

# Trace elements in groundwater near an abandoned mine tailings dam and health risk assessment (NE Zimbabwe)

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Groundwater from shallow hand-dug wells at an abandoned gold mine tailings dam was characterised for selected physicochemical parameters during dry and wet seasons of 2018 and 2019. Health risk exposure of the local population (adults and children) through ingestion and dermal exposure was assessed. Groundwater quality parameters were lower than international drinking water quality guidelines ( $p < 0.05$ ). The parameters were significantly influenced by season of the year (As, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>), nature (As, Cd, Cl<sup>-</sup>, Fe, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>), depth (Cd, Cl<sup>-</sup>, Fe, Ni, SO<sub>4</sub><sup>2-</sup>) and direction of the well (Cu, Cl<sup>-</sup>, Fe, NO<sub>3</sub><sup>-</sup>, Pb, SO<sub>4</sub><sup>2-</sup>) ( $p < 0.05$ ) relative to the tailings dam. Groundwater did not pose non-carcinogenic risk due to studied trace elements. However, arsenic had the potential to cause medium to high cancer risk to the local population. We propose re-vegetation of the tailings dam, diversion of surface tailings drainage to a containment pond and the provision of continuous piped water supplies.

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## INTRODUCTION

Groundwater is of excellent natural quality and conveniently available close to where it is required (Sujatha and Reddy, 2003). However, it is threatened by over-exploitation and pollution (Carrard et al., 2019). Groundwater quality is influenced by geogenic and anthropogenic processes (Javaid et al., 2020). It is naturally found in association with geologic minerals containing dissolved salts (Chacha et al., 2018). Human activities such as mining introduce contaminants into groundwater (Zhu et al., 2020).

Metalliferous mineral exploration and beneficiation leave behind large heaps of overburden material, waste rock and tailings rich in potentially toxic elements (PTEs) (Kiventerä et al., 2018). The nature of the PTEs in the waste streams depends on the mineralogy of the ore body and mineral recovery processes. Gold is naturally found mixed with varying proportions of other elements including Ag, As, Cu, Fe, Pb, S and Zn (Fomchenko and Muravyov, 2020), which eventually constitute mine tailings after extractive processes (Gitari et al., 2017). PTEs from mine tailings dams (MTDs) can be transported into the environment through runoff or by atmospheric dispersal. They can leach into groundwater as surface recharge. In the environment PTEs impact on the quality of air, water, soil and plants (Mohapatra and Kirpalani, 2017).

The establishment of human settlements without matching infrastructural development for water supply and wastewater treatment facilities, a seemingly common practice in small towns of low-income settings, leaves people exploiting groundwater from shallow hand-dug wells (SHDWs) for potable use, and using make-shift sanitation facilities. The quality of groundwater from SHDWs is influenced by season of the year (Ganiyu et al., 2018), depth to water level (Chuang et al., 2019) and distance (Yan et al., 2017) relative to the source of contamination. Groundwater near mine sites was reported to be contaminated with PTEs (Kim et al., 2017; Singh and Kamal, 2017; Akoto et al., 2019). Reported MTD failures (Kossoff et al., 2014; Armstrong et al., 2019) demonstrate the importance of carefully planned post-mining waste management, and the associated public health effects.

The local population that relies on groundwater from SHDWs may be exposed to PTEs through dermal contact and oral routes. Human health problems associated with exposure to As, Cd, Ni and Pb include damage to body organs and systems, and cancer (Wuana and Okieimen, 2011; Jaishankar et al., 2014). Therefore, an understanding of the variation of groundwater quality in developing settlements near abandoned mine sites without centralised water supply systems is critical to public health.

Studies on groundwater quality in Zimbabwe appear focused on peri-urban and urban informal settlements (e.g., Ndoziya et al., 2019; Zingoni et al., 2005). No local studies were done to assess the quality of groundwater near abandoned mine sites and the associated potential human health risk to PTEs. Further, there are no published cases of ill-health associated with exposure to PTEs retraced to mine wastes for the local population. This is against a background where groundwater for potable use is rarely treated and monitoring boreholes (usually > 40 m deep) placed at active mine sites are not used for monitoring groundwater quality after mine operations have ceased.

In the current work we report an investigation of the potential contamination of groundwater from SHDWs near an abandoned gold MTD and the associated human health risks to PTEs by dermal

and oral exposure. The selected groundwater quality parameters assessed are those normally used in similar studies, naturally associated with gold ores and tailings, or have significant health effects. It was assumed that (i) groundwater from SHDWs had higher concentrations of PTEs than international drinking water quality guidelines, and (ii) the local population living near the MTD was at health risk due to PTEs in groundwater by oral and dermal exposure.

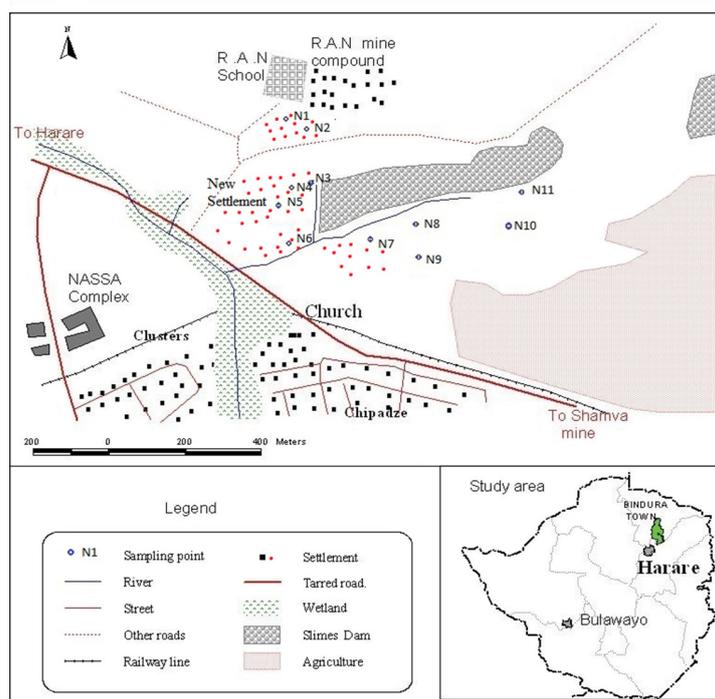
## METHODS AND MATERIALS

### Description of the study site

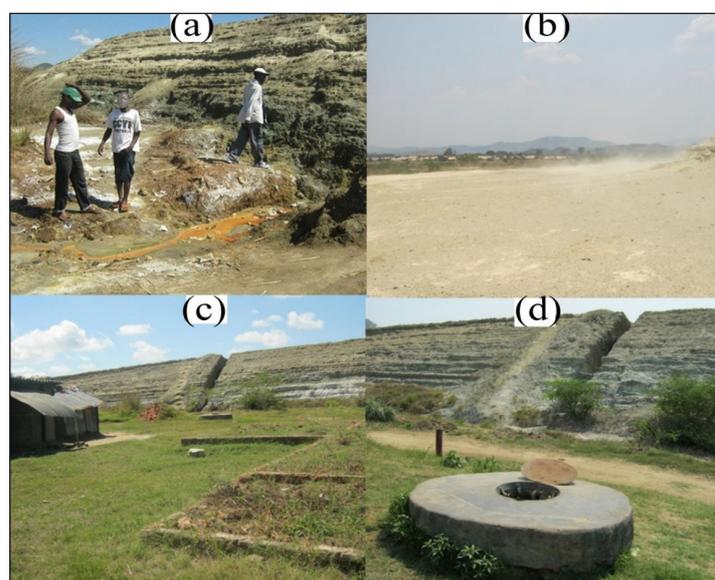
The study area was previously described (Kanda et al., 2019). Bindura town is found 60 km within the greenstone zone of the geology of Zimbabwe in the vicinity of the Great Dyke. The greenstone contains Fe, Au, Ni and Cu (Locmelis et al., 2010). The RAN ore channels consist of granodiorite stock that intrudes meta-sediments of the Mazowe formation (Shamvianian Group)

constituting mainly quartz and plagioclase feldspar with biotite and actinolite (Kalbskopf, 2002). Gold mining operations in Bindura began in the early 1900s (RAN mine, Freda Rebecca) and nickel mining in mid-1900 (Trojan Nickel Mine). RAN mine was commissioned in 1921 and operated for 77 years. It was decommissioned without rehabilitation. The MTD (17°18'05"S; 31°19'49"E) is located less than 30 m from newly established human settlements (Fig. 1).

The stability of the MTD is threatened by the development of gullies due to runoff exacerbated by illegal mineral exploration activities (Fig. 2d). Tailings are eroded from bare embankments by easterly winds in dry weather (Fig. 2b) towards human settlements and a primary school. In wet weather, runoff transports eroded tailings (Fig. 2a) into the ambient environment. The toe of the MTD is not designed to receive tailings drainage and channel it away. Surface runoff from elsewhere combines with that originating from the MTD and is transported downslope to a wetland.



