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 (HO) 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_{oral} 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_{dermal} 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; Tiwari et al. 2017; Wright et al. 2018; Habib et al. 2019). Coal mining processes are well-known sources of water pollution because of mine drainage to local waterways (Wright et al. 2017, 2018). Usually, discharged mine water is highly acidic depends on the sulfur content in coal that could generate H₂SO₄ which leaches and mobilizes heavy metals from the surrounding geology (Zhao et al. 2007, Bhuiyan et al. 2010a; Wright et al. 2018). 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, 2010b; Zakir et al. 2013; Hossain et al. 2015; Linn et al. 2016; Howladar et al. 2017; Liao et al. 2017; Gao et al. 2019; Dong et al. 2020).

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; McCullough and Lund 2006; Petritz et al. 2009; Gammons et al. 2009; Howladar et al. 2017). 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; Renz et al. 2009). 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). The Gondwana coal-bearing sedimentary unit (Gondwana formation) is highly saturated with groundwater (Bhuiyan et al. 2010a). 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). 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, 2010b; Zakir et al. 2013; Howladar et al. 2014; Hossain et al. 2015; Sultana et al.

2016; Khan et al. 2017). Hence, the public health of the surrounding area is under threat because of the probable metals transferring into the food chain.

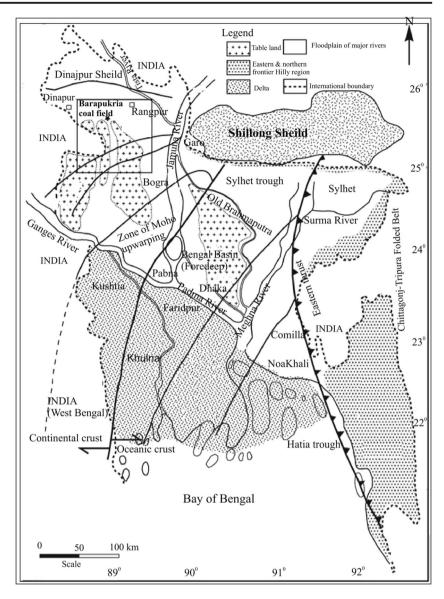
Certain parts of the underground mine subsided and developed fractures and lineaments on the mine roof and surface area. Therefore, surface water along with rainwater may be infiltrated into the lithologic units through these weak points and contaminated the in situ mine water. The infiltrated water 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). The area is relatively flat, considered as major rice-producing (> 60%) zones in Bangladesh (Bhuiyan et al. 2010a). 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). 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

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).

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). 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; Alam et al. 2003). 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). 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; Habib et al. 2019). 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; Howladar et al. 2014).

However, the unconsolidated Dupi Tila Formation is influencing the water flow in seam VI (Table 1), 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		•	
	(a) Upper section	Relatively aquiclude	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 gneiss	31

 Table 1
 Stratigraphy and hydrostratigraphic units of the Barapukuria coal mine area (Bakr et al. 1987; CMC 1994; Khan et al. 2017)

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. 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). 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). 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). The Gondwana sandstones are separated into upper and lower sections by seam VI (Islam and Hayashi 2008). The thickness of the upper section is 156 m, placed at the center part in the basin (Khan et al. 2017). 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; CMC 1994; Howladar et al. 2014). The thickness of the lower unit differs from 107 to 244 m from north-west to the southeastern part.

Materials and methods

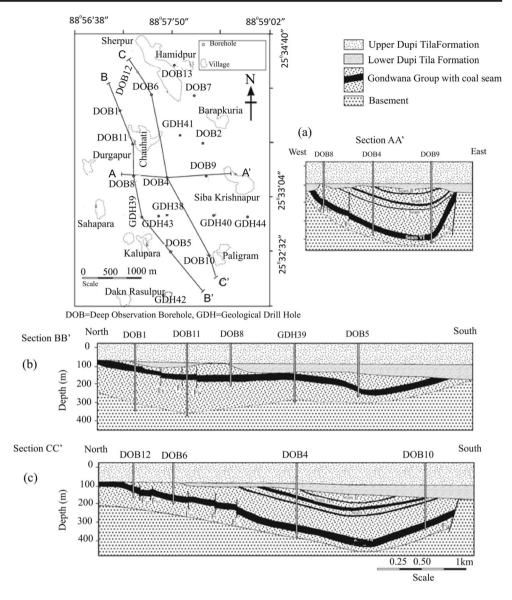
Sampling and analytical procedure

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). Coliform colonies (TC) were identified and counted following the standard procedure (APHA 1995). 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 **Fig. 2** Subsurface geology of Barapukuria coal mine area of Bangladesh



