RESEARCH ARTICLE



# Appraisal of pollution scenario, sources and public health risk of harmful metals in mine water of Barapukuria coal mine industry in Bangladesh

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## Abstract

This study was conducted to assess the accumulation and sources of harmful metals and associated public health risk from the usage of underground mine water of Barapukuria coal mine in Bangladesh, keeping in mind the optimum reuse. Thirty underground mine water samples had been analyzed for assessing temperature, pH, EC, TC, DO, BOD, COD, Ca, K, S, Ti, Mn, V, Fe, Co, Ni, Cu, Zn, Br, Rb, Sr, and Pb. Numerous pollution evaluation and health risk assessment indices along with multivariate statistical tools were employed in this study to apprise the pollution scenario, controlling factors, and probable health risk. The chronic or persistent health risk of metals via oral and dermal exposure of adults and children was determined using the hazard quotient (HQ) and hazard index (HI). The results showed that the content of physicochemical parameters and potentially harmful elements in water samples was many folds higher than the national and international standards. The results of pollution evaluation indices indicate that coal seam-leached mine water is highly concentrated by potentially harmful metals and not suitable for drinking, agriculture, and aquatic lives. The correlation coefficients and multivariate analysis illustrate both the geological and anthropogenic factors controlling the variability of metals in mine water. Results of  $HQ<sub>oral</sub>$  value suggest that V, Co, and Pb are significant health risk for adults and Mn, V, Co, Cu, and Pb are for children. Vanadium is found potential for dermal effects, and HI<sub>dermal</sub> value directs 33%, and 70% samples exceed the safe limit for adults and children, respectively. The HI value suggests that oral exposure to harmful metals creates more harm than dermal absorption, and children are more vulnerable than adults. It is anticipated that the outcomes of this study would deliver expedient insights to initiate necessary steps to minimize the public health risk by applying appropriate environmental protocols.

Keywords Coal mining . Heavy metals . Mine water . Multivariate statistics . Pollution indices

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## Introduction

Due to rapid economic growth and growing call for energy, underground coal mining operations are increasing worldwide, which pose severe effects on environment (Hossain et al. [2015](#page-15-0); Tiwari et al. [2017;](#page-16-0) Wright et al. [2018](#page-16-0); Habib et al. [2019](#page-15-0)). Coal mining processes are well-known sources of water pollution because of mine drainage to local waterways (Wright et al. [2017](#page-16-0), [2018](#page-16-0)). Usually, discharged mine water is highly acidic depends on the sulfur content in coal that could generate  $H_2SO_4$  which leaches and mobilizes heavy metals from the surrounding geology (Zhao et al. [2007,](#page-17-0) Bhuiyan et al. [2010a;](#page-15-0) Wright et al. [2018](#page-16-0)). Though ecology and environment face huge challenges, in most cases, the mining technology or operational methods do not maintain environmental management protocols. Therefore, the largescale operation from the coal mining industries creates enormous adverse impacts on nearby surface and groundwater as well as the surrounding environment (Bhuiyan et al. [2010a,](#page-15-0) [2010b](#page-15-0); Zakir et al. [2013](#page-17-0); Hossain et al. [2015](#page-15-0); Linn et al. [2016](#page-16-0); Howladar et al. [2017](#page-16-0); Liao et al. [2017;](#page-16-0) Gao et al. [2019](#page-15-0); Dong et al. [2020](#page-15-0)).

To promote the socio-economic and environmental sustainability, reuse of mine water in agricultural and industrial sectors is encouraging nowadays. Many potential exploits were found with mine discharge water in industry, irrigation, recreational, waterfowl habitat, and tourism in many parts of the world (Otchere et al. [2004;](#page-16-0) McCullough and Lund [2006](#page-16-0); Petritz et al. [2009](#page-16-0); Gammons et al. [2009;](#page-15-0) Howladar et al. [2017\)](#page-16-0). Some projects might be exemplified worldwide which were evaluating the prospect of an underground mine discharged water for use in the heating system (Raymond and Therrien [2008;](#page-16-0) Renz et al. [2009](#page-16-0)). However, theoretically, mine effluent is a composition of coal seam-leached water with geologic formation. The Pleistocene geological formation is considered as the major aquifer system in the Barapukuria coal mine (BCM) area of Bangladesh (Khan et al. [2017\)](#page-16-0). The Gondwana coal-bearing sedimentary unit (Gondwana formation) is highly saturated with groundwater (Bhuiyan et al. [2010a\)](#page-15-0). Therefore, there is a potential source of groundwater production in this mining industry. The BCM underground mine operation has, thus, perturbed the waterbearing sedimentary formation in different ways such as subsidence, depletion of groundwater level, quality of water, and environmental condition of the surrounding area. The annual average mine drainage in BCM is around 4,000,000 l. In Bangladesh, BCM authority usually discharges this mine effluent to the natural environment without any treatment (Bhuiyan et al. [2010b\)](#page-15-0). So, streams, canals, and water bodies draining to the eastern and western parts of the Barapukuria coalfield are polluted and pose a risk for irrigation and aquatic ecology (Bhuiyan et al. [2010a,](#page-15-0) [2010b](#page-15-0); Zakir et al. [2013](#page-17-0); Howladar et al. [2014;](#page-16-0) Hossain et al. [2015;](#page-15-0) Sultana et al.

[2016;](#page-16-0) Khan et al. [2017\)](#page-16-0). Hence, the public health of the surrounding area is under threat because of the probable metals transferring into the food chain.

Certain parts of the underground mine subsided and developed fractures and lineaments on the mine roof and surface area. Therefore, surface water along with rainwater may be infiltrated into the lithologic units through these weak points and contaminated the in situ mine water. The infiltrated water contain heavy metals from anthropogenic sources. Hence, the application of a multivariate approach may be a suitable method to apportion the sources of heavy metals in the collected water samples. Therefore, a comprehensive assessment regarding the evaluation of harmful metals' pollution, their source, and health risk is important. The aims of this study were to (i) determine the potentially harmful elements and their sources in mine-seepage water and (ii) delineate the pollution scenarios along with the public health risk assessment of the BCM area. In line with study aims, several pollution evaluation indices were employed to assess the pollution status and suitability of usages for drinking, irrigation and aquaculture. Multivariate statistical analysis was employed to interpret the data set for a better explanation of the pollution status and identifying the probable pollution sources. Furthermore, human health risk was assessed by considering the oral and dermal exposure of metals to evaluate the degree of detrimental health effects. The collective application of these methods will provide the wholestic picture of sources, contamination level, and exposure of heavy metals to human health. It is expected that the outcome of this study will help to aware the mining authority and policymaker to take necessary steps for environmental safe management of mine water for sustainable water resource management and public health safety.

