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EVALUATION AND SPATIAL DISTRIBUTION OF HUMAN HEALTH RISK ASSOCIATED WITH HEAVY METALS IN SURFACE AND GROUND WATER FROM WASTE DISPOSAL SITE AT KHULNA

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ABSTRACT

The heavy metal releases from waste disposal site contains extensive ranges of carcinogen and non-carcinogenmetal compounds that signify a potential risk to public health. The main focus of this study was to evaluate the non-carcinogen health risk associated withheavy metals in surface and groundwater nearby waste disposal site. To these attempts, fifteen surface water and fifteen groundwater samples were collected from different selected production wells located adjacent to the waste disposal site at Rajbandh, Khulna, Bangladesh.In the laboratory, the concentrations ofheavy metals of Fe, Mn, Cr, Cu, Pb, Zn, Ni, Cd, Na, K, Ca and As in water were measured through the standard test methods. To assess the health risk, chronic daily intake (CDI), hazard quotient (HQ) and hazard index (HI) were computed using exposure and risk models proposed by US.EPA (1989).The exposure routessuch as dermal and ingestion were considered. The inhabitants were categorized as adult and child. According to US.EPA, limit of HI/HQfor non-carcinogen is less than unity and carcinogen risk is more than unity.Result reveals that the values of CDI, HQ and HI for metal in surface and ground water for rainy season was found comparatively higher than that of dry reason. Results indicated that the values of HQ and HI were found to be higher for child than that of adult as well as reasonable maximum exposure (RME) displayed higher values of HI than that of central tendency exposure (CTE).In this study, Pearson's correlation and principal component analysis (PCA) were performed using XLSTAT and results indicated that Fe, Mn, Cu and Ca were generated from anthropogenic sources except Cd, Cr, Ni, Pb, K and As from natural sources. The concentration of heavy metals, CDI, HQ and HI was distributed spatially. The uncertainty of exposure and risk parameters were analyzed using 1-D Monte Carlo Simulation @risk 7.5 with 10000 iterations.

Keywords: *Waste disposal site, water, chronic daily intake, reference dose, hazard quotient, hazard index, health risk.*

1. INTRODUCTION

Decision making around the disposal of municipal solid waste (MSW) is complex and becomes more difficulty in developing countries. The only safe solution is to dispose in a way that environment is not affected. This requires significant investments from the already squeezed budgets of governments, which continue to have other pressing priorities for spending (Manyin et al., 2009). In most of developing countries MSW management services take third chance in municipal priorities after water supply and sanitation.Drinking water is a major issue in human life. No one can survive without drinking water. Bangladesh is vulnerable to water insecurity partially because of its environmental circumstances. Heavy metals are important pollutants in surface waters, causing persistent environmental hazardsthat can seriously harm human and ecological health (Lin et al., 2007;Atli et al., 2008; Perianez et al., 2009). Heavy metals in surface waters originatefrom natural processes, such as atmospheric deposition and geological weathering, and fromanthropogenic activities (in emissions such as industrial wastewater and domestic sewage). Thecontributions of these sources are different in different regions and in different seasons, so heavy metalconcentrations in surface water and groundwatercan vary both spatially and seasonally. Information on these variationsis important for decision makers involved in environmental risk management (Huang et al., 2012;Kumar et al., 2013;Li et al., 2010; Matache et al., 2009; Lenoble et al., 2013). Evaluating environmental impact of contaminants in soils must start with a robust determination of theirconcentration and spatial distribution.

GIS based spatial distribution map is generally used to display the distribution of metal contamination has been widely used toassist the interpretation of environmental data and to distinguish between natural and anthropogenic inputs(Manta 2002).There are two main sources of heavy metals in the soil (Li et al.,2009b): (i) natural background, which represents the heavy metalconcentration derived from parent rocks; (ii) anthropogenic contamination, including agrochemicals, organic amendments, animal manure, mineral fertilizer, sewage sludge and industrial wastes. In the last severaldecades, the natural input of several heavy metals to soils due to pedogenesis has been exceeded by the human input, even on global andregional scales (Facchinelli et al., 2001; Nriagu and Pacyna, 1988).The main objectives of this study were to know the hazard index of selected chemical heavy metals in different routes within two seasons of a year at the Rajbandh near a landfill site, to know the sources of metal contamination by principal component analysis (PCA), to show the distribution of chemical metal concentrations, chronic daily intake (CDI) values of heavy metals, hazard index (HQ) values of heavy metals using ArcGIS.

2. METHODOLOGY

The sampling of water, measurement of the concentrations of heavy metals in water, models used for assessing health risk, principal component analysis and Monte Carlo simulation used in this study are presented and hence described in the following articles.

2.1 Sampling of Groundwater and Surface Water

Before collecting water samples the bottle was washed by distilled water several times. Then the bottles were air or sun dried. Then 2-3 mL a solution was used as preservative. The preservative was prepared by mixing concentrated nitric acid and distilled water at a ratio of 1:1. Then the bottle was kept for 24 hours at room temperature. After that the bottles were prepared for collecting water sample. In this study, fifteen groundwater samples were collected from selected production wells or tube wells located adjacent to the waste disposal site at Rajbandh of Khulna, Bangladesh. Moreover, fifteen surface water samples were collected from pond located at the mentioned locations. All the sampling points were gathered with the help of GPS and shown in Figure 1. These study periods covered both the dry and rainy seasons.

2.2 Laboratory Investigations

Both the water samples were collected from the site and then brought to DPHE, Khulna, Bangladesh. The concentrations of heavy metals of Fe, Mn, Cr, Cu, Pb, Zn, Ni, Cd, Na, K, Ca, As in water were measured through atomic absorption spectrophotometer (AAS).