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

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), 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; Shrestha and Kazama

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

Table 2 Pollution evaluation indices used to determine mine water pollution status

2007). It is also minimizing the contribution of less significant variables generated from PCA (Shrestha and Kazama 2007; Bodrud-Doza et al. 2020). 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; Bodrud-Doza et al. 2019b, 2020). 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).

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; Giri and Singh 2015; Bodrud-Doza et al. 2020). This evaluation is generally performed based on the risk level determination and expressed by carcinogenic and non-carcinogenic health risks (USEPA 2009). 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; Wu et al. 2009).

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

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

where CDI_{Oral} and CDI_{Dermal} 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.

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). When the value of HQ > 1, there is an unacceptable risk of adverse non-carcinogenic effects on health, while if HQ < 1, it was considered an acceptable level (USEPA 2001).

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

where RfD is the reference dose (μ g/Kg/day) of a specific metal or metalloids considered from the risk-based concentration table (USEPA 2001), 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), which is calculated using Eq. 4.

$$HI = HQ_1 + HQ_2 + \dots + HQ_n \tag{4}$$

where HI < 1 means it is expected that the dose is chronic risks free, and if it is HI > 1, it indicates the non-carcinogenic or chronic health risk (USEPA 1989).

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. 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) and FAO (1972) for the irrigation water standard,

 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)	µg/kg/day	_	-	Karim (2011); Yang et al. (2012)
CW (conc. of trace metal in water)	μg/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)	USEPA (2004)
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 = $ED \times 365 = 10,950$ (adult), 2190 (children) Carcinogenic effects $AT = 70 \times 365 = 25,550$	Wu et al. (2009); USEPA (2004); Karim (2011); Wongsasuluk et al. (2014); Bodrud-Doza et al. 2019b)
SA (skin-surface area)	cm ²	_	18,000 (adult) 6600 (children)	USEPA (2004)
K_p (permeability coefficient)	cm/h	_	0.001 (Mn), 0.001 (V), 0.001 (Fe), 0.0004 (Co), 0.0002 (Ni), 0.001 (Cu), 0.0006 (Zn), 0.001 (Sr), 0.0001 (Pb)	USEPA (2004)
CF (conversion factor)	L/cm ³	_	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). 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; Bodrud-Doza et al. 2020). EC of the water samples ranges from 240 to 840 μ S/cm with a mean of 495 μ S/cm. Though the mean value lies within the standard, around 23.34% of samples exceed the limit (Table 4). 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). 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). 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), 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; Caboi et al. 1999) was employed in this study to determine the classification of mine water (Fig. 3). 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, 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). Oxidation of sulphides (type MS_2 , where M = Mn, Fe, Co, Ni, Cu, Zn)

 Table 4
 Descriptive statistics of measured parameters of the Brapukuria coal mine water samples and their comparison with standard limit

Parameters	Descriptive	e statistics (n	= 30)				Standards: BMAL, ^a FAO	% of samples exceed standards
	Minimum	Maximum	Mean	Std. deviation	Skewness	Kurtosis		
Temp. (°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
pН	7.00	8.60	7.70	0.54	0.42	-1.47	6.50-8.50	6.67% (2)
EC (µS/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	-	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	-	NA
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 ^a	66.67% (20) ^a
Fe (mg/L)	0.04	2.90	1.35	0.82	0.26	-0.85	0.3–1.0, 5 ^a	70% (21)
Co (mg/L)	0.07	0.71	0.39	0.20	-0.39	-1.07	0.05 ^a	100% (30) ^a
Ni (mg/L)	0.03	0.54	0.17	0.14	1.72	2.90	0.1, 0.2 ^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 ^a	66.67% (20), 13.34 (4) ^a
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	-	NA
Rb (mg/L)	0.05	0.23	0.09	0.06	1.06	-0.29	-	NA
Sr (mg/L)	0.06	0.27	0.14	0.05	0.70	0.49	-	NA
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), ^aFAO (1972) 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; Li and Wu 2017). 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; Zakir et al. 2017; Dong et al. 2020). 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;

