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Human Health Risk Evaluation of Dissolved Metals in Water Resources in and Around MSW Disposal Site in Kolkata

Vandana Parth*, Somnath Mukherjee
Environmental Engineering Division, Department of Civil Engineering, Jadavpur University, Kolkata, India
Source(s) of support: University Grants Commission, New Delhi, India.
Email id: Vandana.parth@gmail.com

Abstract: ***- Geochemical study was undertaken to assess the extent and health impact of toxic heavy metals intrusion to surface and sub-surface water resources in and around uncontrolled solid waste dumpsite of a metropolitan city in India. Water samples were collected from the vicinity of the site with different metal concentration scenario. The concentration (μgL^{-1}) of toxic metals/metalloids viz Cr 27.5±15.4, Mn 257.2±330.9, Co 0.5±0.5, Ni 4.0±2.3, As 18.4±12.6, Cu 5.3±5.0, Zn 292.7±466.7, Cd 0.2±0.5, Pb 1.0±0.4, V 36.8±16.8) were observed in above samples through estimation in dynamic reaction cell inductively coupled plasma mass spectrometry (DRC-ICP-MS). Human health was evaluated for potential non-carcinogenic hazard quotient (HQ) and carcinogenic risk (CR) for the nearby habitants considering all possible exposure routes. The cumulative hazard index (dermal and ingestion) for child (Σ HI = 4.26E+00) and adult (2.59E+00) reveals that toxic risk is beyond tolerable limit (HQ > 1.00). Arsenic was identified as the most significant pollutant of concern among the ten heavy metals; both its values of the HQ and CR indicated potentially undesirable health risks for the local residents. The estimated risk level for As exposure (3.56E-04) is exceeding the safe standard for cancer (CR=1.0E-6) unveil that residents confront higher risks, with carcinogenic effects that average 4 in 10,000. However, the cancer risk due to Pb exposure (1.11E-07) is within the target level.***

Keywords: *Solid waste dumpsite; Water samples; Heavy metals; Health risk assessment; Hazard quotient; Cancer risk.*