#### Study area

#### Geographical settings

The BCM is situated at Parbatipur Upazila of the Dinajpur district in Bangladesh. Geographically, it lies between 25,031′ 46″ N-25033′ 5″ N and 88,057′ 48″ E-88058′ 53″ E (Fig. [1\)](#page-2-0). The area is relatively flat, considered as major riceproducing (> 60%) zones in Bangladesh (Bhuiyan et al. [2010a\)](#page-15-0). Parbatipur area is considered a major part of the Tista River floodplain, which lies at the northeastern margin of Barind Tract (Pleistocene Terrace). The Little Jamuna, Atrai, Karatoa, and Banglai rivers are flowing over the study area originating from the Himalayas (Bhuiyan et al. [2010b\)](#page-15-0). These rivers are draining out water to the Little Jamuna in the west and Jabuneswari in the east. Ashorar Beel (300 ha), an ecologically important aquatic body, is surrounded by the <span id="page-2-0"></span>Fig. 1 Location, geology, and geotectonic map of BCM of northwestern Bangladesh



national Shal forest (217 ha), which receives the mine water from the BCM industry (Banglapedia [2016\)](#page-15-0).

#### Geology and tectonic settings

The study area (Dinajpur) lies in the northern part of the Bengal Basin, one of the major sedimentary basins in the world. It has three major tectonic components, i.e., (i) Dinajpur shield and basement, (ii) central deep sedimentary basin, and (iii) Tripura-Sylhet-Chittagong folded belt (Khan and Chouhan [1996\)](#page-16-0). The Gondwana coal-bearing formation is situated in the Dinajpur Shield, fenced by Himalayan Foredeep, Shillong Massif, and the Indian Peninsular Shield to the north, east, and west, respectively. The Gondwana coal basins are situated in the corridor of the Garo-Rajmahal gap, locally named as "Rangpur Saddle" (Khan [1991](#page-16-0); Alam et al. [2003\)](#page-15-0). The Rangpur Saddle is the thinnest sedimentary cover (128 m at Madhyapara) over the basement. The Dinajpur slope is 64 km wide and inclines to Sub-Himalayan Foredeep. The alignment of the basement up to Bogra is gentle, which further increases towards the southeast. Gondwana sediments were trapped in the faulted troughs in the complex basin (Alam et al. [2003\)](#page-15-0). Therefore, the BCM area belongs to complex hydrogeological systems comparing with other areas in Bangladesh. The major components of the aquifer in this area are sandstones of unconsolidated Dupi Tila Formation (36 m thick coal seam), which extends thousands of square kilometers (Islam and Hayashi [2008;](#page-16-0) Habib et al. [2019\)](#page-15-0). This average thickness of the aquifer system is about 100 m, which showed 185 m in the southern part covering of Barind Clay residuum (Islam and Hayashi [2008;](#page-16-0) Howladar et al. [2014\)](#page-16-0).

However, the unconsolidated Dupi Tila Formation is influencing the water flow in seam VI (Table [1\)](#page-3-0), corresponding to a probable threat to underground mine flood. The

Age	Lithologic unit	Hydrostratigraphic unit	Lithology	Thickness (m)
Pleistocene	Barind Clay Residuum	Aquiclude	Clay and sandy clay	10
	Upper Dupi Tila	Aquifer	Medium sand inter-bedded with fine sand, pebbly grit and thin clay	104
Pliocene	Lower Dupi Tila	Aquiclude	Weathered residual clay, clay silts, sandstone inter-bedded with silty mudstone and coarse-grain quartz	80
	Gondwana	Aquifer	Medium to coarse-grained sandstone and pebbly sandstone, inter-bedded with coal seam I to V; also siltstone and mudstone	156
Permian	(I) Sandstone of seam VI Roof Aquifer		Medium to coarse-grained sandstone, grit stone, inter-bedded with thin medium- to fine-grained sandstone, siltstone and mudstone are some times	140
	(II) Sandstone of Seam VI Floor	Aquifer	Fine-grained sandstone, medium- to fine-grained sandstone inter-bedded with siltstone, carbonaceous mudstone and 2/3 beds of tuffy siltstone	67
Archean	Basement complex			
	Relatively aquiclude (a) Upper section		Sedimentary, igneous, and metamorphic rocks with sandy and muddy fragments inter-bedded with fine-grained sandstone, carbonaceous mudstones and molted mudstones at bottom	53
	(b) Lower Section	Aquifuse	Granodiorite, quartz diorite and diorite gness	31

<span id="page-3-0"></span>Table 1 Stratigraphy and hydrostratigraphic units of the Barapukuria coal mine area (Bakr et al. [1987](#page-15-0); CMC [1994](#page-15-0); Khan et al. [2017\)](#page-16-0)

stratigraphic unit of the study area is divided into the following units: (a) Basement complex, (b) Gondwana Group, (c) Dupi Tila Formation, and (d) Barind Clay residuum (Table 1) are illustrated in Fig. [2.](#page-4-0) The Carboniferous Gondwana Formation mainly comprises of Alluvium (0–0.50 m), Barind Clay residuum (0.5–6.0 m), Dupitila Formation (6.0– 120.0 m), Tura Sandstone (120.0–121.0 m) and shale, and Gondwana sand (160.0–259.0 m) which consider as the major coal sequence (Table 1 and Fig. [2](#page-4-0)). According to hydrogeological classification, the Barind Clay unit is known as an aquiclude. Its thickness is 10 m from the surface with an infiltration rate of about 1.5 mm/day. However, the upper Dupi Tila aquifer (UDT) constitutes the major groundwater reservoir (Howladar et al. [2014](#page-16-0)). The thickness and depth of the units are 102 and 136 m, respectively. The Lower Dupi Tila (LDT), 80 m thick, is also known as an aquiclude with a depth range from 115 to 118 m (Fig. [2](#page-4-0)). The Gondwana sandstones are separated into upper and lower sections by seam VI (Islam and Hayashi [2008](#page-16-0)). The thickness of the upper section is 156 m, placed at the center part in the basin (Khan et al. [2017\)](#page-16-0). It is composed of a mixture of pebbly sands, medium to coarse sands, and silts and mud, inter-bedded with coal seam I–V (Bakr et al. [1987;](#page-15-0) CMC [1994;](#page-15-0) Howladar et al. [2014](#page-16-0)). The thickness of the lower unit differs from 107 to 244 m from north-west to the southeastern part.

# Materials and methods

# Sampling and analytical procedure

A total of 30 water samples were collected from different points of the inner part of the mine complex, coal seams,

and litho-stratigraphic units. The depth of the coal mine ranged from 120 to 500 m from the surface. Pre-conditioned 500 mL plastic bottles were used for sampling, which were rinsed sufficiently by sample water before taking the samples. However, temperature, dissolved oxygen (DO), electrical conductivity (EC), and pH were measured onsite by field kits (HANNA instruments, model: HI 9143; pH meter: model 3051; and EC meter: model 4070 of JENWAY instruments). Moreover, biochemical oxygen demand (BOD) and chemical oxygen demand (COD) was measured by the standard method (APHA [1992](#page-15-0)). Coliform colonies (TC) were identified and counted following the standard procedure (APHA [1995](#page-15-0)). However, the collected water samples were filtered using a filter membrane of 0.45 μm in size (Gelman Inst. Co, London) and preserved following standard methods for elemental analysis. In this study, elemental analysis of the collected water samples was performed using Perkin-Elmer AAS (Model: 3110). SRM-1640 (groundwater) and NIST reference standard materials were used to verify the analytical precision of the AAS analytical technique, where the precision was within 8% for the analyzed elements. The detection limit of the studied elements was measured from the average and standard deviations of 7 blanks. The minimum detection limits (MLDs) of metals were  $(mg/L)$  0.035(Ca), 0.06(K), 2.791(S), 0.131(Ti), 0.091(V), 0.071(Cr), 0.047(Mn), 0.037(Fe), 0.071(Co), 0.028(Ni), 0.026(Cu), 0.027(Zn), 0.027(As), 0.036(Br), 0.047(Rb), 0.057(Sr), and 0.01(Pb).