$$2\text{FeS}_2 + 2\text{H}_2\text{O} + 7\text{O}_2 = 2\text{Fe}^{2+} + 4\text{SO}_4^{2-} + 4\text{H}^+_{(aq)} \qquad (5)$$

Some protons consume during the partial oxidation process from ferrous (Fe^{2+}) to ferric (Fe^{3+}) ions;

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

For further pyrite oxidation or hydrolysis process, Fe³⁺ 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;

$$4\text{FeS}_2 + 14\text{H}_2\text{O} + 15\text{O}_2 = 4\text{Fe} (\text{OH})_3 + 8\text{SO}_4{}^{2-} + 16\text{H}^+{}_{(\text{aq})} \tag{9}$$

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

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

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

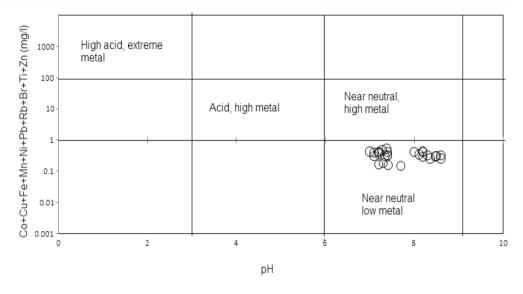


Fig. 3 Ficklin diagram of the sum of metals vs. pH, showing the mine water classification

Sakurovs et al. 2007; Bhuiyan et al. 2010a; Dong et al. 2020). While kaolinite, in general, comprises rutile and anatase as impurities. Ti may substitute for Al in tetrahedral sites (Bhuiyan et al. 2010a). During the carbonization process, kaolinite decomposition would free TiO₂ which shows that kaolinite is considered the main source of TiO₂. Generally, siltstone and claystone contain Rb (Fralick and Kronberg 1997). Lead (Pb) is a common association in coal, mainly with sulfides, for example, clausthalite (PbSe), galena (PbS), and pyrite (Hower and Robertson 2003; Wang et al. 2003). It is reported that Pb may be associated with OM indicates lower rank of coals (Bhuiyan et al. 2010a). 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; Bhuiyan et al. 2010a).

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. The HPI was applied in this study to determine the heavy metal pollution level (Mohan et al. 1996; Bhuiyan et al. 2010a). 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).

However, HEI introduced by Edet and Offiong (2002) 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). 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).

The degree of contamination adopted from Backman et al. (1997) 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, agriculture, and aquaculture use, respectively (Table 5). 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

Table 5 Results of pollution evaluation indices of mine water to determine the usability of drinking, irrigation, and aquatic lives