2.3 Risk Assessment Methodology

In this study, for assessing health risk, the risk modelsproposed by Li and Zhang (2010); US.EPA (2004) and Wu et al. (2009)were used. Human can get exposed to water contaminations inthreemain pathways including dermal absorption, direct ingestion, and inhalation through nose.Among them dermal and ingestion are vital in health risk for both groundwater and surface water (US.EPA, 1989; US.EPA, 2004; Wu et al., 2009). In this study, for assessing health risk from ground and surface water, the exposure routes of dermal and ingestion were considered and hence discussed in the followings.

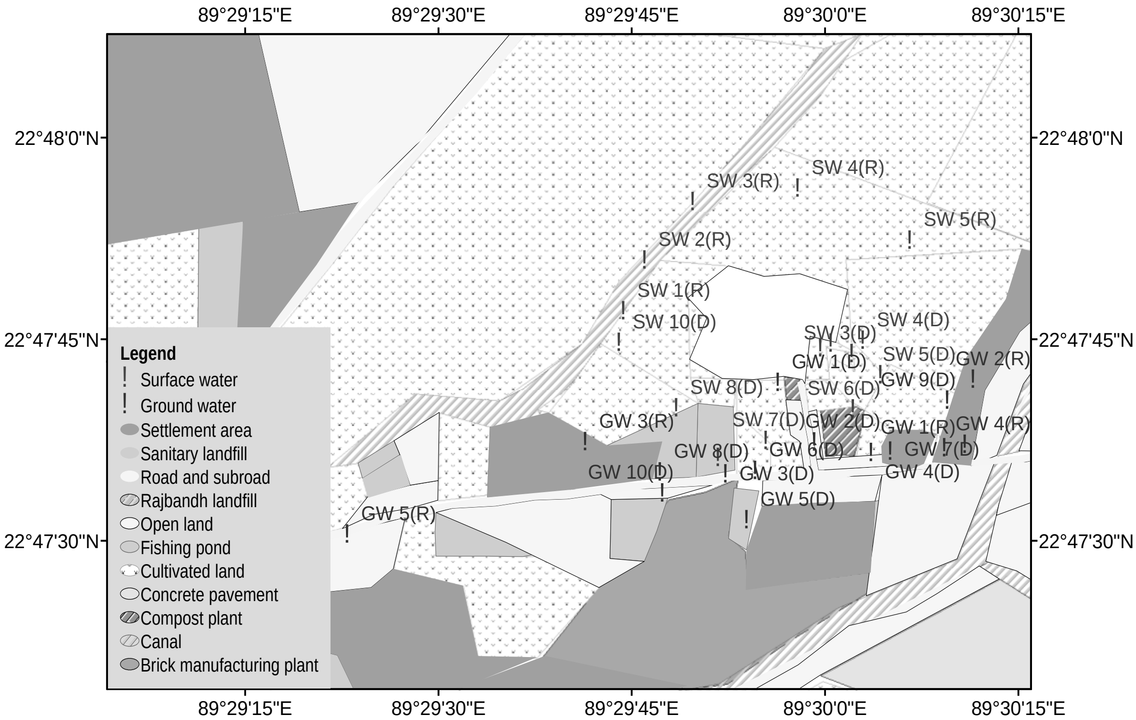

Figure 1: Surface and groundwater sampling locations nearby waste disposal site at Rajbandh, Khulna

2.3.1 ExposureModel for Incidental Ingestion

$$CDI_{ing} = \frac{C_W * CR * ABS_s * ET * EF * ED}{BW * AT}$$

Where, CDI_{ing} = Chronic daily intake for ingestion,Cw = metal concentration in water (mg/L),CR = contact rate (L/hr),ABS_s = absorption factor (%), ET = expose time (hr/event),EF = exposure frequency (days/year),ED = exposure duration (years),BW = body weight (kg),AT = average time (days).

2.3.2 ExposureModel for Dermal Absorption

$$CDI_{dermal} = \frac{C_W * CF * SA * PC * ABS_s * ET * EF * ED}{BW * AT}$$

Where,CDI_{derm} = chronic daily intake from dermal contact with heavy metals in water, Cw = concentration of estimated heavy metals in water (mg/L), SA = skin surface area availablefor contact (cm^2), CF = volumetric conversion factor for water (L/cm^2), PC = metal specific dermal permeability constant (cm/hr), ABS_s = absorption factor (%).

2.3.3 Risk Model for Hazard Quotient

$$HQ_{ing/dermal} = \frac{CDI_{ing/dermal}}{RfD_{ing/dermal}}$$

Where,HQ $_{ing/derm}$ is hazard quotient via ingestion or dermal contact (unitless) andRfD$_{ing/derm}$ is oral/dermal reference dose (mg/kg-day). The RfD$_{ing}$ and RfD$_{derm}$ values were obtained from literature elsewhere (Li and Zhang, 2010; US.EPA,1989; Wu et al., 2009; Liang et al., 2011).Recognized reference dose values are tabulated in Table1.

2.3.4 Risk Model for Hazard Index

$$HI = \sum_{i=1}^{n} HQ_{ing/dermal}$$

Where, HI$_{ing/derm}$ is hazard index via ingestion or dermal contact (unitless). When HQ/HI exceeds unity, there may be a concern for potential human health risks caused by exposure to non-carcinogenic elements (US.EPA, 1989).

Table1: The permeability, dermal and ingestion reference doses of heavy metals

Chemicals	**Permeability ,PC (cm/hr)**	**Reference dose, RfD$_{ing}$ (mg/kg-day)**	**Reference dose, RfD$_{derm}$ (mg/kg-day)**	**References**
Fe	1.00E-03	7.00E-01	1.40E-01	Li and Zhang,2010;US.EPA,1989;Wu et al.,2009;Liang et al., 2011
Mn	1.00E-03	2.40E-02	9.60E-04	
Cr(+6)	2.00E-03	3.00E-03	7.50E-05	
Cu	1.00E-03	4.00E-02	8.00E-03	
Pb	4.00E-03	1.40E-03	4.20E-04	
Zn	6.00E-04	3.00E-01	6.00E-02	
Ni	4.00E-03	2.00E-02	5.40E-03	
Cd	1.00E-03	5.00E-04	2.50E-05	
Na	1.00E-03	2.00E-02	1.60E-02	
K	1.00E-03	2.00E-02	1.60E-02	
Ca	1.00E-03	2.00E-02	1.60E-02	
As	1.00E-03	3.00E-04	1.23E-04	

3. Results and Discussion

The results of exposure and risk models, principal component analysis, geostatistical analysis and Monte Carlo simulation are discussed in the following sections.