#### Pollution evaluation indices

To understand the pollution status, numerous indices, including the heavy metal pollution index (HPI), heavy metal evaluation index (HEI), and the degree of contamination <span id="page-4-0"></span>Fig. 2 Subsurface geology of Barapukuria coal mine area of Bangladesh



(Cd), were employed in this study using the strategies defined in Table [2.](#page-5-0) HPI was employed to illustrate the pollution potential in terms of heavy metal concentration (Bhuiyan et al. [2010a\)](#page-15-0). It is generally described as an inverse relation to the standards values for individual elements (Mohan et al. [1996\)](#page-16-0). For drinking water, the critical value of HPI is considered as 100 (Prasad and Bose [2001](#page-16-0); Bhuiyan et al. [2010a;](#page-15-0) Tiwari et al. [2017](#page-16-0)). Again, HEI provides an understanding of the overall quality of mine water in the context of heavy metals (Edet and Offiong [2002](#page-15-0)). The degree of the HEI became categorized into 3 classes, i.e.,  $HEI \le 10$ : low,  $HEI$  (10–20): medium, and  $HEI > 20$ : high (Edet and Offiong [2002](#page-15-0); Bodrud-Doza et al. [2019b](#page-15-0)). Moreover,  $C_d$  was considered as a reference for assessing the magnitude of metal pollution (Backman et al. [1997](#page-15-0)). The pollution degree became classified into the following categories;  $C_d < 1$ : low,  $C_d$  (1–3): medium, and  $C_d > 3$ : high (Edet and Offiong [2002](#page-15-0); Islam et al. [2016](#page-16-0); Bodrud-Doza et al. [2019b\)](#page-15-0).

#### Statistical analysis

A set of statistical techniques, together with the Pearson correlation matrix (PCM), principal component analysis (PCA), and cluster analysis (CA), were applied in this study by using SPSS software (version 22.0), for windows. PCA was carried out to understand the probable origin of metals. Exploratory factor analysis (FA) was executed using the varimax rotation method (Howitt and Cramer [2005](#page-16-0)), which reduced the variable number with high loading on each component, thus easing the interpretation of the results of PCA. Principal components (PCs) identify the most significant parameters from the entire data set and reduce the data with minimum modification of the original records (Vega et al. [1998;](#page-16-0) Shrestha and Kazama

Evaluation indices	Equations	Description	References
Heavy metal pollution index (HPI)	$HPI = \frac{\sum\limits_{i=1}^{n} WiQi}{\sum\limits_{i=1}^{n} Wi}$	Here, $W_i$ = unit weight of $i^{th}$ elements, sub-index $Q_i$ = the $i^{th}$ elements, Mohan et al. (1996), Bhuiyan et al. $M_i$ = observed value in the $i^{th}$ elements, $I_i$ = ideal value of the $i^{th}$ elements, and $S_i$ = standard value of the $i^{th}$ parameters	$(2010a)$ , Tiwari et al. $(2017)$
	where, $Qi = \sum_{i=1}^{n} \times$ $\frac{\{Mi(-)Ii\}}{(Si–li)} \times 100$		
Heavy metal evaluation index (HEI)	HEI = $\sum_{i=1}^{n} \frac{H_c}{H_{\text{mac}}} H_{\text{mac}}$	Here, $H_c$ = monitored value, and $H_{\text{mac}}$ = maximum admissible concentration of $ith$ parameters	Edet and Offiong (2002), Bodrud-Doza et al. (2019a, 2019 <sub>b</sub>
The degree of	the degree of $C_d = \sum_{i=1}^{n} C_{fi}$ where, $(C_d)$ $C_{fi} = \frac{C_{ai}}{C_{mi}} C_{ni}$	Here, $C_{fi}$ = contamination factor, $C_{ai}$ = analytical value, and $C_{ni}$ = upper permissible concentration for the <i>i</i> <sup>th</sup> component, and $n =$ normative value. $C_{ni} =$ maximum admissible concentration of heavy metal in water	Backman et al. (1997), Islam et al. $(2016)$ , Bodrud-Doza et al. (2019a, 2019b).

<span id="page-5-0"></span>Table 2 Pollution evaluation indices used to determine mine water pollution status

[2007\)](#page-16-0). It is also minimizing the contribution of less significant variables generated from PCA (Shrestha and Kazama [2007](#page-16-0); Bodrud-Doza et al. [2020\)](#page-15-0). CA was used to describe groups of samples with similar heavy metal contents and to provide supportive information to the results attained from PCA. CA was calculated by following the Ward-algorithmic method, and a squared Euclidean distance was considered to find out the distance among clusters of similar metals (Lattin et al. [2003;](#page-16-0) Bodrud-Doza et al. [2019b](#page-15-0), [2020](#page-15-0)). Furthermore, PCM was applied to guide the outcomes acquired from PCA and to determine the constituent variances that can be described by its inter-relationship with others (Liu et al. [2003\)](#page-16-0).

### Human health risk assessment

Health risk evaluation is the process of assessing the likelihood of the occurrence of any known degree of detrimental health consequences over a specified period of time (Bortey-Sam et al. [2015](#page-15-0); Giri and Singh [2015](#page-15-0); Bodrud-Doza et al. [2020\)](#page-15-0). This evaluation is generally performed based on the risk level determination and expressed by carcinogenic and non-carcinogenic health risks (USEPA [2009](#page-16-0)). However, oral and dermal exposure routes were considered for the assessment, and the chronic daily intake (CDI) of metals was calculated by the following Eq. 1 and Eq. 2 (USEPA [1989;](#page-16-0) Wu et al. [2009](#page-17-0)).

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

$$
CDI_{\text{Dermal}} = \frac{(CW \times SA \times K_p \times ET \times EF \times ED \times CF)}{(BW \times AT)} \tag{2}
$$

where  $CDI<sub>Oral</sub>$  and  $CDI<sub>Dermal</sub>$  indicate the exposure dose (mg/kg/day) through oral ingestion and dermal absorption pathway, which is computed using the value of the parameter presented in Table [3](#page-6-0).

To appraise the non-carcinogenic or chronic health risk of metals, hazard quotient (HQ) were calculated (Eq. 3) for both oral and dermal pathway (USEPA [1989\)](#page-16-0). When the value of  $HO > 1$ , there is an unacceptable risk of adverse noncarcinogenic effects on health, while if  $HQ < 1$ , it was considered an acceptable level (USEPA [2001](#page-16-0)).

$$
HQ = \frac{CDI}{RfD}
$$
 (3)

where RfD is the reference dose (μg/Kg/day) of a specific metal or metalloids considered from the risk-based concentration table (USEPA [2001](#page-16-0)), provided in the supplementary information of Table S1.