**Figure 1.** Map of the study area showing the tailings dam and sampling sites



**Figure 2.** Authors' photographs showing (a) surface runoff and (b) wind erosion during wet and dry seasons, and (c) developing human settlements using (d) shallow hand-dug wells at the abandoned RAN mine tailings dam

## Sampling and sample analyses

Seven existing wells (either in use or not) within 75 m of the nearest toe of the MTD were purposively chosen for water sampling. Three new wells (N2, N7 and N9) were dug to depths until water appeared (3.4, 1.9 and 2.8 m, respectively), at chosen sites relative to the MTD and existing wells (Table 1). An existing reference well (REF) located 2 000 m from the MTD was included in the study.

Upon getting consent from house owners to take water samples for analysis (after explaining the purpose of the study), 2 groundwater samples (2 x 500 mL) were taken from each well once a month for 3 consecutive months from September to November 2018 (dry season) and repeated from January to March 2019 (wet season). Samples for total PTE analyses were preserved with drops of ultra-pure conc. 65% HNO<sub>3</sub> (pH < 2) to eliminate microbial activity (Hussain et al., 2019). Groundwater pH was determined on site for unacidified samples using a calibrated multi-parameter tester 35 (Eutech Instruments, USA). Other parameters were determined using standard methods for water analysis; SO<sub>4</sub><sup>2-</sup> (turbidimetry), NO<sub>3</sub><sup>-</sup> (Cd reduction) and Cl<sup>-</sup> (argentometry) (APHA, 2005). Groundwater samples were prepared and analysed for total recovery of PTEs following recommended procedures (USEPA, 2001). An acid-preserved groundwater sample (100 mL) was acid digested (HNO<sub>3</sub>/HCl; 2:1, 3 mL, v/v) over a hot plate (85°C) in a fumehood to 20 mL. Cooled digests were diluted to volume (50 mL) with reagent water and analysed for As, Cd, Cr, Cu, Ni and Pb by ICP OES (Spectro-Arcos FHS12).

## Health risk assessment

The health risk assessment was done for ingestion and dermal contact exposure to PTEs in groundwater (USEPA, 1989; USEPA, 2011) using Eqs 1–5. Parameters and input assumptions for exposure assessment to PTEs through ingestion and dermal pathways were obtained from literature (USEPA, 2011; Walpole et al., 2012; Al-Hwaiti et al., 2018; Haque et al., 2018; Mohammadi et al., 2019) (Table 2). The studied local group were adults and children. The average daily dose (ADD) of PTEs through ingestion and dermal contact was used to determine the human exposure dose to PTEs using Eqs 1 and 2 (see Table 1 for definition of variables in all equations):

$$\text{ADD}_{\text{ing}} = \frac{C_w \times I_r \times EF \times ED}{B_w \times AT} \quad (1)$$

$$\text{ADD}_{\text{dermal}} = \frac{C_w \times SA \times ABS \times EF \times AF \times ED \times CF}{B_w \times AT} \quad (2)$$

The Hazard Quotient (HQ) and the Hazard Index (HI) were used to determine the potential non-carcinogenic health risks due to ingestion and dermal contact for the studied PTEs in groundwater from SHDWs using Eqs 3 and 4:

$$\text{HQ} = \frac{\text{ADD}}{\text{RfD}} \quad (3)$$

$$\text{HI} = \sum_{n=1}^6 \text{HQ} \quad (4)$$

The Target Cancer Risk (TCR) was used to determine the carcinogenic risk due to arsenic. This is the possibility of a human being to develop carcinogenic effects in a lifetime exposure (USEPA, 1989). The ADD and the oral cancer slope factor (SF) were used to compute the TCR for PTE i:

$$\text{TCR} = \text{ADD} \times \text{SF} \quad (5)$$

## Statistical analysis

A one-sample Student's t-test was used to compare mean groundwater parameters against international drinking water quality guidelines (WHO, 2017). The effects of nature of well, direction, season (categorical variables), distance and depth (covariates) on the concentrations of PTEs were tested using generalized linear models (GLMs) with interaction terms for categorical variables. The proper distribution was determined by plotting observed deviance residuals against quantiles of the estimated distribution (Ben and Yohan, 2004). As a result, all GLMs were run with a Poisson error distribution (or a negative binomial distribution where residuals showed over-dispersion) combined with a log-link function. Type 1 log-likelihood ratios were used to analyse main effects. To counteract data over-dispersion and to adjust the statistics, the scale parameter was estimated by dividing the square root of the Pearson's Chi-square statistic by the degrees of freedom (McCullagh and Nelder, 1989). Pairwise differences within each factor were analysed using a post-hoc test based on least-square means. Analyses were performed using the library MASS (Venables and Ripley, 2002) and car (Fox and Weisberg, 2011) in R version 3.6.0 (R Core Team, 2019). A Spearman's correlation analysis was run to examine the relationship among water quality parameters.

**Table 1.** Characteristics of shallow hand-dug wells around RAN mine tailings dam, NE Zimbabwe

Well	Direction from the tailings dam	Shortest distance from the toe of the tailings dam (m)	Depth to water (m)	Nature of hand-dug well
N1	NW	73	3.6	Existing
N2	NW	55	3.4	New
N3	W	12	2.1	Existing
N4	W	24	2.3	Existing
N5	W	42	2.7	Existing
N6	SW	29	2.2	Existing
N7	S	20	1.9	New
N8	S	15	1.6	Existing
N9	S	50	2.8	New
N10	SE	33	3.1	Existing
REF	NE	2 000	3.2	Existing
Mean*		35.30±19.53	2.57±0.66	

\*excluding the reference well (REF)

**Table 2.** Health risk assessment model parameters and input data to Equations 1–5

Parameter	Unit	Ingestion	Dermal contact	Reference
Concentration of PTE (Cw)	mg·L <sup>-1</sup>	From study	From study	This study
Ingestion rate (I <sub>r</sub> )	L·d <sup>-1</sup>	2 adult, 1 child	-	USEPA (2011)
Exposed skin surface area (SA)	cm <sup>2</sup>	-	5 700 adult, 2 800 child	USEPA (2011)
Exposure frequency (EF)	d·yr <sup>-1</sup>	365	350	USEPA (2011)
Exposure duration (ED)	yr	30 adult, 6 child	30 adults, 6 child	USEPA (2011)
Adherence factor (AF)	mg·cm <sup>-2</sup>	-	0.07 adult, 0.2 child	USEPA (2011)
Body weight (Bw)	kg	60 adult, 15 child	60 adult, 15 child	Walpole et al. (2012)
Dermal absorption factor in water (ABS)		-	0.03 (As), 0.001 (each element)	USEPA (2011)
Averaging time for carcinogens (ATc)	D	25 550 (365 * 70)	25 550 (365 * 70)	USEPA (2011)
Averaging time for non-carcinogens (ATn)	D	10 950 adult, 2 190 child	10 500 adult, 2 100 child	USEPA (2011)
Reference dose for PTEi (RfDi)	mg·kg <sup>-1</sup> ·d <sup>-1</sup>	As: 0.0003, Cd: 0.0005, Cr: 0.003, Cu: 0.04, Pb: 0.0014, Ni: 0.02	As: 0.000285, Cd: 0.000005, Cr: 0.000075, Cu: 0.012; Ni: 0.0054, Pb: 0.00042.	Al-Hwaiti et al. (2018); Haque et al. (2018); Mohammadi et al. (2019)
Oral cancer slope factor for PTE (SFi)	kg·d <sup>-1</sup> ·mg <sup>-1</sup>	As: 1.5	-	Al-Hwaiti et al. (2018)

## RESULTS

### Variability of selected groundwater quality from shallow hand-dug wells

The variation of measured groundwater quality parameters from 11 wells and their comparison with international drinking water quality guidelines are shown in Table 3. The lowest groundwater pH was recorded for N3 during the dry season. Excluding the reference, 80% of the groundwater wells (N3–N10) had significantly higher concentrations of SO<sub>4</sub><sup>2-</sup> during the dry season ( $p < 0.05$ ). Similarly, the dry season had 40% of the wells having higher concentrations of Cl<sup>-</sup> than the wet season ( $p < 0.05$ ). All wells (including the reference) had lower concentrations of NO<sub>3</sub><sup>-</sup> in groundwater during both seasons than the international guideline of 50 mg·L<sup>-1</sup> ( $p < 0.05$ ). The concentrations of PTEs in groundwater were higher during the dry than wet season for arsenic (Wells N3–N5), Cd (N2, N4, N7), Cr (N1, N3, N4), Cu (N2, N6–N9), Fe (N4, N6, N7, N9), Ni (N3, N7) and Pb (N7) ( $p < 0.05$ ).