3.1 Health Risk Analysis of Selected Chemicals in Water

The suitability of water samples mainly depends upon some heavy metals as stated earlier (WHO, 2008). From analysis of HQ it was quite clear that the water sample labeled as GW-1, collected in rainy season in RME condition was more hazardous for child .The computed HQ and HI for different heavy metals for child in RME for different exposure routes are provided in Table 2and computed HQ is represented in Figure 2. From Table 2 it was noticed that for total hazard, chemical hazardous sequel should be like as Ca> k> Na> Cd> Cr> Pb> Mn> Cu> As> Fe> Ni> Zn and the route of ingestion showed comparatively the higher hazardous effect than that of dermal route. Figure 2 shows that individual Ca possess more hazards in RME condition as it has maximum HQ value compared to others.

Table 2: Computed HQ and HI of GW-1 in rainy season (RME condition for child)

		HQ		
Chemicals Name	**Water Concentrations (mg/L)**	**Water Dermal contact**	**Incidental Water Ingestion**	**HI**
Fe	4.6	2.25E-02	4.38E-02	6.64E-02
Mn	0.351	2.51E-01	9.76E-02	3.48E-01
Cr(+6)	0.04	7.32E-01	8.90E-02	8.20E-01
Cu	0.95	8.14E-02	1.58E-01	2.40E-01
Pb	0.05	3.27E-01	2.38E-01	5.65E-01
Zn	0.15	1.03E-03	3.34E-03	4.36E-03
Ni	0.062	3.15E-02	2.07E-02	5.22E-02
Cd	0.029	7.96E-01	3.87E-01	1.18E+00
Na	12	5.14E-01	4.00E+00	4.52E+00
K	23	9.86E-01	7.67E+00	8.66E+00
Ca	65	2.79E+00	2.17E+01	2.45E+01
As	0.004	2.23E-02	8.90E-02	1.11E-01
Total HI		6.55E+00	3.45E+01	

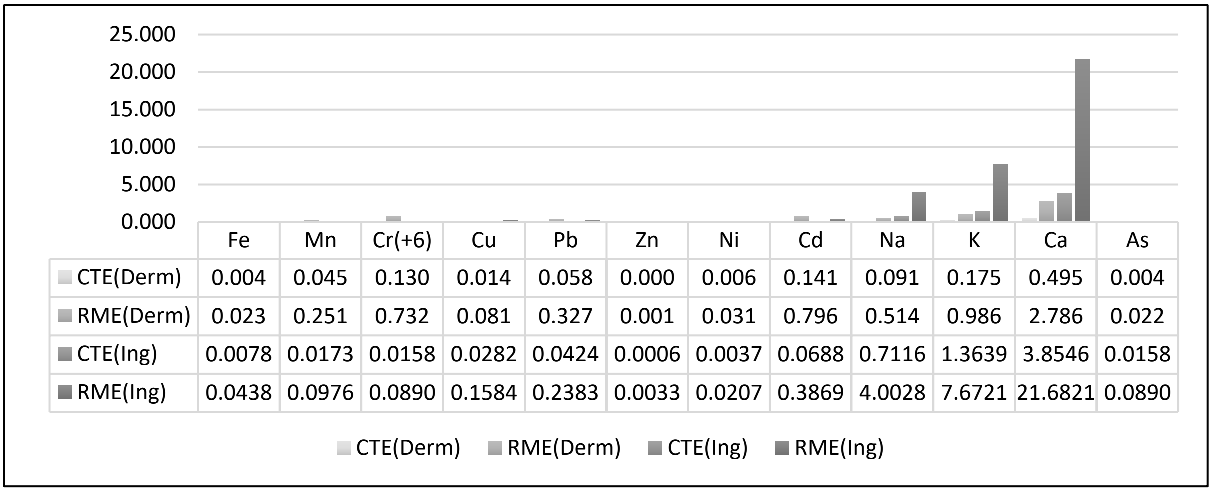

Figure 2: HQ for child in CTE and RME condition of groundwater for rainy season

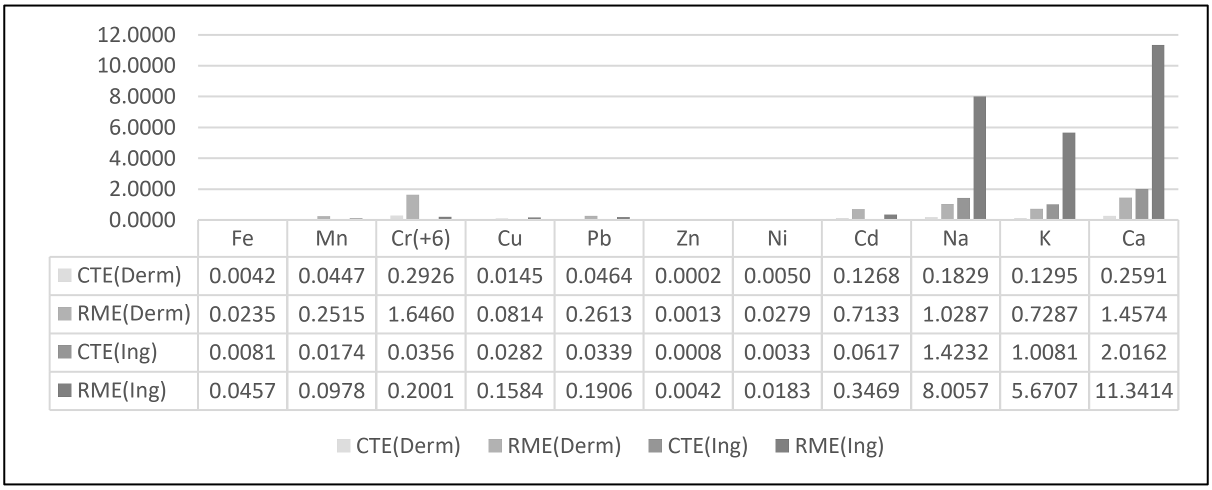

Figure 3: HQ for child in CTE and RME condition of surface water for rainy season