Introduction

Intimidation to the water resources from the unlined and uncontrolled solid waste dumpsite exist in many parts of the world. In India urbanisation and industrialisation is proceeding at an unprecedented rate. Such development is often unbalanced with much of the disposable municipal expenditure in addition to infrastructure with waste disposal and waste management coming well down the list of priorities in terms of allocation of funding. Consequently, open dumping of waste/landfill has become a preferred method of disposal for municipal solid waste (MSW) due to its complimentary economics. However, poorly designed landfills or an open waste dumpsite can result in contamination of valuable groundwater resources. In Kolkata the prevailing method for the disposal of municipal and domestic refuse is usually open dumping, often coupled with waste burning and with minimal effort directed towards sanitary land filling (use of daily cover) practice. Further, the site selection is normally done on the basis of geographical and lands availability rather than geo and hydro geological considerations, i.e. the closer the site to the source of the waste the better in terms of logistics; it is not uncommon therefore to find waste disposal sites within municipal boundaries surrounded by residential areas and agricultural lands. Consequently, such sites pose a serious health risk associated with proximity to litter, feral animals, scavenging birds, vermin and airborne contamination from mobilisation of aerosols. The most commonly reported danger to the human health from such dumpsite is the use of groundwater that has been contaminated by leachate.[3,12,10] A number of incidences have been reported in the past, where leachate had contaminated the surrounding soil and polluted the underlying groundwater aquifer or nearby surface water.[29,30,3,9,12,14] Strength of leachate from MSW dumpsite varies with the progress of biological activity stirring in waste. The rate and characteristics of leachate produced depends on factors such as solid waste composition, particle size, degree of compaction, hydrology of site, age of landfill, moisture and temperature conditions, available oxygen etc. During the course of stabilization of wastes, non-conservative constituents of leachate (primarily organic in nature) tend to decompose and stabilize with time, whereas conservative constituents (mainly heavy metals) remain long even after stabilization of waste. Metals often are precipitated within the waste and are infrequently found at high concentrations in leachate. These heavy metals have a tendency to reach surface and groundwater in appreciable levels resulting adverse effect to human health.[19,4,5,22] One of the most important environmental issues today is the level of groundwater contamination with heavy metals and metalloids, because of their acute toxicity even at low concentrations.[13] In order to quantitatively assess the potential risk associated with metal toxicity, human health risk assessment has become a largely applied methodology to evaluate the likely risks from exposure to environmental pollutants, not only contained in water,[6] but also in other environmental media, such as soil or air.[8,16,21,32] An understanding of leachate composition and an integrated strategy for risk assessment are very much crucial and necessary to correctly face the problem and for making projections on the long-term impact of a dumpsites, with particular attention on the estimation of possible adverse effects on human health.[2] In the case of Kolkata MSW dumpsite (Dhapa), the risk assessment on water resources has not been conducted previously. As a consequence, a risk evaluation study was undertaken for metals and metalloids in water resources in the vicinity of dumpsite which is a part of wetland ecosystem and most likely is contaminated by leachate. The risk of cancer is estimated quantitatively, whereas the non cancer risk is determined taking into account uncertainty by using safety factors. This separation derives from the assumption that cancer risks can be estimated using models that are linear through zero dose, whereas non cancer effects are subject to a threshold.[7] In terms of non cancer risks, “if the overall Hazard quotient (HQ) or Hazard index (HI) value is less than one, public health risk is considered to be very low.” However, “if the HI value is equal to or greater than one, then the exposure assessment and hazard characterization should be investigated more thoroughly” as discussed by Rodriguez and Grant.[20] Risk estimates for carcinogens are expressed as the incremental probability of developing cancer (e.g., an additional one in a million chance of developing cancer) over a lifetime of exposure to potential carcinogens. The United States Environmental Protection Agency (USEPA) has identified a risk level range of 1×10^{-6} to 1×10^{-5} as an appropriate risk management goal for the general population, as long as the most sensitive population is protected at 1×10^{-4}.[15] The manifestation of carcinogenic effects in contaminated areas may not be clearly demonstrated, since it normally takes decades of exposure duration to develop cancer. [17]

The goals of the present study were (1) to investigate the levels of heavy metals (Cr, Mn, Co, Ni, As, Cu, Zn, Cd, Pb, and V) in water in the surrounding areas of Kolkata dumpsite; and (2) to evaluate the health risks associated with these metals by examining the primary human exposure pathways.

Materials and methods

Study area

The survey area chosen for present study is located in the eastern fringes of Kolkata metropolis, falling in Survey of India Toposheet No. 79B/6. The area around Dhapa (geographical coordinates 88°24′N:22°32′E) has been covered since long time with municipal dump with numerous low altitude highs and consequent

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depression filled with water bodies. The highs are been used as agricultural lands whereas, the water bodies are used as fisheries which are vulnerable to contamination by leachate intrusion. Dhapa is a part of the wetlands (approximate area of 10,000 ha), out of which 24.71 ha are used for dumping solid waste. The city forms a part of the lower deltaic alluvial plains of the Ganga-Bhagirathi river system. It is a typical deltaic flat land with surface elevation ranging between 3.5 to 6m above mean sea level (MSL). Kolkata has a Tropical wet-and-dry climate. The annual mean temperature is 24.8 °C (80 °F); monthly mean temperatures range from 15 °C to 30 °C (59 °F to 86 °F). The mean annual precipitation is 1647mm. The location of dumpsite and sampling points has been shown in Figure 1.