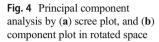
Sample ID	HPI			HEI			C_d		
	Drinking	Irrigation	Aquatic lives	Drinking	Irrigation	Aquatic lives	Drinking	Irrigation	Aquatic lives
CLW 1	113.53	97.84	1283.41	4.32	4.33	42.89	1.66	0.44	36.89
CLW 2	112.19	94.86	1925.94	1.85	3.76	47.98	0.00	0.44	42.02
CLW 3	1532.39	609.24	1268.22	6.05	16.39	42.93	3.50	13.20	38.32
CLW 4	807.80	587.45	2079.92	6.18	17.36	62.35	2.00	13.00	56.35
CLW 5	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
CLW 10	4033.58	510.61	4997.48	19.49	16.46	159.46	15.21	11.00	153.46
CLW 11	113.63	97.84	1153.84	4.41	4.33	40.79	1.76	0.44	34.79
CLW 12	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
CLW 15	1645.01	463.17	2934.58	8.20	13.98	84.54	4.50	9.30	78.54
CLW 16	3499.75	650.01	4332.32	14.40	18.43	128.43	10.30	13.60	122.43
CLW 17	1502.90	559.78	2525.80	10.38	16.58	83.17	6.20	11.60	77.17
CLW 18	2776.12	373.47	2847.94	13.80	11.82	98.84	9.67	7.15	92.84
CLW 19	1372.32	319.43	2228.82	8.74	11.29	70.89	4.60	6.55	64.89
CLW 20	4066.37	473.59	5668.01	20.25	16.08	174.56	15.91	10.60	168.56
CLW 21	113.84	98.37	1021.75	4.59	4.37	38.09	1.96	0.44	32.09
CLW 22	112.22	95.97	2190.19	1.91	3.88	53.48	0.00	0.44	47.52
CLW 23	1467.94	485.06	1254.78	5.65	14.59	41.33	3.10	11.60	36.72
CLW 24	841.01	476.34	3148.05	7.32	14.75	85.45	3.05	9.70	79.45
CLW 25	2325.27	518.21	3392.29	13.35	17.29	105.66	9.40	12.20	99.66
CLW 26	3532.04	650.01	3946.01	14.62	18.42	121.76	10.60	13.60	115.76
CLW 27	1518.81	560.33	2531.99	10.26	16.60	83.60	6.07	11.60	77.60
CLW 28	2791.50	460.55	2719.02	13.09	13.67	94.28	8.97	9.15	88.28
CLW 29	1309.21	327.53	2328.39	9.73	12.00	72.93	5.59	7.05	66.93
CLW 30	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), 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; Bodrud-Doza et al. 2019b, 2020). 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). Rhodochrosite and siderite perhaps played a crucial role in buffering mine water, as well as the dissolved Fe²⁺ and Mn²⁺ concentrations in water (Allison et al. 1991; Williams et al. 2002).

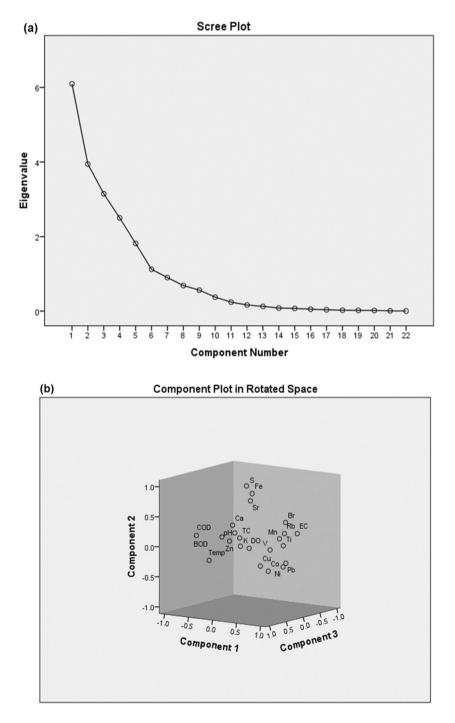
Table 6	Pearson correlations matrix among the elements of mine water collected from BCM of Bangladesh	relations n	natrix amc	ing the elei	ments of m	ine water	collected fi	om BCM	of Bangla	ıdesh										021) 2
Parameters pH	μd	EC	DO	BOD	COD	S	K	Ca	Ti	Mn	Λ	Fe	Co	Ni	Cu	Zn	Br	Rb Sr	r Pb	
μd	1																			1
EC	512**	1																		Z I Z.
DO	-0.075	0.017	1																	2
BOD	-0.298	-0.244	0.313	1																
COD	0.193	666**	0.039	.502**	1															
S	0.231	0.1	0.064	0.173	0.166	1														
K	-0.346	0.191	0.272	0.071	0.305	-0.052	1													
Са	0.337	-0.26	0.279	0.3	0.351	.439*	-0.011	1												
Ti	-0.344	.646**	0.086	-0.332	479**	-0.056	0.165	415* 1	1											
Mn	0.09	.419*	0.006	604**	384*	0.104	0.078	-0.173	.579**	1										
Λ	0.068	-0.046	-0.36	-0.077	-0.239	-0.176	510**		-0.118	- 0.086 1	1									
Fe	0.204	0.149	0.109	-0.168	0.068	.730**	0.143	0.15	0.12	0.254	-0.259	1								
Co	-0.009	-0.002	451*	525**	-0.309	538**	-0.023	365*	0.094		.476**	-0.245	1							
Ni	-0.298	.520**	0.015	528**	601**	367*	0.026	388*	.716**	.653**	-0.101	-0.107	.364*	1						
Cu	0.087	0.223	0.177	560**	$.560^{**} - 0.258$	-0.315	0.161	-0.183	.387*		455*	-0.009	0.053	.657** 1	1					
Zn	0.295	-0.122	0.11	-0.313	0.246 0.122 0.28	0.122	0.28	0.07	0.144	.384*	728** 0.29		-0.265	0.263	.722**	1				
Br	-0.227	.554**	0.287	0.096	508**	0.295	-0.332	-0.022	0.143	0.049	0.277	0.281	-0.25	0.059	-0.131	433*	1			
Rb	386*	.606**	-0.013	-0.153	416*	0.045	.391*	-0.219	0.185	0.145	0.137	0.179	0.224	-0.016 - 0.112	-0.112	372*	.374*	1		
\mathbf{Sr}	.391*	0.191	0.097	-0.008	0.066	.771**	0.111	.447*	0.086	0.229	365*	.515**	501**	-0.207	-0.091	0.293	0.048	0.021 1		
Pb	479**	.615**	-0.25	516**	569**	378*	0.341	456*	.634**	.475**	0.008	-0.161 .573**	.573**	.695** 0.329	0.329	-0.031 - 0.136 .488**	-0.136		-0.18 1	
**Correlation is significant at the 0.01 level (2-tailed)	on is signif	icant at the	e 0.01 lev	el (2-tailed	~															
*Correlation is significant at the 0.05 level (2-tailed)	n is signifio	cant at the	0.05 level	l (2-tailed)																