Figure 3 reveals that the values of HQ in case of ingestion route for surface water were found to be higher than that of dermal condition. In addition, RME showed the higher values of HQ in compare to CTE. The metal of Ca possess more hazards in RME condition during rainy season as it has maximum HQ value compared to others. Based on results of HQ, the chemical hazardous sequel should be like as Ca> Na> K> Cr> Cd> Pb> Mn> Cu> Fe> Ni> Zn and among two routes ingestion possess more hazardous effect for surface water.

Table-3: Correlation coefficient matrixfor CDI and HQ(CHILD/RME/INGESTION) of metals in ground water for dry season (below the diagonal) and rainy season (above the diagonal)

Variables	Fe	Mn	Cr(+6)	Cu	Pb	Zn	Ni	Cd	Na	K	Ca	As
Fe	**1**	0.681	-0.437	0.530	-0.241	0.743	0.264	-0.638	-0.901	-0.248	0.732	0.698
Mn	0.739	**1**	-0.363	0.970	-0.464	0.977	0.632	0.123	-0.461	0.438	0.969	0.513
Cr(+6)	0.404	0.586	**1**	-0.171	0.791	-0.326	0.492	0.306	0.617	0.483	-0.310	0.000
Cu	0.777	0.939	0.797	**1**	-0.330	0.948	0.767	0.309	-0.248	0.632	0.947	0.428
Pb	0.485	0.682	0.929	0.825	**1**	-0.309	0.233	-0.102	0.461	0.065	-0.279	-0.250
Zn	0.751	0.982	0.546	0.913	0.636	**1**	0.644	0.014	-0.472	0.373	0.999	0.464
Ni	-0.543	-0.642	-0.062	-0.474	-0.184	-0.758	**1**	0.379	0.098	0.816	0.652	0.456
Cd	0.611	0.144	0.434	0.397	0.285	0.142	0.046	**1**	0.751	0.844	0.022	-0.344
Na	-0.751	-0.565	-0.199	-0.536	-0.423	-0.605	0.504	-0.154	**1**	0.521	-0.448	-0.692
K	0.861	0.819	0.749	0.926	0.773	0.832	-0.529	0.607	-0.569	**1**	0.382	0.065
Ca	0.384	0.727	0.452	0.631	0.559	0.745	-0.716	-0.019	-0.157	0.619	**1**	0.437
As	-0.550	-0.138	-0.233	-0.301	-0.199	-0.241	0.320	-0.686	0.499	-0.553	0.009	**1**

3.2 Pearson's correlation Analysis

In this study, Pearson's correlationcoefficients were calculated forCDI and HQ of selected heavy metals. The values of correlation between the selected heavy metals for CDI and HQ values of selected heavy metals were shown in Table 3. Interpretation of Table 3, the interrelationship studies between different variables are very helpful tools in promoting research and opening new frontiers of knowledge. The study ofcorrelation reduces the range ofuncertainty associated with decisionmaking (Patil and Patil, 2010).

Fromresults of Pearson's correlations matrix on groundwater duringdry season, it was observed the highpositively correlated values between Cu and Mn(0.939), Pb and Cr(0.929), Zn and Mn(0.982), Zn and Cu(0.913), K and Cu(0.926), and in rainy season between Cu and Mn (0.970) , Zn and Mn (0.977), Ca and Mn (0.969) , Cu and Zn (0.948) , Ca and Cu (0.947), Ca and Zn (0.999) were observed. In contrast, in dry season low negatively correlated values between Cr and Ni (-0.062), Cd and Na (-0.154), Cd and Ca (-0.019), As and Mn (-0.138) were observed. Interpretation of Table 4, fromresults of Pearson's correlations matrix on groundwater for CDI values of different selected heavy metals duringdry season, it was observed the highpositively correlated values between Mn and Fe (0.74), Cu and Fe (0.78), Zn and Fe (0.75), K and Fe (0.86), Cu and Mn (0.94), Zn and Mn (0.98), K and Mn (0.82), Zn and Cu (0.91), Pb and Cu (0.82), K and Cr (0.75), Cu and Cr (0.82), Pb and Cr (0.93), K and Pb (0.77), K and Zn (0.83), Ca and Zn (0.75). In rainy season strongly correlated values between Mn and Cu (0.97), Cr and Pb (0.79), Cu and Zn (0.95), Mn and Zn (0.98), Cu and Ni (0.77), Cd and Na (0.75), Ni and K (0.82), Cd and K (0.84), Mn and Ca (0.97), Cu and Ca (0.95) were observed.

3.3 Principal Component Analysis (PCA)

Principal component analysis (PCA) was employed in order to understand the association among the heavy metals.PCA can be used to identify thesources of contamination (Facchinelli et al., 2001).Natural and anthropogenic sources are one of the root cause of metal element contamination which has caused widespread and variable the hazardous possibilities of environmental and health effect. Moreover, some previous investigations indicated first principal component (PC1) and second component (PC2) refers to the contamination of water due to anthropogenic or human activities and natural parent materials, respectively (Tahir et al., 2007).