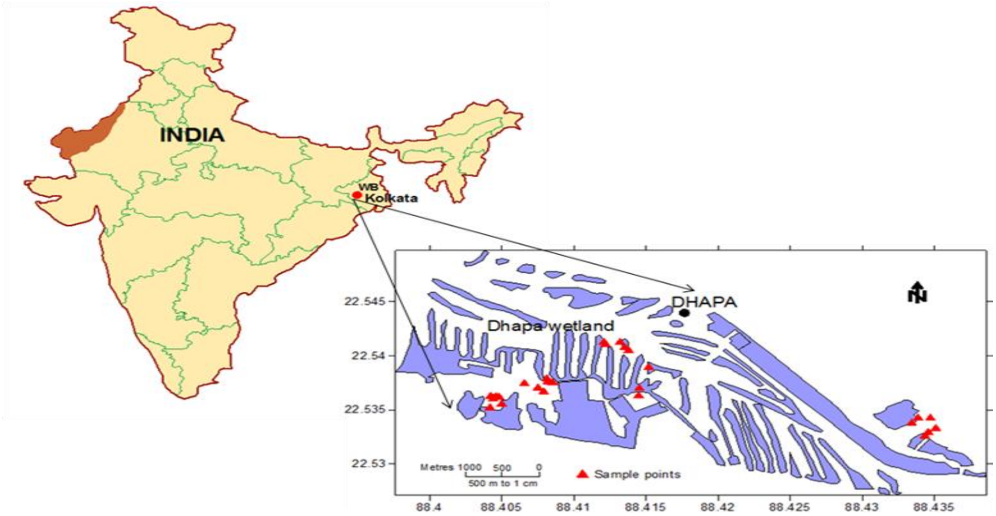

Figure 1 Site location and distribution of sampling points

Sampling and analytical methods

A total number of twenty six water samples (*n* 26) were collected from tube-wells (from depth span of 30-60 m; diameter 200-250 mm), dug-wells, ponds, running water and lakes located in the surrounding areas of dumpsite at the target interval of 200m to 400m as shown in (Figure 1). The samples were collected during April and May (summer) when the water levels are low and the mineral contents in water are likely to reach the maximum. Samples were preserved in pre-cleaned high-density polyethylene bottles from representative locations distributed throughout the area. The collected samples were filtered using Whatmann filter paper no. 42, and acidified with nitric acid (AR grade) to pH < 2 (0.2% v/v). On-site observations like location, source and depth of the tube-wells were recorded. Parameters such as pH, total dissolved solids and temperature were measured instantly by corresponding WTW test kit. Metal content in the samples were analysed by ICPMS model ELAN DRC II, Perkin-Elmer Sciex instrument. Calibration of the instrument was performed using certified reference material NIST 1640 (National Institute of Standards and Technology, USA), to minimize the matrix and other associated interference effects. All the samples were analyzed in three replicates. The quality control of analytical accuracy was also carried out by reagent blanks. The precision obtained in most cases was better than 5% RSD with comparable accuracy.[1] Blanks were analyzed along with the samples and rectifications were carried out accordingly.

The risk assessment

Risk assessment is a function of the hazard and exposure. It is defined as the processes of estimating the probability of occurrence of any given probable magnitude of adverse health effects over a definite time period which involves a multi-level process such as; (a) data compilation and scrutiny, (b) exposure review (evaluation of the extent of potential or actual contacts), (c) toxicity assessment (detrimental human health

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consequences due to chronic exposure to various substances are determined) and (d) the risk classification (sums up the results of (b) and (c) above) defined by US EPA.[27] The health risk assessment of each potentially toxic metal is usually based on the quantification of the risk level and is expressed in terms of a carcinogenic or a noncarcinogenic health risk. The two principal toxicity risk factors evaluated are the slope factor (SF) for carcinogen risk characterization and the reference dose (RfD) for noncarcinogenic risk characterization.[11] The SF is a conservative estimate of the incremental probability of an individual developing cancer as a result of exposure over a lifetime, and RfD is the estimated amount of the daily exposure level for the population that is likely to be without an appreciable risk of deleterious effects during a lifetime. The toxicity indices of each potentially toxic metal are shown in Table 1.