To determine the sum of the potential for chronic effects that occurred from multiple elements, are known as hazard index (HI) (USEPA [1989\)](#page-16-0), which is calculated using Eq. 4.

$$
HI = HQ1 + HQ2 + \dots + HQn
$$
 (4)

where HI < 1 means it is expected that the dose is chronic risks free, and if it is  $H1 > 1$ , it indicates the non-carcinogenic or chronic health risk (USEPA [1989](#page-16-0)).

# Results and discussions

#### Quality of in situ mine water

The summary of both physicochemical properties and trace metals of the mine water samples with standard limits is presented in Table [4](#page-7-0). It is found that among the physicochemical parameters, the values of temperature (temp.), BOD, and COD are relatively higher than the recommended level of DoE [\(1997\)](#page-15-0) and FAO [\(1972](#page-15-0)) for the irrigation water standard,

<span id="page-6-0"></span>Table 3 Parameters for exposure assessment of metals through oral ingestion and dermal absorption pathway

Parameters	Unit	Oral values	Dermal values	Reference
CDI (chronic daily intake)	$\mu$ g/kg/day –			Karim (2011); Yang et al. (2012)
CW (conc. of trace metal in water)	$\mu$ g/L			Study data
IR (ingestion rate)	L/day	$2.2$ (adult) 1 (children)		Wu et al. (2009); Karim (2011); Giri and Singh (2015); Bodrud-Doza et al. 2019a)
EF (exposure frequency)	Days/year 365		350	USEPA (2004); Karim (2011); Bodrud-Doza et al. (2020)
ED (exposure duration)	Year	$70$ (adult) 10 (children)	30 (adult) 6 (children)	USEPA (2004); Wu et al. (2009); Karim (2011); Bortey-Sam et al. (2015)
ET (exposure time)	h/event		$0.58$ (adult) $1.0$ (children)	<b>USEPA (2004)</b>
BW (body weight)	Kg	$70$ (adult) 15 (children)	$70$ (adult) 15 (children)	USEPA (2004); Wu et al. (2009); Karim $(2011)$ ; Giri and Singh (2015)
AT (average time)	Days	25,550 (adult) 3650 (children)	Noncarcinogenic effects = $ED \times 365 = 10,950$ Wu et al. (2009); USEPA (2004); (adult), 2190 (children) Carcinogenic effects $AT = 70 \times 365 = 25,550$	Karim (2011); Wongsasuluk et al. $(2014)$ ; Bodrud-Doza et al. 2019 <sub>b</sub>
SA (skin-surface area)	$\text{cm}^2$		18,000 (adult) 6600 (children)	<b>USEPA (2004)</b>
$K_n$ (permeability coefficient)	cm/h		0.001 (Mn), 0.001 (V), 0.001 (Fe), 0.0004 $(C0)$ , $0.0002$ (Ni), $0.001$ (Cu), $0.0006$ (Zn), $0.001$ (Sr), $0.0001$ (Pb)	<b>USEPA (2004)</b>
CF (conversion factor)	L/cm <sup>3</sup>		1/1000	Wu et al. (2009); Karim (2011); Yang et al. (2012)

and almost 100% of the samples exceeded the standard limit. The mean concentration of DO, BOD, and COD was found 8.85 mg/L (7.13–10.45 mg/L), 20.89 mg/L (15.36–26.70 mg/ L), and 229.63 mg/L (186–280 mg/L) respectively (Table [4\)](#page-7-0). The pH of the samples was varied from 7.0 to 8.60, with a mean value of 7.70 indicating neutral to slightly alkaline in nature. It was found that nearly 6.67% of samples exceed the standard limit of pH (6.50–8.50) of BMAL. Conductivity is one of the key parameters of water to determine the suitability for drinking and agricultural use (Islam et al. [2017;](#page-16-0) Bodrud-Doza et al. [2020\)](#page-15-0). EC of the water samples ranges from 240 to 840  $\mu$ S/cm with a mean of 495  $\mu$ S/cm. Though the mean value lies within the standard, around 23.34% of samples exceed the limit (Table [4](#page-7-0)). Furthermore, total coliform and DO levels are found within permissible limits. Moreover, the mean concentration of trace metals in water samples is found to exceed the values of natural surface water: SW1 and SW2 (reference values were taken from Bhuiyan et al. [2010a](#page-15-0)). It is found that trace element contents in the studied water samples followed the order of  $Ca > S > K > Fe > Ti > Co > Br > Pb >$  $Zn > V > Ni > Mn > Sr > Rb > Cu$ . Except for Ca, the mean concentration of the measured parameters lies above the recommended limits (Table [4\)](#page-7-0). Mn, V, Fe, Co, Ni, Cu, and Pb concentration is found comparatively higher, and 76.67, 66.67, 70, 100, 76.67, 66.67, and 80% samples exceed the standard of BMAL respectively (Table [4\)](#page-7-0), which may pose serious ecological and human health hazards.

#### Classification and chemical evaluation of mine water

Ficklin diagram (the sum of metals vs. pH) (Ficklin et al. [1992;](#page-15-0) Caboi et al. [1999\)](#page-15-0) was employed in this study to determine the classification of mine water (Fig. [3\)](#page-8-0). Generally, this classification is adopted through total metal loads vs. pH. The metal load of the water samples was calculated by summing up the analyzed metals, i.e.,  $Fe + Mn + Co + Cu + Zn + Ti +$  $Ni + Sr + Pb$  (mg/L). Pyrite and other sulfide mineral oxidation resulted in acid solutions, where heavy metals were highly mobile. From Fig. [3,](#page-8-0) it was shown that coal seam-leached mine water was felt within the field of near neutral-low metal.

However, the oxidation process occurs in minerals of reduced state in mine, when they come into the open air with mine discharge. A similar chemical reaction occurs when minerals from a reducing environment are carried to the surface and deposited in spoil tips (Wiggering [1993](#page-16-0)). Oxidation of sulphides (type  $MS_2$ , where  $M = Mn$ , Fe, Co, Ni, Cu, Zn)

<span id="page-7-0"></span>Table 4 Descriptive statistics of measured parameters of the Brapukuria coal mine water samples and their comparison with standard limit