### Factors influencing the quality of groundwater from shallow hand-dug wells

The variations of overall groundwater quality parameters from SHDWs within a distance of 75 m from a MTD are presented in Tables 4, 5 and 6. Groundwater quality was influenced by the season of year (dry, wet) for As, Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup>; nature of well (new, existing) for As, Cd, Cl<sup>-</sup>, Fe, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>; cardinal points direction for Cl<sup>-</sup>, Cu, Fe, NO<sub>3</sub><sup>-</sup>, Pb and SO<sub>4</sub><sup>2-</sup>; depth for Cd, Cl<sup>-</sup>, Cr, Fe, Ni and SO<sub>4</sub><sup>2-</sup>; and distance from the MTD for Cl<sup>-</sup>, Cr, Cu, Fe, Ni and SO<sub>4</sub><sup>2-</sup> ( $p < 0.05$ ). The interactions of factors that influenced groundwater quality were largely evident on the concentrations of SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup>, and to some extent on NO<sub>3</sub><sup>-</sup>, Cu, Fe, and Ni (Table 6).

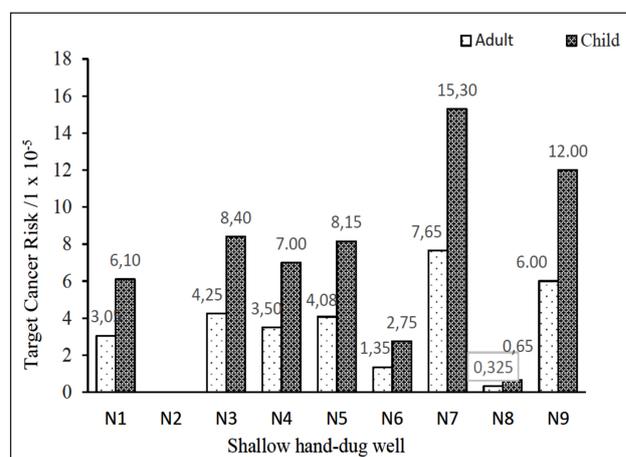
### Correlation analysis

The correlations between PTEs and other measured groundwater quality parameters are shown in Table 7. The concentration of SO<sub>4</sub><sup>2-</sup> was positively correlated with all studied PTEs, except

Pb, and negatively correlated with pH ( $p < 0.05$ ). All PTE–pH correlations were negative and significant ( $p < 0.05$ ), except for Pb. Ni and As had positive and significant correlations with other PTEs ( $p < 0.01$ ).

### Health risk assessment

The average daily dose (ADD) for children and adults in the study area shows their exposure to PTEs in groundwater from each well (Table 8). Well N10 and the reference had no values for the concentrations of PTEs. Overall, results show that children were more exposed to all the PTEs in groundwater than adults through both routes. Results show that the highest ADDs for each PTE from oral and dermal exposure to PTEs in groundwater at each well for children and adults occurred at N3 (Cr, Ni), N4 (Pb), N7 (As, Cd) and N9 (Cu). All Hazard Quotient (HQ) and Hazard Index (HI) values for both exposure routes were less than one ( $< 1$ ) and higher for children than adults. The target cancer risk (TCR) was determined only for arsenic (Fig. 3).



**Figure 3.** Target cancer risk due to oral exposure to arsenic in groundwater from shallow hand-dug wells

**Table 3.** Parameters of groundwater samples taken from individual wells around a gold mine tailings dam during dry and wet seasons. Values are means of 3 monthly measurements expressed as mean±standard deviation. Mean values (µg·L<sup>-1</sup>, unless specified) are compared to international guidelines for drinking water quality.

Parameter	Season	N1	N2	N3	N4	N5	N6	N7	N8	N9	N10	REF	WHO'
As	Dry	1.22±0.15	ND	1.04±0.13*	1.28±0.07*	1.06±0.12*	0.42±0.62	1.87±0.19	0.07±0.01	1.55±0.25	ND	ND	
	Wet	ND	ND	0.64±0.10*	0.12±0.05*	0.57±0.11*	0.12±0.06	1.19±0.37	0.06±0.02	0.85±0.18	ND	ND	10
Cd	Dry	ND	0.07±0.01*	0.33±0.06	0.66±0.08*	0.18±0.08*	ND	1.15±0.12*	0.05±0.01	0.10±0.05	ND	ND	
	Wet	ND	ND	0.14±0.06	0.25±0.09*	0.49±0.18*	0.06±0.02*	ND	0.53±0.01	0.14±0.02	ND	ND	3
Cr	Dry	0.05±0.01*	ND	3.29±0.46*	1.18±0.07*	0.42±0.57	ND	2.22±0.44	0.40±0.58	0.68±0.16	ND	ND	
	Wet	ND	ND	1.68±0.23*	0.85±0.12*	1.21±0.13	ND	1.28±0.18	0.09±0.01	0.33±0.08	ND	ND	50
Cu	Dry	0.09±0.02	0.18±0.05*	1.43±0.17	1.03±0.22	0.27±0.18	0.07±0.02*	1.09±0.21*	0.09±0.03*	2.31±0.18*	ND	ND	
	Wet	0.07±0.02	ND	1.06±0.07	0.89±0.19	0.09±0.01	ND	0.26±0.09*	ND	1.93±0.29*	ND	ND	2 000
Fe	Dry	0.43±0.11	0.85±0.08	4.68±0.67	2.25±0.12*	1.18±0.07	0.31±0.03*	7.49±0.60*	4.25±0.38	3.75±0.24*	0.99±0.24	0.65±0.12	
	Wet	0.28±0.02	0.64±0.10	3.79±0.33	1.28±0.07*	1.15±0.03	0.24±0.04*	4.64±0.43*	2.94±0.28	2.14±0.31*	0.99±0.22	0.60±0.11	None
Ni	Dry	ND	ND	3.82±0.63*	0.78±0.58	0.26±0.05	0.47±0.02	1.12±0.13*	0.06±0.01	0.66±0.11	ND	ND	
	Wet	ND	ND	1.51±0.24*	0.69±0.12	0.20±0.05	0.47±0.03	0.74±0.11*	0.03±0.01	0.41±0.13	ND	ND	70
Pb	Dry	ND	ND	0.35±0.06	1.08±0.18	0.35±0.10	ND	1.19±0.11*	0.39±0.55	0.82±0.66	ND	ND	
	Wet	ND	ND	0.54±0.13	0.85±0.08	0.55±0.07	0.07±0.01*	ND	ND	ND	ND	ND	10
Cl <sup>-</sup>	Dry	47.3±14.6	77.7±22.1	140.3±22.2*	130.7±15.2*	88±23*	45.3±5.1	109±13*	92.3±24	79±19.9	35±9.9	17±6.25	
	Wet	44.3±8.7	39.3±6.5	47±6.6*	41±3*	21.7±3.1*	34±2	12±3.6*	30±3	26.3±5.5	33.3±4	9.3±1.555	None
SO <sub>4</sub> <sup>2-</sup>	Dry	59.4±27.1	253.2±100	1 600±490*	1 298.3±315.5*	1189.3±303*	534.3±166.2*	875.2±242.7*	365.3±100.1*	915±327.5*	473±108.8*	261.3±72.5	
	Wet	12.8±2.8	10.4±2.0	122.7±4.2*	83.7±5.9*	62±4.58*	47±4*	143.7±8.6*	102±4.58*	72±3*	215±7.6*	134±8	None
NO <sub>3</sub> <sup>-</sup>	Dry	2.25±0.09	1.20±0.07	0.08±0.03	0.15±0.07	0.06±0.01*	1.87±0.44*	1.39±0.07*	1.23±0.06*	1.59±0.13*	1.36±0.09	0.43±0.12	
	Wet	ND	ND	0.09±0.02	0.12±0.06	0.14±0.03*	0.06±0.01*	2.77±0.15*	0.46±0.22*	3.06±0.15*	ND	ND	50
pH (+)	Dry	7.23±0.06	6.90±0.10	5.60±0.10*	6.50±0.10*	5.17±0.32	6.67±0.31	6.77±0.15*	6.73±0.15	5.83±0.71	6.63±0.21	7.07±0.15	
	Wet	7.00±0.10	7.00±0.10	4.43±0.15*	5.13±0.31*	6.23±0.15	6.73±0.31	5.47±0.23*	6.07±0.65	5.70±0.10	6.63±0.15	7.20±0.10	None

'WHO (2017) Guidelines for Drinking Water Quality; ND – not detected; None – no health-based guideline is proposed in drinking water; \*denotes significantly different parameter values for the dry and wet seasons within a row for a particular parameter for a well (p < 0.05)

**Table 4.** Effects of season, nature of well, direction and their interactions on groundwater quality parameters around an abandoned mine tailings dam. Values are expressed as mean±SD of replicates (µg·L<sup>-1</sup> unless otherwise specified).