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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). 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;



Ahmed et al. 2019). EC was generally originated from the geogenic process like weathering of bedrock materials (e.g., feldspar) in the underground water (Habib et al. 2019). 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; Bodrud-Doza et al. 2019b; Habib et al. 2019). The presence of Pb might also be derived from mining activities like plumbing systems (Linn et al. 2016). 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; Bodrud-Doza et al. 2019b; Habib et al. 2019). 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; Kumar 2014). 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).

Furthermore, hierarchical CA was also performed to visualize the physicochemical and elemental grouping of mine water (Fig. 5). It seems that variables in the same cluster are likely to be originated from the similar origin (Bhuiyan et al. 2010a; Ahmed et al. 2019; Habib et al. 2019). 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; Jaishankar et al. 2014). Among them, oral ingestion and dermal absorption are the key pathway to enter groundwater into the human body (USEPA 2004; Wu et al. 2009). 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). 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. 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).

It is found that HQ_{oral} 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), indicating these elements could pose low health hazards (Giri and Singh 2015; Bodrud-Doza et al. 2020). 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_{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). 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_{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). 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_{dermal}, value is found less than 1. The immense risks induced due to dermal contact (HQ_{dermal}) for the adults and children were 0.47 (0.18-1.103) and 1.40 (0.55-3.25), respectively (Table 7). 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), 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). However, for children, the results of oral HI raised concerns about non**Fig. 5** Dendrogram showing the hierarchical clusters (R-mode) of analyzed parameters of mine water