Table4: Principal component loadings for heavy metals in groundwater for both seasons

	Dry season			**Rainy season**		
	PC1	PC2	PC3	PC1	PC2	PC3
Eigenvalue	7.1	1.9	1.5	5.9	3.7	1.7
Variability (%)	59.3	16.0	12.8	49.0	30.9	14.1
Cumulative %	59.3	75.3	88.1	49.0	79.9	94.0
Fe	0.86	0.28	-0.29	0.81	-0.48	0.31
Mn	0.91	-0.32	-0.01	0.98	0.14	-0.16
Cr(+6)	0.72	0.11	0.65	-0.36	0.67	0.63
Cu	0.96	-0.05	0.19	0.92	0.38	-0.13
Pb	0.79	-0.03	0.51	-0.43	0.34	0.73
Zn	0.93	-0.30	-0.13	0.97	0.12	-0.03
Ni	-0.63	0.37	0.59	0.61	0.70	0.37
Cd	0.44	0.81	0.12	-0.07	0.85	-0.49
Na	-0.65	-0.14	0.47	-0.61	0.76	-0.15
K	0.97	0.18	0.07	0.30	0.94	-0.09
Ca	0.69	-0.55	0.06	0.96	0.13	-0.03
As	-0.46	-0.70	0.34	0.65	-0.19	0.45

Principal component (PC) loadings for heavy metals in groundwater for both dry and rainy season are shown in Table 4.In dry season, three principal components (PCs) with eigenvalues>1that explains about 88% of the total variance of the dataset were obtained. PC1accounted as almost 59% of the total variance, and Fe, Mn, Cr, Cu, Pb, Zn, K and Ca are closely associated to it and these heavy metals were generated from anthropogenic sources. The PC2 accounted almost 16% of the total variance, and Cd is closely associated to it and generated from natural sources. In Rainy season, three PCs with eigenvalues>1 that explains about 94% of the total variance of the dataset were obtained. The PC1 accounted of almost 49% of the total variance, and Fe, Mn, Cu, Zn, Ca and Aswere closely associated to it. PC2 accounted almost 31% of total variance, and Cr, Ni, Cd, Na and K were closely associated to it.

Figure 4 shows the graphical representation of PCA output, at scree plot (Figures 4a and 4b) it represents the eigenvalues of all the nine factors where in Table 4 only three factors having eigenvalues>1 were shown. Figure 4c, represents the factor loading values for dry season. Where horizontal axis represents PC1 and vertical axis represents PC2, when the distance of a point is far from center and close to a positive side of an axis then the metal represented by this point is closely related to that factor what was represented by this axis. From Figure 4c, the heavy metals of Fe, Mn, Cr, Cu, Pb, Zn, K and Ca representing points are closely attached to positive PC1 axis staying near the circumferential line. So they aresourced from PC1 i.e from anthropogenic sources. When a point is exactly orthogonal to an axis, it represents that there is no relation between the point and axis. Accordingly, Figure 4d shown for rainy season represents the similar type of pattern as dry season.

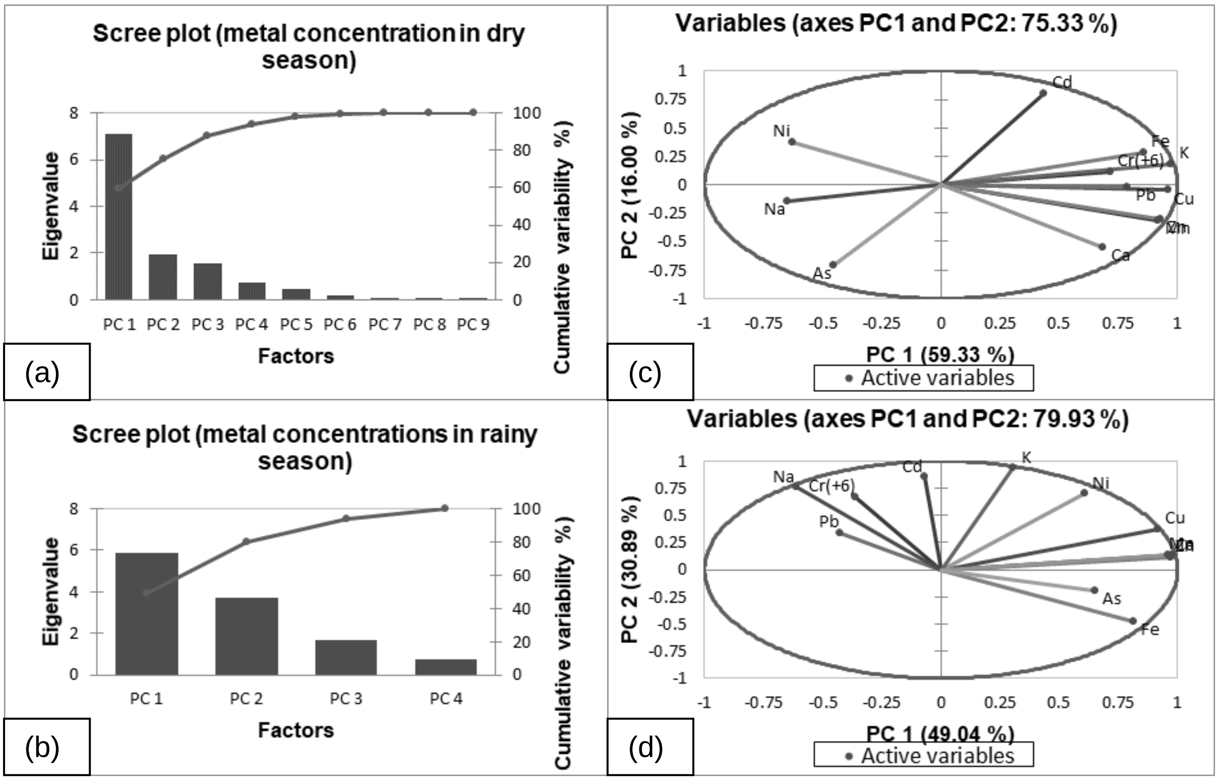

Figure4: Graphical representation of PCA output (a) scree plot for dry season; (b) scree plot for rainy season; (c) variables for dry season and (d) (c) variables for rainy season.