Factor	Level	SO <sub>4</sub> <sup>2-</sup> (mg·L <sup>-1</sup> )	pH	Cl <sup>-</sup> (mg·L <sup>-1</sup> )	NO <sub>3</sub> <sup>-</sup>	As	Cd	Cu	Fe (g·L <sup>-1</sup> )	Cr	Ni	Pb
Season	Wet	91.38±10.27	6.56±0.09	30.76±2.22	0.96±0.28	0.51±0.09	0.19±0.04	0.70±0.17	1.70±0.25	0.91±0.14	0.52±0.11	0.51±0.09
	Dry	712.11±91.19	6.06±0.16	78.33±7.09	1.06±0.13	0.94±0.13	0.36±0.09	0.73±0.15	2.44±0.39	1.18±0.25	0.96±0.28	0.70±0.11
Nature of well	Existing	410.58±74.08	6.32±0.12	53.54±5.67	0.64±0.12	0.54±0.09	0.25±0.04	0.50±0.10	1.63±0.21	1.02±0.19	0.75±0.21	0.52±0.07
	New	378.21±96.62	6.28±0.16	57.22±8.66	2.00±0.21	1.14±0.16	0.37±0.14	1.15±0.23	3.25±0.58	1.13±0.22	0.73±0.08	1.01±0.19
Direction from tailings dam	S	481.88±98.67	6.03±0.13	65.04±8.80	1.35±0.22	0.87±0.14	0.34±0.09	1.09±0.17	3.59±0.38	0.88±0.14	0.56±0.08	0.87±0.11
	SW	745.75±213.81	5.36±0.20	74.25±14.14	0.10±0.01	0.83±0.07	0.28±0.09	0.69±0.18	2.70±0.48	1.65±0.33	1.45±0.45	0.45±0.04
	W	290.67±117.12	6.70±0.11	39.67±2.91	0.97±0.42	0.27±0.17	0.06±0.01	0.07±0.01	0.27±0.02	ND	0.05±0.01	0.07±0.01
	NW	83.95±32.64	7.03±0.04	52.17±5.74	1.73±0.24	0.24±0.06	0.07±0.01	0.11±0.02	0.55±0.07	0.05±0.01	ND	ND
Season x Direction	Wet x S	100.33±8.33	6.43±0.13	27.33±3.27	1.60±0.40	0.56±0.15	0.15±0.03	1.03±0.25	2.75±0.38	0.64±0.14	0.47±0.09	0.85±0.04
	Wet x W	92.33±13.6	5.92±0.15	34.33±5.96	0.12±0.01	0.61±0.04	0.31±0.09	0.53±0.23	2.47±0.60	1.45±0.12	0.86±0.30	0.55±0.04
Wet x SW	Wet x SW	47.00±2.31	6.73±0.18	34.00±1.15	0.06±0.003	0.12±0.03	0.06±0.01	ND	0.24±0.02	ND	0.05±0.02	0.07±0.02
	Wet x NW	11.61±1.03	7.07±0.08	41.83±3.03	ND	ND	ND	0.07±0.009	0.46±0.09	ND	ND	ND
Wet x SE	Wet x SE	174.50±18.33	6.85±0.11	21.33±5.48	ND	ND	ND	ND	0.80±0.10	ND	ND	ND
	Dry x S	863.42±119.07	5.63±0.17	102.75±7.40	1.09±0.17	1.19±0.21	0.49±0.14	1.13±0.24	4.43±0.58	1.12±0.23	0.65±0.14	0.87±0.14
Dry x W	Dry x W	1399.17±173.76	4.80±0.19	114.17±14.37	0.07±0.01	1.05±0.05	0.26±0.04	0.85±0.27	2.93±0.80	1.86±0.67	2.04±0.81	0.35±0.03
	Dry x SW	534.33±95.96	6.67±0.18	45.33±2.96	1.87±0.25	0.42±0.36	ND	0.07±0.01	0.31±0.02	ND	0.05±0.09	ND
Dry x NW	Dry x NW	156.28±50.92	7.0±0.04	62.40±9.64	1.73±0.03	0.24±0.02	0.07±0.01	0.13±0.02	0.64±0.01	0.05±0.001	ND	ND
	Dry x SE	367.17±58.13	6.92±0.14	26.00±5.03	0.89±0.11	ND	ND	ND	0.82±0.10	ND	ND	ND

ND – not detected

**Table 5.** Variation of water quality parameters with depth of well and distance from the tailings dam

Factor		SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Cd	Cu	Cl	Cr	Ni
Depth	R	-0.78	-0.63	-0.81		-0.85	-0.75	-0.78
	R <sup>2</sup>	0.61	0.40	0.66		0.72	0.56	0.61
Distance	R	-0.83	-0.7		-0.79	-0.77	-0.79	-0.70
	R <sup>2</sup>	0.69	0.49		0.62	0.59	0.6241	0.49

Significant ( $p < 0.05$ ) regression results with at least 50% variation being explained by the model

**Table 6.** Generalized linear model results with negative binomial errors. Values are log likelihood ratios (LRT) with level of significance.

Model	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	As	Cd	Cu	Fe	Cr	Ni	Pb
Season	1 109.9***	358.97 ***		4.29*						
Nature	7.24*	22.11***	9.17 ****	9.83 *	9.83*		9.17*			
Direction	100.92***	20.16***	5.13 ***			2.67*	10.18 ***			5.51*
Depth	151.97 ***	60.48***			10.61 **		10.01*	6.59*	6.1*	
Distance	478.65***	237.87***				6.20*	29.39 ***	16.51***	23.53***	
Distance x direction	6.78***	18.80***	5.41**			4.84*	3.93*		5.66*	
Distance x depth	119.26***	49.46 ****	6.7**							
Direction x depth	18.31***	15.82***								
Distance x season	14.69***	14.90***								
Direction x season	11.3***	5.67*								

\*significant at  $p < 0.05$ ; \*\*significant at  $p < 0.001$ ; \*\*\*significant at  $p < 0.0001$

**Table 7.** Spearman's correlation coefficient ( $r$ ) of groundwater parameters at RAN mine

	SO <sub>4</sub> <sup>2-</sup>	pH	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	As	Cd	Cu	Fe	Cr	Ni
pH	-0.55**									
Cl <sup>-</sup>	0.55**	-0.54**								
NO <sub>3</sub> <sup>-</sup>	-0.06	0.30*	-0.24							
As	0.55**	-0.50**	0.32*	0.18						
Cd	0.32*	-0.59**	0.33*	-0.09	0.67**					
Cu	0.43**	-0.57**	0.32*	0.02	0.54**	0.09				
Fe	0.48**	-0.66**	0.34**	0.10	0.45**	0.20	0.60**			
Cr	0.39*	-0.49**	0.23	-0.30	0.43**	0.65**	0.24	0.56**		
Ni	0.42**	-0.54**	0.37*	-0.08	0.63**	0.58**	0.58**	0.59**	0.76**	
Pb	0.26	-0.16	0.26	0.52**	0.53**	0.56**	0.24	0.28	0.11	0.30

\*significant at  $p < 0.05$ ; \*\*significant at  $p < 0.01$

**Table 8.** Estimated average daily dose (ADD, mg·kg<sup>-1</sup>·d<sup>-1</sup>) of PTEs for adults and children through ingestion and dermal contact with groundwater from shallow hand-dug wells