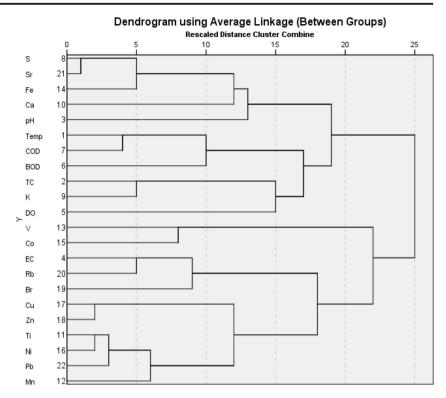
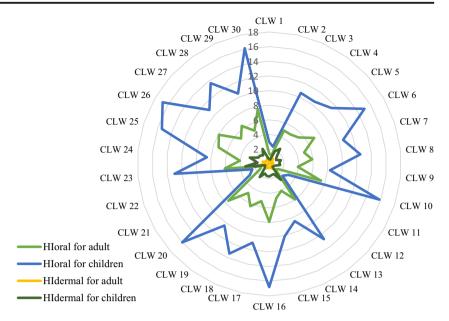


Table 7Summary of the chronichealth risk (HQ and HI) ofcontaminants in mine waterthrough the oral and dermal routesfor adults and children

Health risk index	Elements	Oral			Dermal			
		Min.	Max.	Mean	Min.	Max.	Mean	
			Adul	lt				
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	
HI	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	
	Sr	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	
	HI	1.13	7.93	4.92	0.202	1.12	0.52	
			Childr	en				
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	
HI	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	

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) because they usually consume more water comparing to their per unit of body weight than adults. Gržetić and Ghariani (2008) 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_{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 HI_{oral} 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_{dermal}, 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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References

- Ahmed N, Bodrud-Doza M, Islam SMDU, Choudhry MA, Muhib MI, Zahid A, Hossain S, Moniruzzaman M, Deb N, Bhuiyan MAQ (2019) Hydrogeochemical evaluation and statistical analysis of groundwater of Sylhet, northeastern Bangladesh. Acta Geochim 38(3):440–455
- Alam M, Alam MM, Curray J, Chowdhury RMLR, Gani MR (2003) An overview of the sedimentary geology of the Bengal basin in relation to the regional tectonic framework and basin-fifill history. Sediment Geol 155:179–208
- Allison JD, Brown K, Novo-Gradac (1991) MINTEQA2/PRODEFA2, a geochemical assessment model for environmental systems: version 3.0 user's manual, EPA/600/3–91/021, pp 105
- APHA (American Public Health Association) (1992) Standard methods for the examination of water and wastewater, 18th edn. American Public Health Association, Washington
- APHA (American Public Health Association) (1995) Standard methods for the examination of water and wastewater, 18th ed, Eaton, AD, Clesceri, LS, and Greenberg, AE (eds.), Washington
- Backman B, Bodis D, Lahermo P, Rapant S, Tarvainen (1997) Application of a groundwater contamination index in Finland and Slovakia. Environ Geol 36:55–64
- Bakr MA, Rahman QMA, Islam MM, Islam MK, Uddin MN, Resan SA, Haider MJ, Sultan-Ul-Islam M, Ali MW, Chowdhury MA, Mannan KH, Anam ANMH (1987) Geology and coal deposit of Barapukuria basin, Dinajpur Districts, Bangladesh. Records Geol Survey Bangladesh 8(1):36
- Banglapedia (2016) National Encyclopedia of Bangladesh (2nd.). Asiatic Society of Bangladesh
- Banks D, Younger PL, Arnesen R-T, Iversen ER, Sheila B (1997) Banks, mine-water chemistry: the good, the bad and the ugly. Environ Geol 32(3):157–174
- Bhuiyan MAH, Islam MA, Dampare SB, Parvez L, Suzuki S (2010a) Evaluation of hazardous metal pollution in irrigation and drinking water systems in the vicinity of a coal mine area of northwestern Bangladesh. J Hazard Mater 179:1065–1077
- Bhuiyan MAH, Islam MA, Dampare SB, Parvez L, Suzuki S (2010b) Heavy metal pollution of coal mine-affected agricultural soils in the northern part of Bangladesh. J Hazard Mater 173:383–394
- Bhutiani R, Kulkarni DB, Khanna DR, Gautam A (2016) Water quality, pollution source apportionment and health risk assessment of heavy metals in groundwater of an industrial area in North India. Expo Health 8:3–18
- Bodrud-Doza M, Bhuiyan MAH, Islam SMDU, Quraishi SB, Muhib MI, Rakib MA, Rahman MS (2019a) Delineation of trace metals contamination in groundwater using geostatistical techniques: a study on Dhaka City of Bangladesh 2019a. Groundw Sustain Dev 9: 100212. https://doi.org/10.1016/j.gsd.2019.03.006
- Bodrud-Doza M, Islam SMDU, Hasan MT, Alam F, Haque MM, Rakib MA, Asad MA, Rahman MA (2019b) Groundwater pollution by trace metals and human health risk assessment in central west part of Bangladesh. Groundw Sustain Dev 9:100219. https://doi.org/10. 1016/j.gsd.2019.100219
- Bodrud-Doza M, Islam SMDU, Rume T, Quraishi SB, Rahman MS, Bhuiyan MAH (2020) Groundwater quality and human health risk assessment for safe and sustainable water supply of Dhaka City dwellers in Bangladesh. Groundw Sustain Dev 10:100374. https:// doi.org/10.1016/j.gsd.2020.100374
- Bortey-Sam N, Nakayama SMM, Ikenaka Y, Akoto O, Baidoo E, Mizukawa H, Ishizuka M (2015) Health risk assessment of heavy