3.4 Spatial Distribution of Heavy MetalsinGroundwater

The spatial distribution of metal concentrations is a useful aid toassess the possible sources of enrichment and to identify hotspotswith high metal concentrations. The estimated maps of Ca and Cd are presented in Figure5a and Figure 5b, respectively; several hotspots of high metal concentration were identified by the geochemical maps. From the distribution Figure 5a, it is found that for Ca highest concentration lies in north and east side of disposal site (denoted by red 0) and south-west is less concentrated with Ca. The metal of Cd showed highest concentration is within the east side of selected disposal site (Figure 5b)

3.5 Spatial distribution of CDI values for surface water

Distribution of CDI for Ca, Fe, K and Na are represented in Figure 6. From the distribution map, it is found that for CDI value of Ca highest intake lies in north and west side of disposal site (denoted by red 0) and less intake values are found to spread within south side of the disposal site. Accordingly distribution for Fe, K and Na is presented in Figure 6. For all the heavy metals CDI values distribution criteria is almost similar which indicates that all the values are from the same source.

3.6 Spatial distribution of HQ values for Groundwater

The most serious issue is the hazard quotient values in health risk analysis. For groundwater a typical distribution of Ca, Cd, K, Na are shown in Figure 7. From distribution, north and east side of disposal site are most hazardous for Ca, and opposite sides are less hazardous.

But hazard quotient values for Cd is very much less compared to Ca. Similarly, for K north side is dangerous compared to other sides and for Na medium type of hazard is exist.