Well	As		Cd		Cr		Cu		Ni		Pb	
	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child
<b>Ingestion</b>												
N1	2.03 x 10 <sup>-5</sup>	4.07 x 10 <sup>-5</sup>	-	-	8.33 x 10 <sup>-6</sup>	1.67 x 10 <sup>-5</sup>	2.67 x 10 <sup>-6</sup>	5.33 x 10 <sup>-6</sup>	-	-	-	-
N2	-	-	1.17 x 10 <sup>-6</sup>	2.33 x 10 <sup>-6</sup>	-	-	3.00 x 10 <sup>-6</sup>	6.00 x 10 <sup>-6</sup>	-	-	-	-
N3	2.80 x 10 <sup>-5</sup>	5.60 x 10 <sup>-5</sup>	7.83 x 10 <sup>-6</sup>	1.57 x 10 <sup>-5</sup>	8.28 x 10 <sup>-5</sup>	1.66 x 10 <sup>-4</sup>	4.15 x 10 <sup>-5</sup>	8.30 x 10 <sup>-5</sup>	8.88 x 10 <sup>-5</sup>	1.78 x 10 <sup>-4</sup>	1.48 x 10 <sup>-5</sup>	2.97 x 10 <sup>-5</sup>
N4	2.33 x 10 <sup>-5</sup>	4.67 x 10 <sup>-5</sup>	1.52 x 10 <sup>-5</sup>	3.03 x 10 <sup>-5</sup>	3.38 x 10 <sup>-5</sup>	6.77 x 10 <sup>-5</sup>	3.20 x 10 <sup>-5</sup>	6.40 x 10 <sup>-5</sup>	2.45 x 10 <sup>-5</sup>	4.90 x 10 <sup>-5</sup>	3.22 x 10 <sup>-5</sup>	6.43 x 10 <sup>-5</sup>
N5	2.72 x 10 <sup>-5</sup>	5.43 x 10 <sup>-5</sup>	1.12 x 10 <sup>-5</sup>	2.23 x 10 <sup>-5</sup>	2.72 x 10 <sup>-5</sup>	5.43 x 10 <sup>-5</sup>	6.00 x 10 <sup>-6</sup>	1.20 x 10 <sup>-5</sup>	7.67 x 10 <sup>-6</sup>	1.53 x 10 <sup>-5</sup>	1.50 x 10 <sup>-5</sup>	3.00 x 10 <sup>-5</sup>
N6	9.00 x 10 <sup>-6</sup>	1.80 x 10 <sup>-5</sup>	1.00 x 10 <sup>-6</sup>	2.00 x 10 <sup>-6</sup>	-	-	1.17 x 10 <sup>-6</sup>	2.33 x 10 <sup>-6</sup>	1.57 x 10 <sup>-5</sup>	3.13 x 10 <sup>-5</sup>	1.17 x 10 <sup>-6</sup>	2.33 x 10 <sup>-6</sup>
N7	5.10 x 10 <sup>-5</sup>	1.02 x 10 <sup>-4</sup>	1.92 x 10 <sup>-5</sup>	3.83 x 10 <sup>-5</sup>	5.83 x 10 <sup>-5</sup>	1.17 x 10 <sup>-4</sup>	2.25 x 10 <sup>-5</sup>	4.50 x 10 <sup>-5</sup>	3.10 x 10 <sup>-5</sup>	6.20 x 10 <sup>-5</sup>	1.98 x 10 <sup>-5</sup>	3.97 x 10 <sup>-5</sup>
N8	2.17 x 10 <sup>-6</sup>	4.33 x 10 <sup>-6</sup>	9.67 x 10 <sup>-6</sup>	1.93 x 10 <sup>-5</sup>	8.17 x 10 <sup>-6</sup>	1.63 x 10 <sup>-5</sup>	1.50 x 10 <sup>-6</sup>	3.00 x 10 <sup>-6</sup>	1.50 x 10 <sup>-6</sup>	3.00 x 10 <sup>-6</sup>	2.32 x 10 <sup>-5</sup>	4.63 x 10 <sup>-5</sup>
N9	4.00 x 10 <sup>-5</sup>	8.00 x 10 <sup>-5</sup>	4.00 x 10 <sup>-6</sup>	8.00 x 10 <sup>-6</sup>	1.68 x 10 <sup>-5</sup>	3.37 x 10 <sup>-5</sup>	7.07 x 10 <sup>-5</sup>	1.41 x 10 <sup>-4</sup>	1.78 x 10 <sup>-5</sup>	3.57 x 10 <sup>-5</sup>	1.37 x 10 <sup>-5</sup>	2.73 x 10 <sup>-5</sup>
<b>Dermal contact</b>												
N1	1.17 x 10 <sup>-7</sup>	6.55 x 10 <sup>-7</sup>	-	-	1.59 x 10 <sup>-10</sup>	8.95 x 10 <sup>-10</sup>	5.10 x 10 <sup>-10</sup>	2.86 x 10 <sup>-9</sup>	-	-	-	-
N2	-	-	2.23 x 10 <sup>-10</sup>	1.25 x 10 <sup>-9</sup>	-	-	5.74 x 10 <sup>-10</sup>	3.22 x 10 <sup>-9</sup>	-	-	-	-
N3	1.61 x 10 <sup>-7</sup>	9.02 x 10 <sup>-7</sup>	1.50 x 10 <sup>-9</sup>	8.41 x 10 <sup>-9</sup>	1.58 x 10 <sup>-8</sup>	8.90 x 10 <sup>-8</sup>	7.94 x 10 <sup>-9</sup>	4.46 x 10 <sup>-8</sup>	1.70 x 10 <sup>-8</sup>	9.54 x 10 <sup>-8</sup>	2.84 x 10 <sup>-9</sup>	1.59 x 10 <sup>-8</sup>
N4	1.34 x 10 <sup>-7</sup>	7.52 x 10 <sup>-7</sup>	2.90 x 10 <sup>-9</sup>	1.63 x 10 <sup>-8</sup>	6.47 x 10 <sup>-9</sup>	3.63 x 10 <sup>-8</sup>	6.12 x 10 <sup>-9</sup>	3.44 x 10 <sup>-8</sup>	4.69 x 10 <sup>-9</sup>	2.63 x 10 <sup>-8</sup>	6.15 x 10 <sup>-9</sup>	3.45 x 10 <sup>-8</sup>
N5	1.56 x 10 <sup>-7</sup>	8.75 x 10 <sup>-7</sup>	2.14 x 10 <sup>-9</sup>	1.20 x 10 <sup>-8</sup>	5.20 x 10 <sup>-9</sup>	2.92 x 10 <sup>-8</sup>	1.15 x 10 <sup>-9</sup>	6.44 x 10 <sup>-9</sup>	1.47 x 10 <sup>-9</sup>	8.23 x 10 <sup>-9</sup>	2.87 x 10 <sup>-9</sup>	1.61 x 10 <sup>-8</sup>
N6	5.17 x 10 <sup>-8</sup>	2.90 x 10 <sup>-7</sup>	1.91 x 10 <sup>-10</sup>	1.07 x 10 <sup>-9</sup>	-	-	2.23 x 10 <sup>-10</sup>	1.25 x 10 <sup>-9</sup>	3.00 x 10 <sup>-9</sup>	1.68 x 10 <sup>-8</sup>	2.23 x 10 <sup>-10</sup>	1.25 x 10 <sup>-9</sup>
N7	2.93 x 10 <sup>-7</sup>	1.64 x 10 <sup>-6</sup>	3.67 x 10 <sup>-9</sup>	2.06 x 10 <sup>-8</sup>	1.12 x 10 <sup>-8</sup>	6.26 x 10 <sup>-8</sup>	4.30 x 10 <sup>-9</sup>	2.42 x 10 <sup>-8</sup>	5.93 x 10 <sup>-9</sup>	3.33 x 10 <sup>-8</sup>	3.79 x 10 <sup>-9</sup>	2.13 x 10 <sup>-8</sup>
N8	1.24 x 10 <sup>-8</sup>	6.98 x 10 <sup>-8</sup>	1.85 x 10 <sup>-9</sup>	1.04 x 10 <sup>-8</sup>	1.56 x 10 <sup>-9</sup>	8.77 x 10 <sup>-9</sup>	2.87 x 10 <sup>-10</sup>	1.61 x 10 <sup>-9</sup>	2.87 x 10 <sup>-10</sup>	1.61 x 10 <sup>-9</sup>	4.43 x 10 <sup>-9</sup>	2.49 x 10 <sup>-8</sup>
N9	2.30 x 10 <sup>-7</sup>	1.29 x 10 <sup>-6</sup>	7.65 x 10 <sup>-10</sup>	4.30 x 10 <sup>-9</sup>	3.22 x 10 <sup>-9</sup>	1.81 x 10 <sup>-8</sup>	1.35 x 10 <sup>-8</sup>	7.59 x 10 <sup>-8</sup>	3.41 x 10 <sup>-9</sup>	1.92 x 10 <sup>-8</sup>	2.61 x 10 <sup>-9</sup>	1.47 x 10 <sup>-8</sup>

**Table 9.** Estimated Hazard Quotient (HQ) and Hazard Index (HI) values of PTEs for adults and children through ingestion and dermal contact with well water