Parameters	Descriptive statistics $(n = 30)$							Standards: BMAL, <sup>a</sup> FAO % of samples exceed standards	
	Minimum	Maximum	Mean	Std. deviation	<b>Skewness</b>	Kurtosis			
Temp. $(^{\circ}C)$	35.50	49.00	43.83	4.94	$-0.51$	$-1.38$	30	$100\%$ (30)	
TC	0.00	2.00	0.70	0.79	0.61	$-1.12$	1000	WSL	
pH	7.00	8.60	7.70	0.54	0.42	$-1.47$	$6.50 - 8.50$	$6.67\%$ (2)	
$EC$ ( $\mu$ S/cm)	240.00	840.00	495.00	200.75	0.35	$-1.48$	$700.0^{\rm a}$	23.34\% $(7)^a$	
DO(mg/L)	7.13	10.45	8.85	0.87	$-0.03$	$-0.75$	Min. 6.0	WSL	
$BOD$ (mg/L)	15.36	26.70	20.89	3.19	0.29	$-0.97$	0.20	$100\%$ (30)	
$COD$ (mg/L)	186.00	280.00	229.63	26.85	0.14	$-1.00$	8.0	$100\%$ (30)	
S(mg/L)	8.95	20.80	13.44	3.38	0.42	$-0.98$	$\overline{\phantom{0}}$	NA	
$K$ (mg/L)	2.30	16.35	8.41	3.64	0.37	$-0.18$	12.0	$16.67\%$ (5)	
$Ca \, (mg/L)$	11.00	68.00	20.91	9.61	4.24	21.27	75.0	WSL	
Ti (mg/L)	0.13	0.98	0.43	0.25	0.37	$-0.65$	$\qquad \qquad -$	<b>NA</b>	
$Mn$ (mg/L)	0.05	0.40	0.17	0.10	0.56	$-0.26$	$0.1, 0.2^a$	$76.67\%$ (23), 36.67\% $(11)^a$	
$V$ (mg/L)	0.09	0.54	0.23	0.12	0.71	0.18	0.1 <sup>a</sup>	$66.67\%$ $(20)^a$	
Fe $(mg/L)$	0.04	2.90	1.35	0.82	0.26	$-0.85$	$0.3 - 1.0$ , $5^{\text{a}}$	$70\% (21)$	
$Co$ (mg/L)	0.07	0.71	0.39	0.20	$-0.39$	$-1.07$	$0.05^{\text{a}}$	$100\%$ (30) <sup>a</sup>	
$Ni$ (mg/L)	0.03	0.54	0.17	0.14	1.72	2.90	$0.1, 0.2^{\rm a}$	76.67 (23), 23.34\% $(7)^a$	
$Cu$ (mg/L)	0.03	0.27	0.12	0.06	0.57	0.89	$0.1, 0.2^{\rm a}$	$66.67\%$ (20), 13.34 (4) <sup>a</sup>	
$Zn$ (mg/L)	0.03	0.60	0.27	0.13	$-0.14$	0.31	$5.0, 2.0^a$	WSL	
Br(mg/L)	0.04	1.42	0.24	0.40	2.72	6.01	$\qquad \qquad -$	NA	
$Rb$ (mg/L)	0.05	0.23	0.09	0.06	1.06	$-0.29$	$\overline{\phantom{0}}$	<b>NA</b>	
$Sr \, (mg/L)$	0.06	0.27	0.14	0.05	0.70	0.49		<b>NA</b>	
$Pb$ (mg/L)	0.01	0.50	0.23	0.18	0.34	$-1.19$	$0.05, 5^a$	$80\%$ (24)	

Here, BMAL means Bangladesh (Govt.) maximum admissible limit (DoE [1997](#page-15-0)), <sup>a</sup> FAO [\(1972\)](#page-15-0) standard for irrigation water, WSL means within standard limit, and NA means not applicable

leads to produce protons, rather than consumption (e.g., acid) in geochemical weathering processes (Temmerman et al. [1993;](#page-16-0) Li and Wu [2017\)](#page-16-0). In coal seams, pyrite is a common mineral in most of the metal sulphide and may occur as a potential associating with other chalcophile elements (Bhuiyan et al. [2010a](#page-15-0); Zakir et al. [2017;](#page-17-0) Dong et al. [2020\)](#page-15-0). Though pyrite experiences a complex cycle of reactions during the oxidation process, however, in a simplified form, the following equations (Eqs. 5 to 9) illustrate the net processes;

$$
2FeS_2 + 2H_2O + 7O_2 = 2Fe^{2+} + 4SO_4^{2-} + 4H^+_{(aq)}
$$
 (5)

Some protons consume during the partial oxidation process from ferrous (Fe<sup>2+</sup>) to ferric (Fe<sup>3+</sup>) ions;

$$
4Fe^{2+} + 4H^{+}{}_{(aq)} + O_2 = 4Fe^{3+} + 2H_2O
$$
 (6)

For further pyrite oxidation or hydrolysis process,  $Fe<sup>3+</sup>$ performs like an electron acceptor, where extra protons are released from both processes as;

$$
FeS_2 + 14Fe^{3+} + 8H_2O = 15Fe^{2+} + 2SO_4^{2-} + 16H^+_{(aq)} (7)
$$

$$
Fe^{3+} + 3H_2O = Fe(OH)_3 + 3H^+_{(aq)}
$$
 (8)

In general, a series of reactions is acid-producing;

$$
4FeS_2 + 14H_2O + 15O_2 = 4Fe (OH)_3 + 8SO_4^{2-} + 16H^+_{(aq)}(9)
$$

In hostile acidic mine water, clay minerals sometimes go under reaction with acidic water and offer a certain neutralizing capacity. For instance,

$$
Al_2Si_2O_5(OH)_4 + 6H^+ = 2Al^{3+} + 2H_4SiO_4 + H_2O \quad (10)
$$

However, metal sulfides usually do not produce acidity; nonetheless, they would set free soluble metal ions into the solution. Sphalerite (ZnS) may discharge Zn by oxidization reaction given below (Eq. 7) (Bhuiyan et al. [2010a](#page-15-0)):

$$
ZnS(s) + 2O_2(aq) \rightarrow Zn^{2+} + SO_4{}^{2-}
$$
 (11)

Likewise, siderite, rutile and anatase, galena, melnikovite, and mispickel, strontianite, and zircon released Mn, Ti, Pb, As, Sr, and Zr respectively (Hower and Robertson [2003;](#page-15-0)

<span id="page-8-0"></span>Fig. 3 Ficklin diagram of the sum of metals vs. pH, showing the mine water classification



Sakurovs et al. [2007;](#page-16-0) Bhuiyan et al. [2010a](#page-15-0); Dong et al. [2020\)](#page-15-0). While kaolinite, in general, comprises rutile and anatase as impurities, Ti may substitute for Al in tetrahedral sites (Bhuiyan et al. [2010a](#page-15-0)). During the carbonization process, kaolinite decomposition would free  $TiO<sub>2</sub>$  which shows that kaolinite is considered the main source of  $TiO<sub>2</sub>$ . Generally, siltstone and claystone contain Rb (Fralick and Kronberg [1997\)](#page-15-0). Lead (Pb) is a common association in coal, mainly with sulfides, for example, clausthalite (PbSe), galena (PbS), and pyrite (Hower and Robertson [2003;](#page-15-0) Wang et al. [2003](#page-16-0)). It is reported that Pb may be associated with OM indicates lower rank of coals (Bhuiyan et al. [2010a](#page-15-0)). The activity of acidophilic sulphide-oxidizing bacteria (e.g., Thiobacillus ferrooxidans) flourishes pH range from 1.5 to 3.0, which plays a role in the oxidation of pyrite in mine waters. Through catalyzing the oxidation of ferrous sulphide to ferric sulphate, this acidophilic sulphide-oxidizing bacterium importantly hastens the slow oxidation of iron sulphide (Banks et al. [1997](#page-15-0); Bhuiyan et al. [2010a](#page-15-0)).