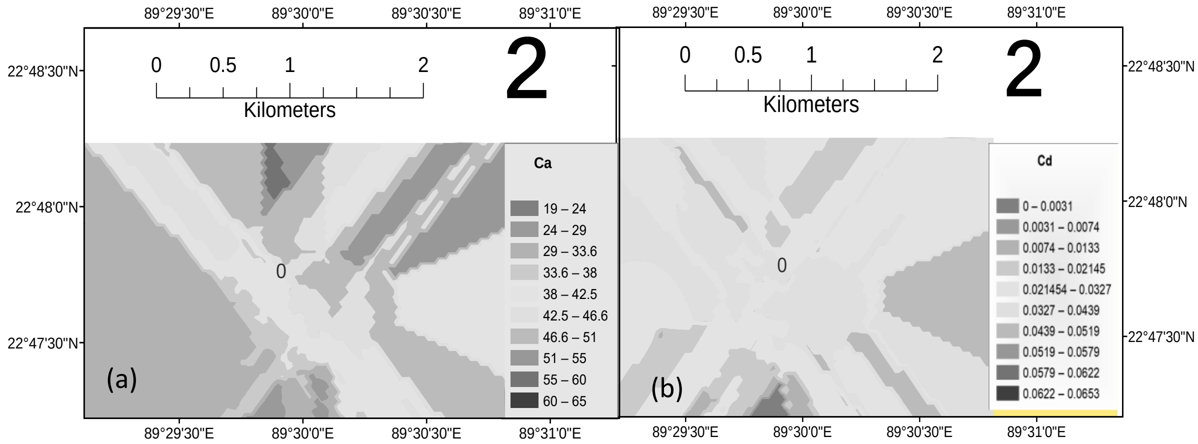

Figure 5: Spatial Distribution of Heavy Metal concentrations in Groundwater

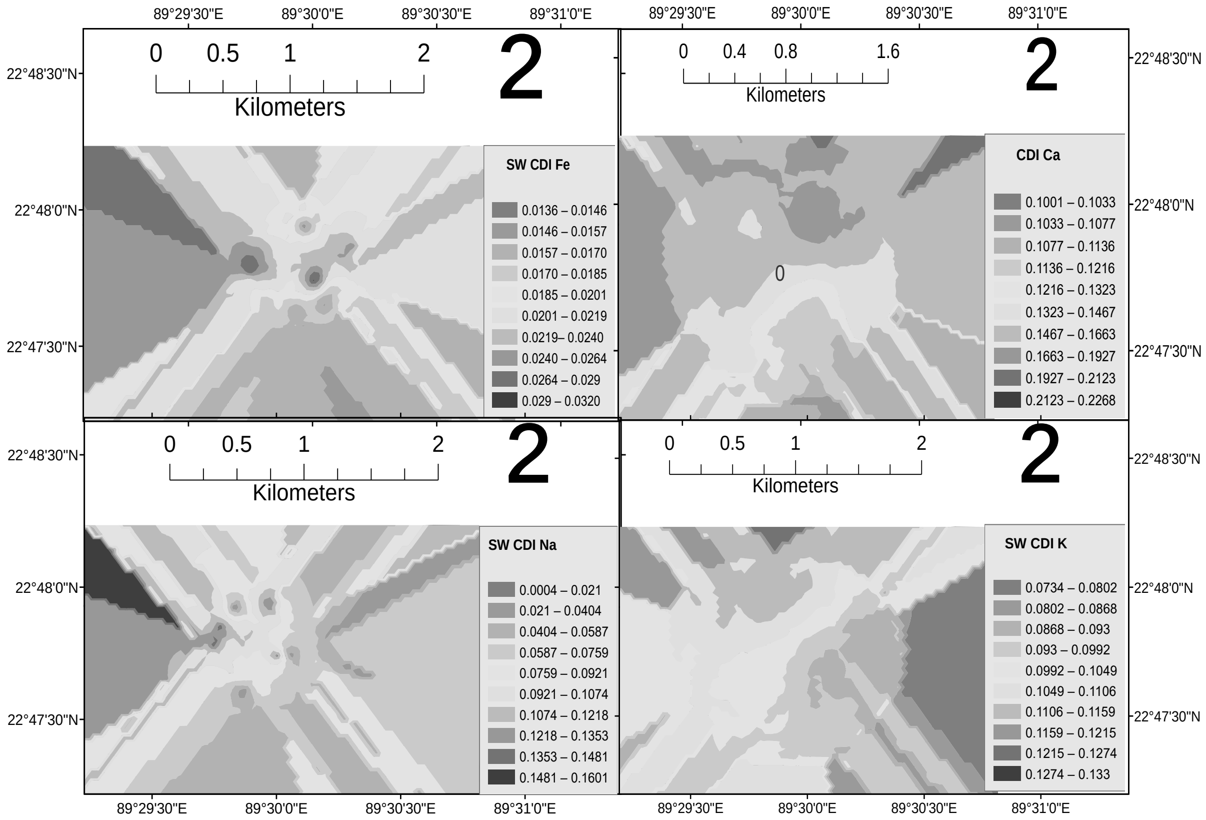

Figure 6:Spatial Distribution of CDI values of surface water

3.7 Uncertainty Analysis (1-D Monte Carlo Simulation)

The term uncertainty is interpreted as a lack ofknowledge about factors affectingexposure or risk models(Iman and Conover 1982;Kilic and Aral 2008).These uncertaintiescan be linked to the parameters used inexposure model (e.g. errors orinaccuracies in the measurement), to risk models input parameters,population characterization) (Lee et al.,2004; US.EPA, 2005; Wang et al., 2007;Chowdhury et al., 2009).The analysis ofuncertainty of exposure parameters and risk outputs (HQ, Hi) were performed using 1-D MCS @RISK 7.5 with 10000 iterations.

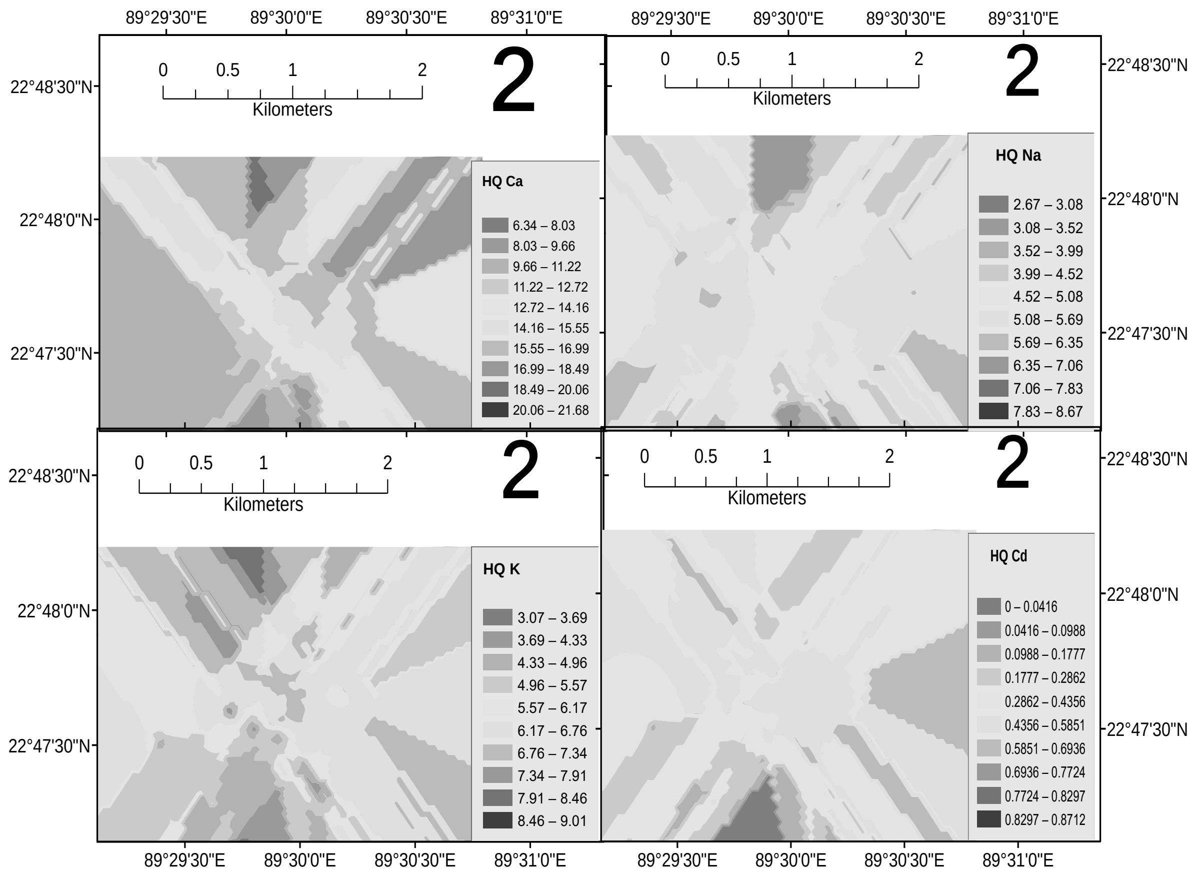

Figure7: Spatial distribution of HQ values for Groundwater

Graphical representation of input parameters (BW) of exposure model is shown in Figure 8. To select and fit a probability distribution, the body weight of the exposure populations was compiled using @RISK 7.5. In the Figure 8, the height of the bars (the yaxis) represents the relative frequency ofbody weight in the population and thespread of the bars (the x-axis) is thevarying amounts body weight (kg).Sincebody weight is a continuous randomvariable, the probability distribution canalso be represented graphically with aprobability density function (PDF) (Figure-8a) as well as CDF (Figure-8b).Theminimum, maximum, mean, standarddeviation and numberof iterations are alsopresented in the box.There is a greater area under the curve (greater probability density) in the 60-80kg range than 0-60kg or 80-90 kg. By selecting a normal distribution to characterize inter-individual variability, we can state more precisely that 60 kg corresponds to the 5th percentile and 80 kg corresponds to the 95th percentile, so approximately 90% (i.e., 0.95–0.05=0.90) of the BW is likely to exist between 60 and 80 kg with a mean value of 70 kg. Theprobabilistic calculation of riskinvolves random sampling fromeach ofexposure variable distributions.