Table 1. Toxicity indices of elements

Chemical	Cas. #	Chronic RfD mg/kg-d	Chronic RfC mg/m^3	Ingestion SF $(mg/kg\text{-}d)^{-1}$	Inhalation Unit Risk $(ug/m^3)^{-1}$	EPA Cancer Classification
Arsenic	7440-38-2	3.00E-04	1.50E-05	1.50E+00	4.30E-03	Group A
Cadmium	7440-43-9	5.00E-04	1.00E-05	Nd	1.80E-03	Group B1
Chromium, total	Nd	nd	Nd	Nd	nd	Group A
Cobalt	Nd	3.00E-04	6.00E-06	Nd	9.00E-03	
Copper	7440-50-8	4.00E-02	Nd	Nd	nd	Group D
Lead	7439-92-1	nd	Nd	8.50E-03	1.20E-05	
Manganese	7439-96-5	2.40E-02	5.00E-05	Nd	nd	
Nickel	Nd	2.00E-02	9.00E-05	Nd	2.60E-04	
Vanadium	Nd	5.04E-03	Nd	Nd	nd	
Zinc	7440-66-6	3.00E-01	Nd	Nd	nd	Group D

nd = index not determined, Cas. # = chemical abstract service registry number.

The estimations of the magnitude, frequency and duration of human exposure to each potentially toxic metal in the environment are typically reported as chronic daily intake (CDI),[24] as shown in equations 1 and 2. The input parameters in CDI are summarised in Table 2.

Table 2 Input parameters to compute CDI

Parameters	Description	Unit	Value
C	Contamination concentration in media	mg/L	
IR	Ingestion rate per unit time		
	Adult	L/day	2.5
	Child	L/day	0.78
EF	Exposure frequency, adult/child	days/years	350
ED	Exposure duration		
	Adult	years	26
	Child	years	6
BW	Body weight		
	Adult	kg	80
	Child	kg	15
AT	Average time	days	30/70x365
SA	Skin surface area		
	Adult	cm^2	20900
	Child	cm^2	6378
AF	Adherence factor	$L/m^2/day$	272.167
ABS	Absorption factor		0.01
CF	Conversion factor		E-06

The toxicity indices equations are

For ingestion

$$CDI = \frac{CW \times CF \times IR \times EF \times ED}{BW \times AT} \quad (1)$$

For dermal contact

$$CDI = \frac{CW \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (2)$$

where CDI is the exposure duration (mg/kg-day), CW is the concentration (mg/l), CF is the conversion factor, IR is the intake rate (l/day), EF is the exposure frequency (day/year), ED is the exposure duration (year), SA is the surface area (m^2), AF is the adherence factor (L/m^2/day), ABS is the absorption factor, BW is the body weight (kg), and AT is the average time (day).

Risk characterization is the final stage of risk assessment. The aim of this stage is to calculate the risk. The results of toxicity and exposure assessment are integrated to arrive at quantitative estimates of cancer risk and hazard indices. Carcinogenic risk is the probability of an individual developing any type of cancer from lifetime exposure to carcinogenic hazards. The acceptable or tolerable risk for regulatory purposes is in the range of 10^{-6} to 10^{-4}. These values mean from one additional case in a population of 1 million to one in 10,000 people is acceptable. Carcinogenic risk is estimated with a linear equation (3) given below

Cancer Risk = CDI (mg/kg/day) ×Slope Factor (mg/kg/day)$^{-1}$ (3)

The potential for non carcinogenic agents is assessed by comparing exposure or average intake of hazardous substances with corresponding RfD as shown in equation 4. Non cancer risk is represented in terms of hazard quotient (HQ) for a single substance or hazard index (HI) for multiple substances and/or exposure pathways as shown in equation (4) and (5) respectively. If the exposure level of a substance exceeds the corresponding RfD i.e., HQ exceeds 1, there may be concern for potential non carcinogenic effects.

$$HQ = CDI/RfD \quad (4)$$

$$HI = \Sigma\, HQ \quad (5)$$

Toxicological values (RfD and SF) for the selected pollutants are obtained from USEPA IRIS,[25,28] database. This database consists of the main toxicological parameters derived from epidemiological and clinical studies for different compounds. In the case of missing human toxicological values, statistics were derived from experimental or predicted values (EC 50, NOEL and LC 50) from animal in vivo studies (rat or mouse) and using dedicated and appropriate safety factors: 10.000 in case of acute toxicity data, 1.000 for NOAEL or sub-acute toxicity data or 100 for chronic data.