## Pollution status of mine water

The results of pollution indices of mine water to determine the usability of drinking, irrigation, and aquatic lives are presented in Table [5.](#page-9-0) The HPI was applied in this study to determine the heavy metal pollution level (Mohan et al. [1996;](#page-16-0) Bhuiyan et al. [2010a\)](#page-15-0). Results demonstrated that the mean value of HPI for drinking, agriculture, and aquatic species was 1802.39 (range 112.19–4098.62), 412.55 (range 94.86–725.80), and 2771.25 (range 1021.75–5950.67), respectively. Except for a few samples, all the samples exceeded the critical limit of pollution. In terms of drinking and aquaculture purposes, 100% of samples were above the critical values, whereas only 20% of samples (i.e., CLW1, CLW2, CLW11, CLW12, CLW21, CLW22) are within critical value in terms of irrigation use (Table [5](#page-9-0)).

However, HEI introduced by Edet and Offiong [\(2002\)](#page-15-0) was also demonstrated in this study to reveal a better understanding of the pollution status. The index range extended from 1.85 to 20.50 (mean 9.71), 3.76 to 19.69 (mean 12.73), and 38.09 to 183.56 (mean 86.15) in terms of drinking, agricultural, and aquaculture use (Table [5](#page-9-0)). In the case of drinking, almost 40% of the samples (CLW5-8, CLW10, CLW16-18, and CLW25-28) fall in the medium category, and 6.67% (CLW20, CLW30) falls in the high category. In the case of irrigation use, 26.66% samples (CLW1-2, CLW8-9, CLW11- 12, CLW21-22) fall low, 73.34% medium (CLW3-7, CLW10, CLW13-20, CLW23-30), and no samples falls in high category. Again, it is noticed that almost all the samples (100%) fall in the high category of pollution levels of aquatic lives (Table [5\)](#page-9-0).

The degree of contamination adopted from Backman et al. [\(1997\)](#page-15-0) was applied to assess the metal pollution level in the sampling sites. The mean value of  $C_d$  for drinking, agriculture, and aquaculture was found 6.16 (range 0–16.11), 8.39 (range 0.44–16.60), and 80.28 (range 32.09–177.56), respectively. It is found that almost 73.24%, 80%, and 100% of samples fall in a high degree of contamination in terms of drinking, agri-culture, and aquaculture use, respectively (Table [5](#page-9-0)). The  $C_d$ values for the mine water indicate contamination of the water is so alarming for the aquatic ecosystem and lives. Overall, the trend of pollution in the  $C_d$  method interestingly coincided with the values evaluated from HPI and HEI, hence illustrating the high level of population.

# Source apportionment of pollution and controlling factors

PCM, PCA, and CA were applied in this study to determine the potential sources of pollution. The results of PCM exhibit some significant positive and negative correlations

<span id="page-9-0"></span>Table 5 Results of pollution evaluation indices of mine water to determine the usability of drinking, irrigation, and aquatic lives

Sample ID	HPI			HEI			$C_d$		
	Drinking	Irrigation	Aquatic lives	Drinking	Irrigation	Aquatic lives	Drinking	Irrigation	Aquatic lives
CLW 1	113.53	97.84	1283.41	4.32	4.33	42.89	1.66	0.44	36.89
CLW <sub>2</sub>	112.19	94.86	1925.94	1.85	3.76	47.98	0.00	0.44	42.02
CLW <sub>3</sub>	1532.39	609.24	1268.22	6.05	16.39	42.93	3.50	13.20	38.32
CLW <sub>4</sub>	807.80	587.45	2079.92	6.18	17.36	62.35	$2.00\,$	13.00	56.35
CLW <sub>5</sub>	2274.01	444.68	3099.32	10.34	13.33	94.32	6.50	8.65	88.32
CLW 6	3516.03	538.35	3811.40	14.61	15.27	118.86	10.60	10.50	112.86
CLW 7	1486.56	470.47	2391.19	10.07	14.39	80.27	5.90	9.50	74.27
CLW 8	2757.91	280.77	2963.18	11.60	9.09	94.88	7.47	5.10	88.88
CLW 9	1292.41	204.03	2048.64	9.22	8.07	68.70	5.10	3.30	62.70
<b>CLW 10</b>	4033.58	510.61	4997.48	19.49	16.46	159.46	15.21	11.00	153.46
<b>CLW 11</b>	113.63	97.84	1153.84	4.41	4.33	40.79	1.76	0.44	34.79
<b>CLW 12</b>	112.22	95.42	2058.94	1.90	3.83	50.98	$0.00\,$	0.44	45.02
CLW 13	1500.18	725.80	1261.50	5.85	19.69	42.13	3.30	16.60	37.52
CLW 14	840.34	635.87	2344.02	5.69	17.50	68.05	$2.00\,$	13.40	62.09
<b>CLW 15</b>	1645.01	463.17	2934.58	8.20	13.98	84.54	4.50	9.30	78.54
<b>CLW 16</b>	3499.75	650.01	4332.32	14.40	18.43	128.43	10.30	13.60	122.43
<b>CLW 17</b>	1502.90	559.78	2525.80	10.38	16.58	83.17	6.20	11.60	77.17
<b>CLW 18</b>	2776.12	373.47	2847.94	13.80	11.82	98.84	9.67	7.15	92.84
<b>CLW 19</b>	1372.32	319.43	2228.82	8.74	11.29	70.89	4.60	6.55	64.89
<b>CLW 20</b>	4066.37	473.59	5668.01	20.25	16.08	174.56	15.91	10.60	168.56
<b>CLW 21</b>	113.84	98.37	1021.75	4.59	4.37	38.09	1.96	0.44	32.09
<b>CLW 22</b>	112.22	95.97	2190.19	1.91	3.88	53.48	$0.00\,$	0.44	47.52
<b>CLW 23</b>	1467.94	485.06	1254.78	5.65	14.59	41.33	3.10	11.60	36.72
<b>CLW 24</b>	841.01	476.34	3148.05	7.32	14.75	85.45	3.05	9.70	79.45
<b>CLW 25</b>	2325.27	518.21	3392.29	13.35	17.29	105.66	9.40	12.20	99.66
<b>CLW 26</b>	3532.04	650.01	3946.01	14.62	18.42	121.76	10.60	13.60	115.76
<b>CLW 27</b>	1518.81	560.33	2531.99	10.26	16.60	83.60	6.07	11.60	77.60
<b>CLW 28</b>	2791.50	460.55	2719.02	13.09	13.67	94.28	8.97	9.15	88.28
<b>CLW 29</b>	1309.21	327.53	2328.39	9.73	12.00	72.93	5.59	7.05	66.93
<b>CLW 30</b>	4098.62	475.86	5950.67	20.50	16.36	183.56	16.11	10.80	177.56
Min	112.19	94.86	1021.75	1.85	3.76	38.09	$0.00\,$	0.44	32.09
Max	4098.62	725.80	5950.67	20.50	19.69	183.56	16.11	16.60	177.56
Mean	1802.39	412.55	2771.25	9.71	12.73	86.15	6.16	8.39	80.29