The output of the exposure assessment process is a distribution of risk estimates. When the calculation of risk (or any other model endpoint) is repeated many times using MCS to sample the variables at random, the resulting distribution of risk estimates can be displayed in a similar fashion. In addition, the normal distribution of HI for the metal of Ca in SW1 for Child is shown in Figure 9. The total normal distribution is represented by both the PDF and CDF which represented the same distribution including summary of statistics, but are useful for conveying different information.

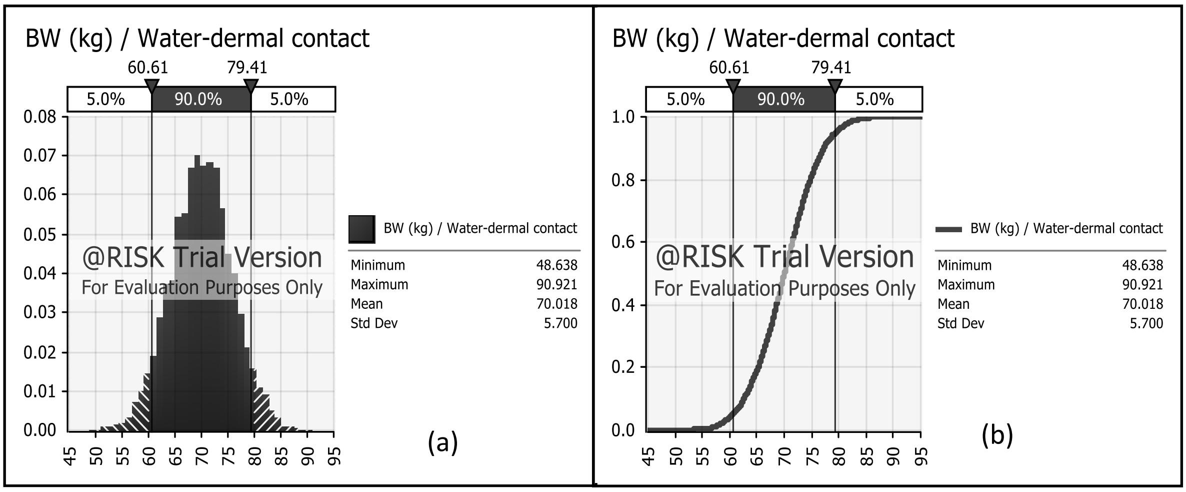

Figure 8: Normal distribution of BW for water dermal contact as (a) Bell-shaped curve represents the PDF and (b) S-shapedcurve represents the CDF

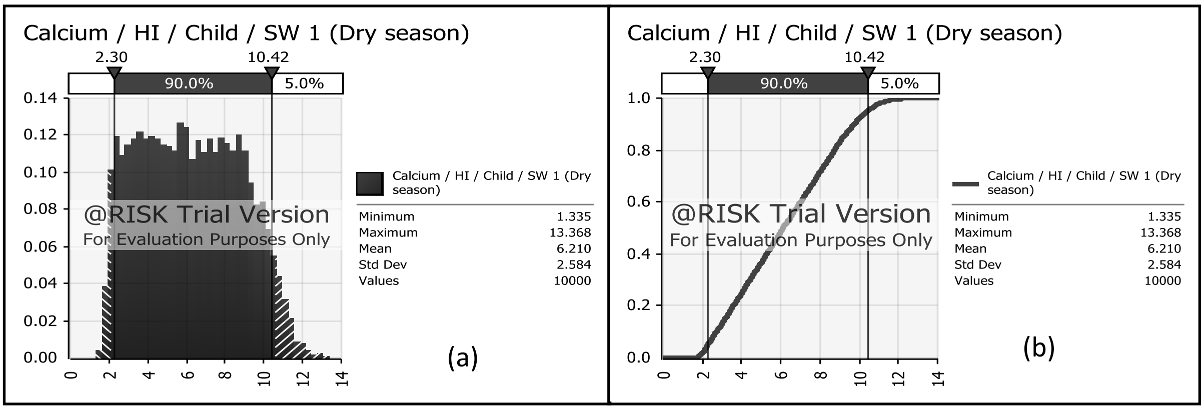

Figure 9: Normal distribution of HI of Calcium inSW1 for Child as (a) Bell-shaped curve represents the PDF and (b) S-shaped curverepresents the CDF.

4. CONCLUSIONS

Result reveals that the values of CDI, HQ and HI for metal in surface and ground water for rainy season was found comparatively higher than dry reason.In addition, HQ for ingestion route was comparatively higher than that of dermal for almost all metals. However, Ca showed the highest values of HI with 2.45E+01, exceed the acceptable non-carcinogen limit of 1. On the basis of HI, metals hazardous sequel should be like as Ca> k> Na> Cd> Cr> Pb> Mn> Cu> As> Fe> Ni> Zn in groundwater. Furthermore, results reveal that the values of HQ in case of ingestion route for surface water were found to be higher than that of dermal condition. In addition, RME showed the higher values of HQ in compare to CTE. Ca possess more hazards in RME condition during rainy season as it has maximum HQ valuefor surface water. Results of Pearson's correlation and PCA indicated that Fe, Mn, Cu and Ca were generated from anthropogenic sources, while, Cd, Cr, Ni, Pb, K and As from natural sources.

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