Estimates of uncertainties

Various sources of uncertainty occur in risk assessment. Uncertainty is inherent in the process even when using the most accurate data and the most sophisticated models. Uncertainties encountered are originally due to fate and transport of pollutants, in a variety of different and variable environments, by processes that are often poorly understood or too complex to quantify accurately and extrapolation of slope factor of cancer risk from in vivo studies to human and from elevated dose to low dose.[23] As a result a particular compound of interest (COI) may not cause cancer at all though calculated risk is positive (>0). The uncertainty related to exposure assessment also contributes to the uncertainty of the risk estimate. Risk analyses in the water resources around dumpsite indicate that the exposure parameters strongly influence the results of the assessment. Moreover, the risk assessment is based on the route of drinking water ingestion and dermal exposure. However, other possible exposure pathways (e.g., the food chain) also need to be considered. With this information, environmental management system would be capable to deal with these uncertainties in making judgment.

Results and discussion

Hydrogeochemistry

The concentrations of metal in surface and groundwater were determined and their corresponding risk values were calculated. The statistical descriptive parameters were computed using SPSS 20.0 software package. Using correlation analysis, the associations between the metal concentrations were then calculated. COI were identified as the following total trace metals: Cr 27.51±15.44 μgL^{-1}, Mn 257.25±330.93 μgL^{-1}, Co 0.49±0.53 μgL^{-1}, Ni 4.00±2.27 μgL^{-1}, As 18.39±12.65 μgL^{-1}, Cu 5.32±5.01 μgL^{-1}, Zn 292.72±466.70 μgL^{-1}, Cd 0.25±0.52 μgL^{-1}, Pb 1.02±0.40 μgL^{-1}, V 36.79±16.76 μgL^{-1}. Maximum concentrations as detected from various samples representing the untreated water were screened against the most conservative Oregon Risk Based Concentrations (RBCs) for groundwater ingestion.[18] If an RBC was not available for a specific chemical, the EPA Regional Screening Levels,[26] were used. RSLs were used for zinc in the present study. All COI were considered as chemicals of potential concern (COPC). The maximum concentrations of Cr and Mn are higher than the permissible limits as prescribed by WHO,[31] but their average concentrations are within the limit. The average concentration of As on the other hand exceeds the acceptable limit. Other metals have lower concentrations than WHO threshold limits. A summary of statistical analysis of the COPC is shown in Table 3. Correlation coefficient of COPC is also depicted in Table 4. A reasonable correlation exists between Cr-Mn (r=0.559), Cr-Ni (r=0.430), Mn-Ni (r=0.625), Cu-Ni (r=0.419), Mn-Co (r=0.787), Cr-Co (r=0.574), Co-Ni (r=0.759), Co-As (r=0.594), Ni-As (r=0.586) and As-V (r=0.616) indicating a common source of origin for these metals. The maximum concentrations of Cr, Mn and As were observed higher than health based guidelines for ingestion of water set by WHO (Table 3).

Table 3 Statistical values of metal (μgL^{-1}) in water around Dhapa landfill site

	Cr	Mn	Co	Ni	As	Cu	Zn	Cd	Pb	V
Max	65.94	992.59	2.23	11.2	46	19.75	1446.61	2.59	2.1	91.78
Min	12.07	0.58	0.14	2.09	2.59	0.69	5.09	0.01	0.595	9.19
Avg	27.51	257.25	0.49	4.00	18.39	5.32	292.72	0.25	1.02	36.79
Std. Dev	15.44	330.93	0.53	2.27	12.65	5.01	466.70	0.52	0.40	16.76
Skew	1.55	1.31	2.26	2.02	0.93	1.57	1.54	4.03	1.47	1.20
Kurt	1.20	0.57	4.55	3.92	-0.15	1.94	0.97	17.87	2.48	3.66
Theshold value*	50	500	-	20	10	200	2000	3	10	-