or pairs among the studied parameters (Table [6](#page-10-0)), which may help to better illustrate the association of contaminants for finding the sources. For instance, COD has a strong positive correlation with BOD and a negative correlation with EC. On the other hand, EC has a significantly positive correlation with Ti, Ni, Br, Rb, and Pb. It seems that a significant positive correlation of the elements may appear from a similar origin (Islam et al. [2017;](#page-16-0) Bodrud-Doza et al. [2019b](#page-15-0), [2020](#page-15-0)). Again, Fe and Sr showed a significant positive correlation with S and negatively correlated with Co. Titanium (Ti) showed a significant correlation with Ni, Pb, and Mn, and Mn showed a correlation with Ni and Cu. The significant correlations among the elemental pairs were Fe–Sr, Co–Pb, and Cu–Zn. Also, BOD is negatively correlated with Pb, Cu, Ni, Co, Mn, and COD; V is negatively correlated with Zn, and Co is negatively correlated with Sr. Though Ca has no significant correlation with others, the leaching of Ca and the precipitation of other heavy metals indicated the evidence of chemical weathering of basement rocks due to mining activities (Bhuiyan et al. [2010a](#page-15-0)). Rhodochrosite and siderite perhaps played a crucial role in buffering mine water, as well as the dissolved  $Fe^{2+}$  and  $Mn^{2+}$  concentrations in water (Allison et al. [1991;](#page-15-0) Williams et al. [2002\)](#page-16-0).



<span id="page-10-0"></span>Environ Sci Pollut Res (2021) 28:22105–22122 22115

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\*\*Correlation is significant at the 0.01 level (2-tailed) \*Correlation is significant at the 0.05 level (2-tailed)

\*\*Correlation is significant at the 0.01 level (2-tailed)<br>\*Correlation is significant at the 0.05 level (2-tailed)

To explore the association and their pathways, both Rmode and Q-mode PCA were carried out using the measured parameters of mine water. Besides, varimax rotation was applied to maximize the sum of the variance of the factor coefficients for a better explanation of the possible sources or groups (Howitt and Cramer [2005\)](#page-16-0). The scree plot was used to identify the number of PCs to recognize the analyzed parameters (Fig. 4a). The loading plot of the first three most influential components of PCA was presented in Fig. 4 b. A total of six factors with eigenvalues > 1 were extracted from



R-mode PCA, exhibiting 84.66% of the total variance. In this study, PC1, PC2, PC3, PC4, PC5, and PC6 were explained by 25.58%, 14.07%, 14.01%, 10.94%, 10.36%, and 9.67% of variance respectively (Supplementary Table S2). The results of R-mode PCA were also allied with Q-mode, where it was found that PC1 was positively loaded with EC, Ti, Mn, Ni, Cu, and Pb, which were distributed in the sampling sites of CLW5-6, CLW9-10, CLW15-16, CLW19-20, CLW25-26, and CLW29-30, indicating mainly derived from geogenic evolution, via rock-water interaction (Bhutiani et al. [2016;](#page-15-0)



Ahmed et al. [2019](#page-15-0)). EC was generally originated from the geogenic process like weathering of bedrock materials (e.g., feldspar) in the underground water (Habib et al. [2019](#page-15-0)). Besides, Mn might be derived from weathering and pedogenic processes, whereas Cu, Ni, and Pb are from leaching of minerals and mine wastes (Bhuiyan et al. [2010a;](#page-15-0) Bodrud-Doza et al. [2019b;](#page-15-0) Habib et al. [2019](#page-15-0)). The presence of Pb might also be derived from mining activities like plumbing systems (Linn et al. [2016\)](#page-16-0). Again, PC2 exhibits 14.07% of the total variance and is highly loaded with S, Fe, and Sr, which may abundant from the geogenic origin. It is reported that Fe is one of the abundant elements in the earth's crust and derived from weathering or leaching of mine effluents (Bhutiani et al. [2016;](#page-15-0) Bodrud-Doza et al. [2019b;](#page-15-0) Habib et al. [2019\)](#page-15-0). These elements were found mostly in the sampling areas of CLW7- 9, CLW11, CLW17-19, CLW21, and CLW27-29. PC3 consists of COD, Cu, and Zn, which may be released via chemical weathering of parental rock. A high level of Zn also has arisen from the cation-exchange of the aquifer (Bhuiyan et al. [2010a](#page-15-0); Kumar [2014](#page-16-0)). Very high loadings of these elements were dominated in the sampling sites of CLW1-2, CLW10-12, CLW-14, CLW20-22, CLW24, and CLW30. PC4 exhibits TC, K, and Rb, mostly distributed in CLW1, CLW6, CLW8, CLW11, CLW16, CLW18, CLW21, CLW26, and CLW28. Again, PC5 consists of DO, BOD, and Br, dominating in CLW1-2, CLW9, CLW11-12, CLW19-20, CLW22, and CLW29. Furthermore, PC6 is highly loaded with pH and Ca, which are mostly distributed in CLW1, CLW4-5, CLW15, and CLW24-25, which indicate that under an alkaline environment, metals may release into the mine water (Díaz et al. [2016](#page-15-0)).

Furthermore, hierarchical CA was also performed to visualize the physicochemical and elemental grouping of mine water (Fig. [5\)](#page-13-0). It seems that variables in the same cluster are likely to be originated from the similar origin (Bhuiyan et al. [2010a;](#page-15-0) Ahmed et al. [2019;](#page-15-0) Habib et al. [2019\)](#page-15-0). Among the six clusters, cluster 1 consists of S, Sr, Fe, Ca, and pH; cluster 2 consists of temperature, COD and BOD; cluster 3 includes TC, DO, and K; cluster 4 includes V, Co; cluster 5 consists of EC, Rb, and Br, and cluster 6 includes Cu, Zn, Ti, Ni, Pb, and Mn. Though there exist a few variations among the associations, the overall grouping of elements in CA agrees with PCM and PCA's results.

#### Probable human health risk

Usually, metals or any harmful elements enter the human body via direct ingestion or oral uptake, inhalation by mouth and nose, and dermal absorption through the skin (Park and Zheng [2012;](#page-16-0) Jaishankar et al. [2014](#page-16-0)). Among them, oral ingestion and dermal absorption are the key pathway to enter groundwater into the human body (USEPA [2004](#page-16-0); Wu et al. [2009\)](#page-17-0). Hence, the chronic or non-carcinogenic health risk of water via oral ingestion and dermal absorption was explored in this study using the model suggested by USEPA [\(1989\)](#page-16-0). Health risk assessment of mine water for adults and children was assessed by using hazard quotients (HQ), and hazard index (HI) and summarized in Table [7.](#page-13-0) It is suspected from the results that the health risk of adults and children may occur greatly by oral ingestion of contaminated mine water, then dermal contact, and children are at high risk (Fig. [6](#page-14-0)).