metals and metalloid in drinking water from communities near gold mines in Tarkwa, Ghana. Environ Monit Assess 187:397. https://doi.org/10.1007/s10661-015-4630-3

- Caboi R, Cidu R, Fanfani L, Lattanzi P, Zuddas P (1999) Environmental mineralogy and geochemistry of the abandoned Pb-Zn Montevecchio-Ingurtosu mining district, Sardinia, Italy. Chronique de la recherche minière 534:21–28
- CMC (Chinese mining Company) (1994) Preliminary geology and exploration report of Barapukuria coal mine, Bangladesh
- DoE (Department of Environment) (1997) Industrial effluents quality standard for Bangladesh, Bangladesh Gazette Additional August, 28, 1997. In: The environment conservation rules 1997. Government of the People's Republic of Bangladesh, Dhaka
- Díaz SL, Espósito ME, del Carmen BM, Amiotti NM, Schmidt ES, Sequeira ME, Paoloni JD, Nicolli HB (2016) Control factors of the spatial distribution of arsenic and other associated elements in loess soils and waters of the southern Pampa (Argentina). Catena 140:205–216
- Dong S, Feng H, Xia M, Li Y, Wang C, Wang L (2020) Spatial-temporal evolutions of groundwater environment in prairie opencast coal mine area: a case study of Yimin Coal Mine, China. Environ Geochem Health 42:3101–3118. https://doi.org/10.1007/s10653-020-00544-z
- Edet AE, Offiong OE (2002) Evaluation of water quality pollution indices for heavy metal contamination monitoring. A study case from Akpabuyo-Odukpani area, Lower Cross River Basin (southeastern Nigeria). GeoJournal 57:295–304
- ENHIS (European Environment and Health Information System) (2007) Exposure of children to chemical hazards in food. Fact Sheet No. 4.4, CODE: RPG4 Food EX1.World Health Organization
- FAO (1972) Overall Study of the Messara Plain. Report on study of the water resources and their exploitation for irrigation in Eastern Crete. FAO Report No. AGL:SF/GRE/31
- Ficklin DJWH, Plumee GS, Smith KS, McHugh JB (1992) Geochemical classification of mine drainages and natural drainages in mineralized areas. In: Kharaka YK, Maest AS (eds) Water–rock interaction, vol 7. Balkema, Rotterdam, pp 381–384
- Fralick PW, Kronberg BI (1997) Geochemical discrimination of clastic sedimentary rock sources. Sediment Geol 113:111–124
- Gao P, Sun X, Xiao E, Xu Z, Sun W (2019) Characterization of ironmetabolizing communities in soils contaminated by acid mine drainage from an abandoned coal mine in Southwest China. Environ Sci Pollut Res 26:9585–9598
- Gammons CH, Harris LN, Castro JM, Cott PA, Hanna BW (2009) Creating lakes from open pit mines: processes and considerations, with emphasis on northern environments. Can Tech Rep Fish Aquat Sci 2826:106
- Giri S, Singh AK (2015) Human health risk assessment via drinking water pathway due to metal contamination in the groundwater of Subarnarekha River Basin, India. Environ Monit Assess 187:63. https://doi.org/10.1007/s10661-015-4265-4
- Gržetić I, Ghariani RHA (2008) Potential health risk assessment for soil heavy metal contamination in the central zone of Belgrade (Serbia). J Serbian Chem Soci 7:923–934
- Habib MA, Islam ARMT, Bodrud-Doza M, Mukta FA, Khan R, Siddique MAB, Phoungthong K, Techato K (2019) Simultaneous appraisals of pathway and probable health risk associated with trace metals contamination in groundwater from Barapukuria coal basin. Bangladesh, Chemosphere 242:125183. https://doi.org/10.1016/j. chemosphere.2019.125183
- Hossain MN, Paul SK, Hasan MM (2015) Environmental impacts of coal mine and thermal power plant to the surroundings of Barapukuria, Dinajpur, Bangladesh. Environ Monit Assess 187:202. https://doi. org/10.1007/s10661-015-4435-4
- Hower J, Robertson JD (2003) Clausthalite in coal. Int J Coal Geol 53: 219–225