***Maximum permissible concentrations as defined by WHO (2004)**

Table 4 Correlation coefficient between metals in water resources around Dhapa

	Cr	Mn	Co	Ni	As	Cu	Zn	Cd	Pb	V
Cr	1									
Mn	0.559	1								
Co	0.574	0.787	1							
Ni	0.430	0.625	0.759	1						
As	0.405	0.220	0.594	0.586	1					
Cu	0.374	-0.079	0.278	0.419	0.464	1				
Zn	-0.313	0.164	-0.089	-0.186	-0.336	-0.389	1			
Cd	-0.037	-0.172	-0.125	0.040	-0.126	-0.081	-0.181	1		
Pb	-0.197	-0.343	-0.071	0.038	0.055	0.391	-0.407	-0.093	1	
V	0.110	0.371	0.560	0.508	0.616	0.351	0.054	-0.179	0.105	1

Table 5 Non carcinogenic health hazard of Dhapa Landfill site for water

Chemical	Child HQ ingestion	Child HQ dermal	Child ΣHQ	Adult HQ ingestion	Adult HQ dermal	Adult ΣHQ
Arsenic	3.06E+00	1.35E-02	3.07E+00	1.84E+00	1.09E-02	1.85E+00
Cadmium	2.49E-02	2.19E-03	2.70E-02	1.49E-02	1.77E-03	1.67E-02
Chromium, total	-	-	-	-	-	-
Cobalt	8.21E-02	1.45E-04	8.22E-02	4.93E-02	1.17E-04	4.94E-02
Copper	6.63E-03	2.93E-05	6.66E-03	3.98E-03	2.37E-05	4.01E-03
Lead	-	-	-	-	-	-
Manganese	5.34E-01	5.90E-02	5.93E-01	3.21E-01	4.77E-02	3.69E-01
Nickel	9.97E-03	2.20E-04	1.02E-02	5.99E-03	1.78E-04	6.17E-03
Vanadium	3.64E-01	6.18E-02	4.26E-01	2.19E-01	4.99E-02	2.69E-01
Zinc	4.87E-02	1.29E-04	4.88E-02	2.92E-02	1.04E-04	2.93E-02
Total HI	**4.13E+00**	**1.37E-01**	**4.26E+00**	**2.48E+00**	**1.11E-01**	**2.59E+00**

Human health risk assessment

Non carcinogenic

The concentrations of Co, Ni, Cu, Zn, Cd, Pb and V in water have been found not exceeding the drinking water standards and hence, the water quality is acceptable (Table 3). Accordingly, the human health risk assessment of these metals showed respective HQ values suggesting an acceptable level of non carcinogenic adverse health risk. This is found to be consistent with the absence of any reports of a significant non carcinogenic risk from these heavy metals by oral or dermal exposure (Table 5). Nevertheless, in contrast, the HQ values of Cr and Mn indicate an acceptable non carcinogenic health risk despite showing their maximum concentrations (Table 3) beyond acceptable levels. However, objectionable HQ was identified for arsenic exposure (ΣHQ_{child} = 3.07E+00, ΣHQ_{adult} = 1.85E+00). The measured cumulative hazard index for child (Σ HI = 4.26E+00) and adult (2.59E+00) reveal that toxic risk is beyond target level (HQ > 1.00) as described in Figure 2.

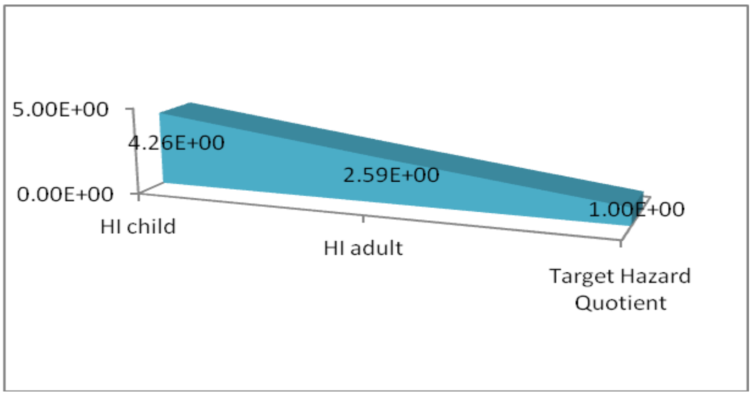

Figure 2 Cumulative HQ for child and adult residents in Dhapa area

Carcinogenic

Individual cancer risks were evaluated for arsenic and lead in the surface and groundwater as shown in Table 6. According to Oregon Administrative Rule (OAR) 340-122-115(2) (a), DEQ[18] the acceptable risk level for excess lifetime cancer risk (ECR) associated with potential exposures to individual compounds is 10E-06 (one in one million). In the present study, the ECRs for lead (1.11E-07) in the resident are below 1.0E-06 or one in one million threshold indicating negative cancer risks due to lead exposure. A carcinogenic

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risk was encountered only for arsenic in water as shown in Figure 3.