Well	Hazard Quotient (HQ)												Hazard Index (HI)					
	As			Cd			Cr			Cu			Ni			Pb		
	Adult	Child																
	HQ (ingestion)																	
N1	6.78 x 10 <sup>-2</sup>	1.36 x 10 <sup>-1</sup>	-	-	2.78 x 10 <sup>-3</sup>	5.56 x 10 <sup>-3</sup>	5.56 x 10 <sup>-3</sup>	6.67 x 10 <sup>-5</sup>	1.33 x 10 <sup>-4</sup>	-	-	-	-	-	-	-	7.06 x 10 <sup>-2</sup>	1.41 x 10 <sup>-1</sup>
N2	-	-	2.33 x 10 <sup>-3</sup>	4.67 x 10 <sup>-3</sup>	-	-	7.50 x 10 <sup>-5</sup>	1.50 x 10 <sup>-4</sup>	1.50 x 10 <sup>-4</sup>	-	-	-	-	-	-	-	2.41 x 10 <sup>-3</sup>	4.82 x 10 <sup>-3</sup>
N3	9.33 x 10 <sup>-2</sup>	1.87 x 10 <sup>-1</sup>	1.57 x 10 <sup>-2</sup>	3.13 x 10 <sup>-2</sup>	2.76 x 10 <sup>-2</sup>	5.52 x 10 <sup>-2</sup>	1.04 x 10 <sup>-3</sup>	2.08 x 10 <sup>-3</sup>	2.08 x 10 <sup>-3</sup>	4.44 x 10 <sup>-3</sup>	8.88 x 10 <sup>-3</sup>	8.88 x 10 <sup>-3</sup>	1.06 x 10 <sup>-2</sup>	2.12 x 10 <sup>-2</sup>	1.06 x 10 <sup>-2</sup>	2.12 x 10 <sup>-2</sup>	1.53 x 10 <sup>-1</sup>	3.05 x 10 <sup>-1</sup>
N4	7.78 x 10 <sup>-2</sup>	1.56 x 10 <sup>-1</sup>	3.03 x 10 <sup>-2</sup>	6.07 x 10 <sup>-2</sup>	1.13 x 10 <sup>-2</sup>	2.26 x 10 <sup>-2</sup>	8.00 x 10 <sup>-4</sup>	1.60 x 10 <sup>-3</sup>	1.60 x 10 <sup>-3</sup>	1.23 x 10 <sup>-3</sup>	2.45 x 10 <sup>-3</sup>	2.45 x 10 <sup>-3</sup>	2.30 x 10 <sup>-2</sup>	4.60 x 10 <sup>-2</sup>	2.30 x 10 <sup>-2</sup>	4.60 x 10 <sup>-2</sup>	1.44 x 10 <sup>-1</sup>	2.89 x 10 <sup>-1</sup>
N5	9.06 x 10 <sup>-2</sup>	1.81 x 10 <sup>-1</sup>	2.23 x 10 <sup>-2</sup>	4.47 x 10 <sup>-2</sup>	9.06 x 10 <sup>-3</sup>	1.81 x 10 <sup>-2</sup>	1.50 x 10 <sup>-4</sup>	3.00 x 10 <sup>-4</sup>	3.00 x 10 <sup>-4</sup>	3.83 x 10 <sup>-4</sup>	7.67 x 10 <sup>-4</sup>	7.67 x 10 <sup>-4</sup>	1.07 x 10 <sup>-2</sup>	2.14 x 10 <sup>-2</sup>	1.07 x 10 <sup>-2</sup>	2.14 x 10 <sup>-2</sup>	1.33 x 10 <sup>-1</sup>	2.66 x 10 <sup>-1</sup>
N6	3.00 x 10 <sup>-2</sup>	6.00 x 10 <sup>-2</sup>	2.00 x 10 <sup>-3</sup>	4.00 x 10 <sup>-3</sup>	-	-	2.92 x 10 <sup>-5</sup>	5.83 x 10 <sup>-5</sup>	5.83 x 10 <sup>-5</sup>	7.83 x 10 <sup>-4</sup>	1.57 x 10 <sup>-3</sup>	1.57 x 10 <sup>-3</sup>	8.33 x 10 <sup>-4</sup>	1.67 x 10 <sup>-3</sup>	8.33 x 10 <sup>-4</sup>	1.67 x 10 <sup>-3</sup>	3.36 x 10 <sup>-2</sup>	6.73 x 10 <sup>-2</sup>
N7	1.70 x 10 <sup>-1</sup>	3.40 x 10 <sup>-1</sup>	3.83 x 10 <sup>-2</sup>	7.67 x 10 <sup>-2</sup>	1.94 x 10 <sup>-2</sup>	3.89 x 10 <sup>-2</sup>	5.63 x 10 <sup>-4</sup>	1.13 x 10 <sup>-3</sup>	1.13 x 10 <sup>-3</sup>	1.55 x 10 <sup>-3</sup>	3.10 x 10 <sup>-3</sup>	3.10 x 10 <sup>-3</sup>	1.42 x 10 <sup>-2</sup>	2.83 x 10 <sup>-2</sup>	1.42 x 10 <sup>-2</sup>	2.83 x 10 <sup>-2</sup>	2.44 x 10 <sup>-1</sup>	4.88 x 10 <sup>-1</sup>
N8	7.22 x 10 <sup>-3</sup>	1.44 x 10 <sup>-2</sup>	1.93 x 10 <sup>-2</sup>	3.87 x 10 <sup>-2</sup>	2.72 x 10 <sup>-3</sup>	5.44 x 10 <sup>-3</sup>	3.75 x 10 <sup>-5</sup>	7.50 x 10 <sup>-5</sup>	7.50 x 10 <sup>-5</sup>	7.50 x 10 <sup>-5</sup>	1.50 x 10 <sup>-4</sup>	1.50 x 10 <sup>-4</sup>	1.65 x 10 <sup>-2</sup>	3.31 x 10 <sup>-2</sup>	1.65 x 10 <sup>-2</sup>	3.31 x 10 <sup>-2</sup>	4.59 x 10 <sup>-2</sup>	9.19 x 10 <sup>-2</sup>
N9	1.33 x 10 <sup>-1</sup>	2.67 x 10 <sup>-1</sup>	8.00 x 10 <sup>-3</sup>	1.60 x 10 <sup>-2</sup>	5.61 x 10 <sup>-3</sup>	1.12 x 10 <sup>-2</sup>	1.77 x 10 <sup>-3</sup>	3.53 x 10 <sup>-3</sup>	3.53 x 10 <sup>-3</sup>	8.92 x 10 <sup>-4</sup>	1.78 x 10 <sup>-3</sup>	1.78 x 10 <sup>-3</sup>	9.76 x 10 <sup>-3</sup>	1.95 x 10 <sup>-2</sup>	9.76 x 10 <sup>-3</sup>	1.95 x 10 <sup>-2</sup>	1.59 x 10 <sup>-1</sup>	3.19 x 10 <sup>-1</sup>
	HQ (dermal)																	
N1	4.09 x 10 <sup>-4</sup>	2.30 x 10 <sup>-3</sup>	-	-	2.13 x 10 <sup>-6</sup>	1.19 x 10 <sup>-5</sup>	4.25 x 10 <sup>-8</sup>	2.39 x 10 <sup>-7</sup>	2.39 x 10 <sup>-7</sup>	-	-	-	-	-	-	-	4.11 x 10 <sup>-4</sup>	2.31 x 10 <sup>-3</sup>
N2	-	-	4.46 x 10 <sup>-5</sup>	2.51 x 10 <sup>-4</sup>	-	-	4.78 x 10 <sup>-8</sup>	2.68 x 10 <sup>-7</sup>	2.68 x 10 <sup>-7</sup>	-	-	-	-	-	-	-	4.46 x 10 <sup>-5</sup>	2.51 x 10 <sup>-4</sup>
N3	5.64 x 10 <sup>-4</sup>	3.17 x 10 <sup>-3</sup>	3.00 x 10 <sup>-4</sup>	1.68 x 10 <sup>-3</sup>	2.11 x 10 <sup>-4</sup>	1.19 x 10 <sup>-3</sup>	6.62 x 10 <sup>-7</sup>	3.71 x 10 <sup>-6</sup>	3.71 x 10 <sup>-6</sup>	3.15 x 10 <sup>-6</sup>	1.77 x 10 <sup>-5</sup>	1.77 x 10 <sup>-5</sup>	6.76 x 10 <sup>-6</sup>	3.79 x 10 <sup>-5</sup>	6.76 x 10 <sup>-6</sup>	3.79 x 10 <sup>-5</sup>	1.09 x 10 <sup>-3</sup>	6.09 x 10 <sup>-3</sup>
N4	4.70 x 10 <sup>-4</sup>	2.64 x 10 <sup>-3</sup>	5.80 x 10 <sup>-4</sup>	3.26 x 10 <sup>-3</sup>	8.63 x 10 <sup>-5</sup>	8.84 x 10 <sup>-4</sup>	5.10 x 10 <sup>-7</sup>	2.86 x 10 <sup>-6</sup>	2.86 x 10 <sup>-6</sup>	8.68 x 10 <sup>-7</sup>	4.87 x 10 <sup>-6</sup>	4.87 x 10 <sup>-6</sup>	1.47 x 10 <sup>-5</sup>	8.23 x 10 <sup>-5</sup>	1.47 x 10 <sup>-5</sup>	8.23 x 10 <sup>-5</sup>	1.15 x 10 <sup>-3</sup>	6.47 x 10 <sup>-3</sup>
N5	5.47 x 10 <sup>-4</sup>	3.07 x 10 <sup>-3</sup>	4.27 x 10 <sup>-4</sup>	2.40 x 10 <sup>-3</sup>	6.93 x 10 <sup>-5</sup>	3.89 x 10 <sup>-4</sup>	9.57 x 10 <sup>-8</sup>	5.37 x 10 <sup>-7</sup>	5.37 x 10 <sup>-7</sup>	2.72 x 10 <sup>-7</sup>	1.52 x 10 <sup>-6</sup>	1.52 x 10 <sup>-6</sup>	6.83 x 10 <sup>-6</sup>	3.84 x 10 <sup>-5</sup>	6.83 x 10 <sup>-6</sup>	3.84 x 10 <sup>-5</sup>	1.05 x 10 <sup>-3</sup>	5.90 x 10 <sup>-3</sup>