It is found that HQ<sub>oral</sub> values for Mn, Fe, Ni, Cu, Zn, and Sr were found less than 1 for adults and Fe, Cu, Zn, and Sr for children (Table [7\)](#page-13-0), indicating these elements could pose low health hazards (Giri and Singh [2015](#page-15-0); Bodrud-Doza et al. [2020\)](#page-15-0). Overall, V, Co, and Pb were found as the most significant pollutants leading to non-carcinogenic worries in the case of oral ingestion. For adults, mean  $HQ_{\text{oral}}$  for V, Co, and Pb were 1.46, 0.61, and 2.11, with the range of 0.58– 3.39, 0.11–1.12, and 0.09–4.49, respectively (Table [7\)](#page-13-0). It is found that about 66.67%, 13.34%, and 70% of samples were contaminated via V, and Pb, which are above the safe limit (Supplementary Table S3). Moreover, for children, Mn, V, Co, Ni, and Pb were found as the main metals for  $HQ_{\text{oral}}$ . The mean values of Mn, V, Co, Ni and Pb were 0.59, 3.1, 1.29, 0.59, and 4.49 with a range of 0.16–1.13, 1.23–7.20, 0.24–2.37, 0.09–1.80, and 0.19–9.52, respectively (Table [7\)](#page-13-0). Among the samples, 16.67% (Mn), 100% (V), 70% (Co), 10% (Ni), and 80% (Pb) were placed above the threshold of concern for adverse health effects (Supplementary Table S3). Again, in the case of dermal absorption except for V all other elements  $HQ_{\text{dermal}}$ , value is found less than 1. The immense risks induced due to dermal contact (HQ<sub>dermal</sub>) for the adults and children were 0.47 (0.18–1.103) and 1.40 (0.55–3.25), respectively (Table [7](#page-13-0)). Around 3.34% and 70% of samples exceed the safe limits of V in the case of dermal exposure of adults and children, respectively (Supplementary Table S4).

Considering the concentration of the average metal, 100% of the sample comprises a HI value greater than 1 for both adults and children (Supplementary Table S3). Among the samples, the HI ranged from 1.13 to 7.93 and 2.39 to 16.80 for adult and child respectively, suggesting that metals posed a hazard by oral intake recommending the mine-induced groundwater inflow. According to the chronic risk classification of USEPA ([1989](#page-16-0)), for oral ingestion, almost 76.67% and 80% of samples fall in the high category and the rest 23.33, and 20% fall in the medium category of risk for adult and children, respectively. Again, in the case of dermal adsorption, V was identified as the only dominating metal for hazard. For adults and children, the results observed that 3.33% and 70%, respectively, of the samples, exceeded the safe limits (Supplementary Table S4). It is found that children in the study area are the main victim of dermal absorption of hazardous elements, where nearly 70% of samples fall in the medium category of risk (USEPA [1989\)](#page-16-0). However, for children, the results of oral HI raised concerns about non<span id="page-13-0"></span>Fig. 5 Dendrogram showing the hierarchical clusters (R-mode) of analyzed parameters of mine water



Table 7 Summary of the chronic health risk (HQ and HI) of contaminants in mine water through the oral and dermal routes for adults and children

Health risk index	Elements	Oral			Dermal			
		Min.	Max.	Mean	Min.	Max.	Mean	
			Adult					
HQ	Mn	0.08	0.63	0.28	0.009	0.072	0.031	
	V	0.58	3.39	1.46	0.188	1.103	0.476	
	Fe	0.01	0.30	0.14	1.00E-04	0.009	0.004	
	Co	0.11	1.12	0.61	3.00E-04	0.003	0.001	
	Ni	0.05	0.85	0.28	2.00E-04	0.003	9.00E-04	
H	Cu	0.02	0.21	0.10	3.00E-04	0.003	0.001	
	Zn	0.003	0.06	0.03	4.00E-05	9.00E-04	4.00E-04	
	<b>Sr</b>	0.003	0.014	0.007	7.00E-05	3.00E-04	2.00E-04	
	Pb	0.09	4.49	2.11	3.00E-04	0.014	0.006	
	H	1.13	7.93	4.92	0.202	1.12	0.52	
			Children					
HQ	Mn	0.16	1.33	0.59	0.0253	0.211	0.0927	
	V	1.23	7.20	3.10	0.555	3.255	1.403	
	Fe	0.01	0.64	0.30	4.00E-04	0.027	0.013	
	Co	0.24	2.37	1.29	8.00E-04	0.007	0.004	
	Ni	0.09	1.80	0.59	5.00E-04	0.008	0.003	
H1	Cu	0.05	0.45	0.21	9.00E-04	0.009	0.004	
	Zn	0.01	0.13	0.06	1.00E-04	0.003	0.001	
	Sr	0.01	0.03	0.02	1.00E-04	6.00E-04	3.00E-04	
	Pb	0.19	9.52	4.49	8.00E-04	0.04	0.019	
	HI	2.39	16.80	10.40	0.595	3.304	1.533	

<span id="page-14-0"></span>Fig. 6 Results of chronic health risk index (HI) for adults and children via oral and dermal exposure



carcinogenic adverse health effects of drinking water if we consider the additive impact of harmful metals. Children are vulnerable to acute, sub-acute, and chronic effects of the ingestion of chemical pollutants (ENHIS [2007\)](#page-15-0) because they usually consume more water comparing to their per unit of body weight than adults. Gržetić and Ghariani [\(2008](#page-15-0)) found that HQ value was 10-folds higher for children comparing the corresponding results for adults. However, it was shown that the ingestion was the main exposure pathway of heavy metals into the human body, following by dermal absorption.

# **Conclusions**

Accumulation, sources, and public health risks associated with metal contamination in in situ mine water were studied in the Barapukuria coal mine industry in Bangladesh. It is found that, among the measured parameters, concentrations of pH, temperature, EC, BOD, COD, K, Mn, V, Fe, Ci, Co, Ni, and Pb exceeded the national and international standard limits in different proportions. The results obtained from pollution evaluation indices suggest that water is contaminated with metal at a certain level. For instance, HPI indicates that almost 100% of samples exceed the critical limit for the use of drinking and aquaculture activities, and 80% for irrigation purpose. The multivariate analysis apportions the sources of the metal in water mostly from geogenic sources (e.g., weathering of parent materials, leaching of minerals) along with certain anthropogenic influences (e.g., mine effluents, mine dust). However, the chronic health risk of metals via oral and dermal exposure of adults and children were further analyzed and found V, Co, and Pb as the most significant pollutants of health risk for adults and Mn, V, Co, Cu, and Pb for children. Around  $66\%$  (V),  $13\%$  (Co), and  $70\%$  (Pb) samples exceed the safe limit of  $HQ_{\text{oral}}$  value for adult, whereas  $17\%$ (Mn), 100% (V), 70% (Co), 10% (Ni), and 80% (Pb) were placed above the threshold limit for children. In the case of HIoral, almost 100% of samples exceed the critical limit, and 77% and 80% of samples fall in the high category of risk for adults and children, respectively. Moreover, for  $HI<sub>dermal</sub>$ , 3% and 70% of samples exceed the safe limit for adults and children, respectively, and nearly 70% of samples fall in the medium category of risk for children. To minimize the health risk of local people, mine water should be treated before use. The findings from this study would contribute immence values to the mine authorities and local government to take necessary actions for the necessary treatment of mine discharge water before drains out to the nearby canals to minimize the health risk and ensure environmental sustainability.

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Author's contribution MAHB, MAR, and BBS have made the concept and study design, collected the samples, performed a laboratory experiment, and prepared the draft manuscript. MB and SMDI performed data analysis and interpretation, and improved the draft manuscript.

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## Compliance with ethical standards

Ethical approval Not applicable.

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