studied region is contaminated with trace elements. Then it would be appropriate to compare the levels of studied elements with the same existing in other segments of environment. For the purpose the levels of trace elements in the groundwater of a few locations in Zone A and C were measured. Detail of these analyses is not being presented as that is beyond the scope of this paper. Similarly, Pandey (1998) had reported the levels of trace elements in the rainwater of Bhilai city.

The segmental differences in the contamination is clearly visible in Figure 3 wherein it can be seen that subsurface water is maximum contaminated and its levels clearly appear to affect the surface water quality of the region. This emphasis on the subsurface origin of contaminant is based on the fact that

all streams in the area are of "gaining type" and hence the groundwater water is providing the base flow to the streams and rivers of the region. In this situation the contamination of surface water derives from groundwater and not vice-versa.

The above conclusion can be further tested by carrying out a correlation analysis. The result of such an analysis is presented in TABLE 9. This calculation shows that the result of trace elements in surface water and sub-surface water correlate significantly and positively (+0.87), whereas the same is not the case if sub-surface water and rainwater are concerned. Secondly, it can also be seen that there exists a correlation (+0.5) between the rain water and surface water but the value of this correlation is lower. This indicates that in an industrially contami-

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TABLE 9 : Correlation matrices in the three environmental segments

	Surface water	Subsurface water	Rain water
Surface water	1.00		
Subsurface water	0.87	1.00	
Rain water	0.56	0.48	1.00

TABLE 10 : Enrichment factors for trace ions (μg/l)in surface water and their source apportionment

Trace metals	Annual mean surface water	Mean sea level	Na in sea	Na in surface water	Enrichment factor	SSF	NSSF
Fe	1300.00	2.000	10770000	45414.67	154146.23	0.001	99.999
Mn	598.33	0.200	10770000	45414.67	709467.89	0.000	100.000
Cu	0.00	0.250	10770000	45414.67	0.00	0.000	100.000
Zn	6.67	4.900	10770000	45414.67	322.65	0.309	99.691
Pb	0.00	0.030	10770000	45414.67	0.00	0.000	100.000

nated location the rainwater can also be a source of the pollution of the surface water. However, the dilution effects of heavy rain etc. lower the significance of such a correlation.

A fourth dimension could be added to make the picture complete is the marine contribution to the observed levels of trace elements in rainwater and surface water. Pandey (1998) had studied the marine and non-marine contribution to the ionic concentration and the enrichment factors observed in the rainwater of Bhilai region by comparing the seawater and rainwater taking sodium as a reference material. Bhilai falls in Zone B area of this study and provides an opportunity for comparing the relative contribution of rain to the levels of contaminants.

Enrichment factors (EF) of the ions in the rain samples, taking sodium as the indicator species, can be calculated using the following formula:

$$EF_{Na}(X) = (X/Na)Rain / (X/Na)Sea$$

where X is the ion under investigation.

Marine contribution, i.e. sea salt fraction (SSF) and non-marine contribution, i.e., non-sea salt fraction (NSSF) of the various ions can be calculated as follows:

$$SSF = Na_R.(X/Na)sea.100 / X_R$$

Here X_R and Na_R, is the concentration of ion X and indicator element sodium (Na) in rainwater and (X/Na)sea is the mass ratio of ion X relative to Na in sea water.

NSSF= 100 - SSF.

In the EF calculations, a factor of unity shows that the particular ion is marine in origin and the EF greater than unity means that other sources are enriching the concentration. Enrichment calculations by Pandey (1998).

In this study, the enrichment calculation has extended with respect to the sea water, taking sodium as a reference element. Assumption of sodium as a reference element for enrichment calculation with respect to the seawater is debatable as the origin of this element is not entirely from sea. On the other hand, the geologic source can be expected to contribute significantly to the observed Na levels in surface water. Still when the values of trace elements are divided by the local sodium levels and then when these values are compared with that calculated from the seawater; a good comparison can be obtained. TABLE 10 presents the results of this calculation.

As expected this results show that the observed levels of the trace elements are almost entirely of non-marine in origin and the elemental levels of rainwater can be expected to affect the observed levels of trace elements in surface water if there exists a major source of aerial contamination.