N6	1.81 x 10 <sup>-4</sup>	1.02 x 10 <sup>-3</sup>	3.83 x 10 <sup>-5</sup>	2.15 x 10 <sup>-4</sup>	-	-	1.86 x 10 <sup>-8</sup>	1.04 x 10 <sup>-7</sup>	1.04 x 10 <sup>-7</sup>	5.55 x 10 <sup>-7</sup>	3.12 x 10 <sup>-6</sup>	3.12 x 10 <sup>-6</sup>	5.31 x 10 <sup>-7</sup>	2.98 x 10 <sup>-6</sup>	5.31 x 10 <sup>-7</sup>	2.98 x 10 <sup>-6</sup>	2.21 x 10 <sup>-4</sup>	1.24 x 10 <sup>-3</sup>
N7	1.03 x 10 <sup>-3</sup>	5.77 x 10 <sup>-3</sup>	7.33 x 10 <sup>-4</sup>	4.12 x 10 <sup>-3</sup>	1.49 x 10 <sup>-4</sup>	8.35 x 10 <sup>-4</sup>	3.59 x 10 <sup>-7</sup>	2.10 x 10 <sup>-6</sup>	2.10 x 10 <sup>-6</sup>	1.10 x 10 <sup>-6</sup>	6.17 x 10 <sup>-6</sup>	6.17 x 10 <sup>-6</sup>	9.03 x 10 <sup>-6</sup>	5.07 x 10 <sup>-5</sup>	9.03 x 10 <sup>-6</sup>	5.07 x 10 <sup>-5</sup>	1.92 x 10 <sup>-3</sup>	1.08 x 10 <sup>-2</sup>
N8	4.36 x 10 <sup>-5</sup>	2.45 x 10 <sup>-4</sup>	3.70 x 10 <sup>-4</sup>	2.08 x 10 <sup>-3</sup>	2.08 x 10 <sup>-5</sup>	1.17 x 10 <sup>-4</sup>	2.39 x 10 <sup>-8</sup>	1.34 x 10 <sup>-7</sup>	1.34 x 10 <sup>-7</sup>	5.31 x 10 <sup>-8</sup>	2.98 x 10 <sup>-7</sup>	2.98 x 10 <sup>-7</sup>	1.06 x 10 <sup>-5</sup>	5.92 x 10 <sup>-5</sup>	1.06 x 10 <sup>-5</sup>	5.92 x 10 <sup>-5</sup>	4.45 x 10 <sup>-4</sup>	2.51 x 10 <sup>-3</sup>
N9	8.05 x 10 <sup>-4</sup>	4.52 x 10 <sup>-3</sup>	8.59 x 10 <sup>-4</sup>	8.59 x 10 <sup>-4</sup>	4.29 x 10 <sup>-5</sup>	2.41 x 10 <sup>-4</sup>	1.13 x 10 <sup>-6</sup>	6.32 x 10 <sup>-6</sup>	6.32 x 10 <sup>-6</sup>	6.32 x 10 <sup>-7</sup>	3.55 x 10 <sup>-6</sup>	3.55 x 10 <sup>-6</sup>	6.22 x 10 <sup>-6</sup>	3.49 x 10 <sup>-5</sup>	6.22 x 10 <sup>-6</sup>	3.49 x 10 <sup>-5</sup>	1.01 x 10 <sup>-3</sup>	5.67 x 10 <sup>-3</sup>
	HI (ingestion + dermal)																	
N1	6.82 x 10 <sup>-2</sup>	1.38 x 10 <sup>-1</sup>	-	-	2.80 x 10 <sup>-3</sup>	5.57 x 10 <sup>-3</sup>	1.00 x 10 <sup>-4</sup>	1.33 x 10 <sup>-4</sup>	1.33 x 10 <sup>-4</sup>	-	-	-	-	-	-	-	7.10 x 10 <sup>-2</sup>	1.43 x 10 <sup>-1</sup>
N2	-	-	2.34 x 10 <sup>-3</sup>	4.92 x 10 <sup>-3</sup>	-	-	1.00 x 10 <sup>-4</sup>	1.50 x 10 <sup>-4</sup>	1.50 x 10 <sup>-4</sup>	-	-	-	-	-	-	-	2.45 x 10 <sup>-3</sup>	5.07 x 10 <sup>-3</sup>
N3	9.39 x 10 <sup>-2</sup>	1.90 x 10 <sup>-1</sup>	1.60 x 10 <sup>-2</sup>	3.30 x 10 <sup>-2</sup>	2.78 x 10 <sup>-2</sup>	5.64 x 10 <sup>-2</sup>	1.00 x 10 <sup>-3</sup>	2.08 x 10 <sup>-3</sup>	2.08 x 10 <sup>-3</sup>	4.40 x 10 <sup>-3</sup>	8.90 x 10 <sup>-3</sup>	8.90 x 10 <sup>-3</sup>	1.06 x 10 <sup>-2</sup>	2.12 x 10 <sup>-2</sup>	1.06 x 10 <sup>-2</sup>	2.12 x 10 <sup>-2</sup>	1.54 x 10 <sup>-1</sup>	3.11 x 10 <sup>-1</sup>
N4	7.83 x 10 <sup>-2</sup>	1.59 x 10 <sup>-1</sup>	3.09 x 10 <sup>-2</sup>	6.40 x 10 <sup>-2</sup>	1.14 x 10 <sup>-2</sup>	2.35 x 10 <sup>-2</sup>	8.01 x 10 <sup>-4</sup>	1.60 x 10 <sup>-3</sup>	1.60 x 10 <sup>-3</sup>	1.20 x 10 <sup>-3</sup>	2.45 x 10 <sup>-3</sup>	2.45 x 10 <sup>-3</sup>	2.30 x 10 <sup>-2</sup>	4.61 x 10 <sup>-2</sup>	2.30 x 10 <sup>-2</sup>	4.61 x 10 <sup>-2</sup>	1.45 x 10 <sup>-1</sup>	2.95 x 10 <sup>-1</sup>
N5	9.11 x 10 <sup>-2</sup>	1.84 x 10 <sup>-1</sup>	2.27 x 10 <sup>-2</sup>	4.71 x 10 <sup>-2</sup>	9.17 x 10 <sup>-3</sup>	1.85 x 10 <sup>-2</sup>	2.00 x 10 <sup>-4</sup>	3.01 x 10 <sup>-4</sup>	3.01 x 10 <sup>-4</sup>	4.00 x 10 <sup>-4</sup>	7.69 x 10 <sup>-4</sup>	7.69 x 10 <sup>-4</sup>	1.07 x 10 <sup>-2</sup>	2.14 x 10 <sup>-2</sup>	1.07 x 10 <sup>-2</sup>	2.14 x 10 <sup>-2</sup>	1.34 x 10 <sup>-1</sup>	2.72 x 10 <sup>-1</sup>
N6	3.02 x 10 <sup>-2</sup>	6.10 x 10 <sup>-2</sup>	2.04 x 10 <sup>-3</sup>	4.22 x 10 <sup>-3</sup>	-	-	1.86 x 10 <sup>-8</sup>	5.84 x 10 <sup>-5</sup>	5.84 x 10 <sup>-5</sup>	8.01 x 10 <sup>-4</sup>	1.57 x 10 <sup>-3</sup>	1.57 x 10 <sup>-3</sup>	8.01 x 10 <sup>-4</sup>	1.67 x 10 <sup>-3</sup>	8.01 x 10 <sup>-4</sup>	1.67 x 10 <sup>-3</sup>	3.38 x 10 <sup>-2</sup>	6.85 x 10 <sup>-2</sup>
N7	1.71 x 10 <sup>-1</sup>	3.46 x 10 <sup>-1</sup>	3.90 x 10 <sup>-2</sup>	8.08 x 10 <sup>-2</sup>	1.95 x 10 <sup>-2</sup>	3.97 x 10 <sup>-2</sup>	6.00 x 10 <sup>-4</sup>	1.13 x 10 <sup>-3</sup>	1.13 x 10 <sup>-3</sup>	1.60 x 10 <sup>-3</sup>	3.11 x 10 <sup>-3</sup>	3.11 x 10 <sup>-3</sup>	1.42 x 10 <sup>-2</sup>	2.84 x 10 <sup>-2</sup>	1.42 x 10 <sup>-2</sup>	2.84 x 10 <sup>-2</sup>	2.46 x 10 <sup>-1</sup>	4.99 x 10 <sup>-1</sup>
N8	7.24 x 10 <sup>-3</sup>	1.46 x 10 <sup>-2</sup>	1.97 x 10 <sup>-2</sup>	4.08 x 10 <sup>-2</sup>	2.72 x 10 <sup>-3</sup>	5.56 x 10 <sup>-3</sup>	2.39 x 10 <sup>-8</sup>	7.51 x 10 <sup>-5</sup>	7.51 x 10 <sup>-5</sup>	1.00 x 10 <sup>-4</sup>	1.50 x 10 <sup>-4</sup>	1.50 x 10 <sup>-4</sup>	1.65 x 10 <sup>-2</sup>	3.32 x 10 <sup>-2</sup>	1.65 x 10 <sup>-2</sup>	3.32 x 10 <sup>-2</sup>	4.63 x 10 <sup>-2</sup>	9.44 x 10 <sup>-2</sup>
N9	1.34 x 10 <sup>-1</sup>	2.72 x 10 <sup>-1</sup>	8.15 x 10 <sup>-3</sup>	1.69 x 10 <sup>-2</sup>	5.46 x 10 <sup>-3</sup>	1.14 x 10 <sup>-2</sup>	1.80 x 10 <sup>-3</sup>	3.54 x 10 <sup>-3</sup>	3.54 x 10 <sup>-3</sup>	9.01 x 10 <sup>-4</sup>	1.78 x 10 <sup>-3</sup>	1.78 x 10 <sup>-3</sup>	9.81 x 10 <sup>-3</sup>	1.95 x 10 <sup>-2</sup>	9.81 x 10 <sup>-3</sup>	1.95 x 10 <sup>-2</sup>	1.60 x 10 <sup>-1</sup>	3.25 x 10 <sup>-1</sup>