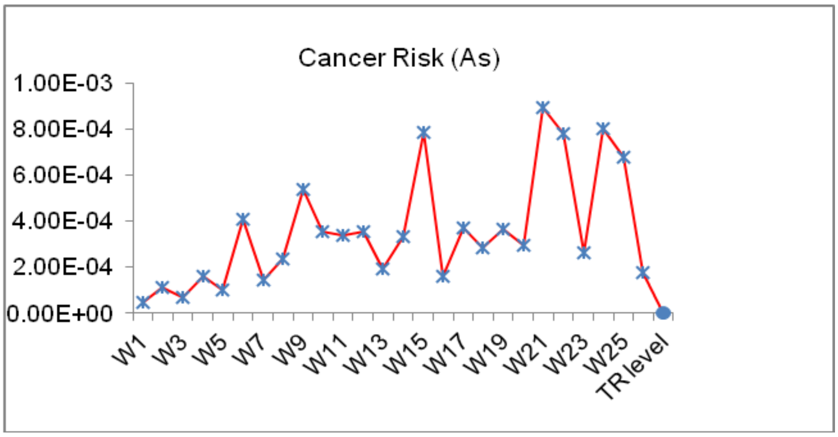

Figure 3 Carcinogenic risk of arsenic due to oral exposure in Dhapa

Where x axis represents the water samples and the y axis signifies the respective carcinogenic risk level. Ingestion-based carcinogenic risk was higher than the critical level (10^{-6}), but the dermal-based risk was low (Table 6). The estimated risk level for arsenic exposure (3.56E-04) in the present case investigation unveil that residents confront higher risks, with carcinogenic effects that average 4 in 10,000.

Table 6 Carcinogenic health risk of Dhapa Landfill site for water

Chemical	Ingestion risk	Dermal risk	Total Cancer risk
Arsenic	3.54E-04	1.97E-06	3.56E-04
Cadmium	Nd	Nd	Nd
Chromium, total	Nd	Nd	Nd
Cobalt	Nd	Nd	Nd
Copper	Nd	Nd	Nd
Lead	1.11E-07	6.19E-11	1.11E-07
Manganese	nd	Nd	Nd
Nickel	nd	Nd	Nd
Vanadium	nd	Nd	Nd
Zinc	nd	Nd	Nd

nd = not detected

Conclusions

The prevalence of metals in surface and subsurface water around Kolkata MSW dumping site and their carcinogenic and non carcinogenic risk levels to human health following dermal and ingestion exposure were determined for adults and children. The area around dumpsite is covered with several low altitude highs and consequent depression filled with water bodies. The highs are been used as farm lands whereas, the water bodies are used as fisheries which are most viable to be contaminated by the leachate. The concentrations of Co, Ni, Cu, Zn, Cd, Pb, and V in the surface and subsurface water were found within the permissible limits. However, the average concentration of arsenic was found exceeding the allowable drinking water standard. In addition, the maximum concentrations of Cr and Mn are also exceeding the prescribed standard, but do not show any health hazard. Arsenic was identified as the most significant pollutant of concern among ten heavy metals; both its values of HQ and CR indicated potentially undesirable health risks for the local residents. The outcome of this study can be applied to decision making action and to communicate about the risk to local people who use shallow groundwater (depth 10-15m) for drinking purpose. In order to reduce the estimated carcinogenic risk and non carcinogenic HIs, the residents are advised to treat their water or find alternative sources for drinking. Local authorities should be made aware of such health risks to provide potable water facilities. Broadly, this study illustrates the greater requirement for risk awareness and communication about heavy metal contamination of shallow groundwater, especially in the surrounding areas of MSW dumpsites.

Acknowledgements

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