Finally, the results of the surface water contamination of the region have been compared with the world median values of fresh surface water as reported by Bowen (1979). The results of % enrichment compared to the world median and enrichment

TABLE 11 : Calculation of %v enrichment and EF$_{(Na)}$ of the studied region

Trace metals	SW CE India (This paper)	World median (Bowen, 1979)	% Enrichment	Na in Surfacewater μg/L	Na in fresh water μ g/L	EF (Na)
Al	371.3	300	123.76	45414.67	6000	0.164
Fe	1300	500	260	45414.67	6000	0.344
Mn	598.33	8	7479.16	45414.67	6000	9.881
Cu	0	3	0	45414.67	6000	0
Zn	6.67	15	44.45	45414.67	6000	0.059
Pb	0	3	0	45414.67	6000	0

TABLE 12 : Comparison of the data with two surface water quality criterias

	Surface	Sub-Surface	Chronic (Spokane tribe)	Chronic (Usepa)	Acute (Spokane Tribe)	Acute (Usepa)
Al	438.78	500	690	87	750	750
As	58.18	127	-	150	340	340
Cu	18.78	205	40	8.6	18	13
Zn	14.86		170	120	120	120
Fe	1286.78	2130	1000	1000	-	-
Mn	610.56	30800	-	-	-	-
Na	45414.67	49200	-	-	-	-
K	6472.11	4100	-	-	-	-

factor with respect to the sodium has been presented in TABLE 11.

The result of this calculation shows that the surface water of the central-eastern Indian region comprising the area of Rajnandgaon-Durg region is enriched by 260%, 7479%, 123% and 44% for Fe, Mn, Al and Zn respectively. The corresponding enrichment values of these elements when compared taking sodium as an indicator species comes out to be 0.344, 9.881, 0.164 and 0.059 for Fe, Mn, Al and Zn respectively. This is a clear testimony of the higher levels of these elements in the studied region.

COMPARISON OF RESULTS TO ASSESS THE AQUATIC LIFE RISK

The Chronic water quality criteria also termed continuous criteria are defined as the highest concentration of a chemical to which aquatic life can be exposed for an extended period of time (4 days) without deleterious effects. Acute criteria also termed maximum criteria are defined as the highest concentration of a pollutant to which aquatic life can be exposed for a short period of time (1 hour) without

deleterious effect (USEPA, 1992).

The results obtained in this work when compared to two such criteria i.e. Spokane Tribe criteria (Chronic and Acute) and USEPA (Chronic and Acute) then it becomes clear that the the studied region is stressed owing to the certain trace elemental values close to or already surpassing the Chronic criteria TABLE 12.

A graphical presentation Figure 4 brings a clearer picture of the comparison. The results also necessitate a regular watch on the water quality parameters to avoid any environmental damage or probable human health
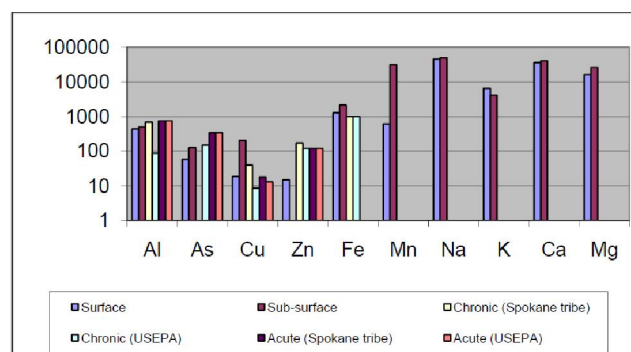

Figure 4 : Comparative graph of the surface water levels of trace elements in surface water with other water quality criteria

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impacts. The Fe and Mn are geological in origin as this area is geologically mineralised with iron ore it can be expected. However, their presence in dissolved form is anomalous and calls for more research on finding the reasons for their solubilisation. This study recommends further research to assess this anomaly, which may be able to generate a greater understanding of the processes governing mineralisation and their mobilisation.

CONCLUSION

Groundwater contamination in Rajnandgaon District of Chhattisgarh is the first major case of arsenic poisoning outside West Bengal and Bangladesh[3]. This work on trace metals on the rivers in the region is showing that even the surface water is being contaminated at certain locations in the region. The SHEONATH-HASDEO-MAHANADI RIVER SYSTEM and its watershed in CHHATTISGARH STATE is one of the most mineral rich part of India where huge mineral excavation and heavy engineering industries viz. power generation, steel and aluminium production are located. The Sheonath and Hasdeo rivers are the main source of water and carrier of the effluents of all these industries.

The contamination of surface water from other heavy metals of the region is established by the results obtained in this study. The surface water of mineralized zone and the Kotri- Indrawati catchments is richer in trace elements as compared to Shivnath catchments downstream of these locations. The Shivnath catchments downstream to Zone A is richer in alkaline and alkaline earth elements. The Fe and Mn are geological in origin as this area is geologically mineralised with iron ore it can be expected. However, their presence in dissolved form is anomalous and calls

for more research on finding the reasons for their solubilisation. The surface water of the studied region is highly influenced by the ground water of this region as it correlated highly (+0.87 correlation values). The trace metals values of surface water are non-marine in origin. The comparison of reported results with aquatic life risk values the Al, Cu and Fe has crossed the chronic level while the arsenic is nearer to the chronic level and Zn is on safe level of chronic criteria. These results necessitate a regular surveillance of the surface water parameters to avoid any aquatic and human health impacts.

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