- no value, concentration of the PTE was below the limit of detection

## DISCUSSION

Overall results show that groundwater had higher concentrations of contaminants during the dry than the wet season. This is consistent with earlier studies for the concentrations of  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  (Ganiyu et al., 2018) and  $\text{NO}_3^-$  (Bexfield and Jurgens, 2014). This could be attributed to change in the intensity of hydrochemical processes (e.g., evaporation, redox and mineral precipitation) in the dry season (Aladejana et al., 2020). Wells are over-exploited during the dry season to meet high water demand which is usually offset by rainfall during the wet season. This reduces groundwater recharge and lowers the water table (Guo et al., 2018). Further, it influences the direction and magnitude of the horizontal and vertical gradients that drive contaminants to wells, changing groundwater quality (Bexfield and Jurgens, 2014; Mohapatra and Kirpalani, 2017).

The main source of  $\text{NO}_3^-$  in groundwater was agricultural activities. This conclusion was also reached in a groundwater assessment study in Andhra Pradesh, India (Sunitha and Reddy, 2019). Urban agriculture is associated with intensive use of artificial fertilisers on small pieces of land (Stewart et al., 2013). Nitrate causes methaemoglobinaemia, significant in children (WHO, 2017). High concentrations of  $\text{SO}_4^{2-}$  in groundwater could be from the oxidation of sulphidic material in tailings. Leaching from tailings results in an increase of oxyanions (e. g.  $\text{SO}_4^{2-}$ ) in solution (Dold, 2014; Kim et al., 2017). Acidic pH (less than 6) recorded in Wells N3, N5 and N9 could result in dissolution of the redox-sensitive Fe (Gad et al., 2016). According to WHO (2017), drinking water quality has no health-based guideline values for pH, Fe,  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$ . However, at some elevated concentrations, Fe,  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  influence the taste of water. The current study had groundwater of slightly acidic, neutral to slightly alkaline pH. This could be a result of either natural or enhanced neutralising potential by treatment of the mine tailings (Petronijević et al., 2020).

Groundwater sampled in open wells (25–30 m deep) during summer near an active coal mine in Dhanbad, India (Prasad et al., 2014), had higher concentrations of Cd, Cr, Cu, Fe and Pb when compared to the current study. Coal mining is generally associated with acid mine drainage laden with PTEs which can leach deep into groundwater (Li et al., 2021). Zhuang et al. (2014) showed that well water from sources near a mining area of Dabaoshan, Guangdong, southern China, had higher concentrations of Cd, Cu and Pb than those found in the current study. Analysis of the hair of local residents showed higher concentrations of Cd and Pb than an unexposed population. However, comparison of the concentration of PTEs in groundwater from various studies may present challenges due to variations in mineral exploitation activities and duration, local climate and geology, soil and well characteristics. Nevertheless, comparing with the maximum allowable limit for drinking water quality guidelines allows water quality assessments and monitoring.

The area between the sector formed by NW and S directions within 75 m from the nearest MTD toe ( $135^\circ$ ) recorded the highest concentrations of contaminants in groundwater. Surface runoff erodes PTEs downslope to the south during the rainy season (Fig. 2a). In dry weather, long-range atmospheric deposition (Barandovski et al., 2015) accounts for PTE dispersal into open SHDWs (Fig. 2b) through the prevailing easterly winds (Kanda et al., 2019). High concentrations of  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ , Cd and Fe were recorded at relatively shallow wells. This is in agreement with observations made by Rajmohan et al. (2017) in a hydrochemical evaluation of 44 wells in the Ramganga Sub-Basin, India, for the same groundwater quality parameters. They attributed the higher concentrations in shallow wells (average 12 m) than in deep wells (average 35 m) to other sources of contamination than mineral

dissolution alone. Dash et al. (2010) showed that salinity of groundwater was high at shallow groundwater depths.

The three new wells recorded higher concentrations of contaminants (except for  $\text{SO}_4^{2-}$ ) than existing wells. However, these wells were not of the same depth, distance and direction relative to the MTD, which all influence groundwater quality. The concentration of  $\text{SO}_4^{2-}$  in groundwater was considered less prone to sorption, precipitation and geochemical reduction (Kim et al., 2017). Erickson et al. (2018) observed that the concentration of As in groundwater from new wells was highly variable relative to that found when sampled months later. Although the concentrations of PTEs were lower than the drinking water quality guidelines (WHO, 2017), some (e.g. Cd and Pb) are cumulative poisons (Jaishankar et al., 2014) while Ni is considered carcinogenic (Wuana and Okieimen, 2011).

The negative and significant correlation of PTEs with pH ( $p < 0.05$ ) may suggest that pH influences the concentration of PTEs in solution (Palansooriya et al., 2020). Generally, strong positive correlations among PTEs may suggest that they originate from the same source or have similar hydrochemical behaviour (Tepanosyan et al., 2017). However, further multivariate statistical analyses are needed for their source apportionment.

Health risk assessment data showed that both HQi and HI were less than one. This implies that there is no possibility that non-carcinogenic impacts may occur to the local population (USEPA, 2011). The use of PTE concentrations for the dry season, when concentrations of PTEs are maximum, to estimate health risk exposure may represent the worst-case exposure scenario. However, average values (dry and rainy season) may represent a more realistic exposure scenario. Although Cd and Ni are human carcinogens (IARC, 1993), they have no established oral carcinogenic slope factors for groundwater, and Pb is a probable carcinogen. The TCR values recorded for arsenic ( $10^{-4}$ ) in some wells indicate medium to high cancer risk levels (Grade V:  $10^{-4}$ ,  $5 \times 10^{-4}$ ) which require the willingness to invest in managing them (Li et al., 2017). According to Tepanosyan et al. (2017) if TCR or total TCR is less than  $10^{-6}$  no carcinogenic risk is expected to health. Further, values greater than  $10^{-4}$  suggest high risk (USEPA, 1989).

### Limitations of the study

The use of the USEPA-recommended risk assessment model in local scenarios may introduce uncertainties. However, local values were used in the model where applicable. The model assumes continuous use of water from the same source in a year and does not indicate when potential risk symptoms may start to show. There were no established local background values for PTEs. A local reference point was used to provide background concentrations.

Concentrations of most PTEs were below the detection limit. Since groundwater quality is influenced by a number of changing local factors, our results may be interpreted with caution for generalisation to similar scenarios. However, they provide a basis for further monitoring and may influence relevant practices and policy for local authorities.

## CONCLUSION

The study highlights the importance of monitoring abandoned mine sites and the ambient environment even years after mineral exploration activities have ended. It encourages the effective enforcement of urban councils' by-laws for human settlement and environmental regulations that govern mining and waste disposal. The findings stress the importance of seasonality in similar studies as the population appears exposed to higher concentrations of potentially toxic elements during the dry than

the wet season. Although the concentrations of potentially toxic elements in groundwater placed no restriction for potable use and posed no significant non-carcinogenic health risks to the local population, arsenic poses medium to high cancer risk. We encourage municipalities of small towns in low-income settings to provide water and sewerage systems to new human settlements before habitation. We propose the establishment of vegetation on slopes of the MTD to reduce erosion by wind and runoff, and prevent the oxidation of pyrite and the mobility of PTEs. Eroded tailings material downslope of the tailings dam may be contained by silt-catchment paddocks. Further studies could be done to investigate the acid generating potential of the tailings and the potential human health risk to potentially toxic elements through inhalation for the local population.

## DISCLOSURE STATEMENT

The authors declare no conflicts of interest with the contents of this article. This study did not receive any external funding.

## ETHICAL CONSIDERATIONS

No human subjects were directly involved. However, verbal consent to take water samples from wells at some households was sought.

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