- Howitt D, Cramer D (2005) Introduction to SPSS in psychology: with supplement for releases 10, 11, 12 and 13. Pearson, Harlow
- Howladar MF, Deb PK, Muzemder ATMSH (2017) Monitoring the underground roadway water quantity and quality for irrigation use around the Barapukuria coal mining industry, Dinajpur, Bangladesh. Groundwater Sustain Develop 4:23–34
- Howladar MF, Deb PK, Muzemder ATMSH, Ahmed M (2014) Evaluation of water resources around Barapukuria coal mine industrial area, Dinajpur, Bangladesh. Appl Water Sci 4:203–222
- Islam MR, Hayashi D (2008) Geology and coal bed methane resource potential of the Gondwana Barapukuria Coal Basin, Dinajpur, Bangladesh. Int J Coal Geol 75(3):127–143
- Islam SMD, Bhuiyan MAH, Rume T, Mohinuzzaman M (2016) Assessing heavy metal contamination in the bottom sediments of Shitalakhya River, Bangladesh; using pollution evaluation indices and geo-spatial analysis. Pollution 2(3):299–312
- Islam SMD, Majumder RK, Uddin MJ, Khalil MI, Alam MF (2017) Hydrochemical characteristics and quality assessment of groundwater in Patuakhali district, southern coastal region of Bangladesh. Expo Health 9:43–60
- Jaishankar M, Tseten T, Anbalagan N, Mathew BB, Beeregowda KN (2014) Toxicity, mechanism and health effects of some heavy metals. Interdiscip Toxicol 7(2):60–72
- Karim Z (2011) Risk assessment of dissolved trace metals in drinking water of Karachi, Pakistan. Bull. Environ Contam Toxicol 86:676– 678
- Khan FH (1991) Geology of Bangladesh. University Press Limited, Dhaka
- Khan AA, Chouhan RKS (1996) The crystal dynamics and the tectonic trends in the Bengal basin. J Geodyn 22(3/4):267–286
- Khan MHR, Seddique AA, Rahman A, Shimizu Y (2017) Heavy metals contamination assessment of water and soils in and around Barapukuria coal mine area, Bangladesh. American J Environ Protec 6(4):80–86
- Kumar PJS (2014) Evolution of groundwater chemistry in and around Vaniyambadi Industrial Area: differentiating the natural and anthropogenic sources of contamination. Chem Erde 25298:11
- Lattin J, Carrol, Green P (2003) Analyzing multivariate data. Duxbury Press, Belmont
- Li X, Wu P (2017) Geochemical characteristics of dissolved rare earth elements in acid mine drainage from abandoned high-As coal mining area, southwestern China. Environ. Sci Pollut Res 24:20540– 20555
- Liao J, Ru X, Xie B, Zhang W, Wu H, Wu C, Wei C (2017) Multi-phase distribution and comprehensive ecological risk assessment of heavy metal pollutants in a river affected by acid mine drainage. Ecotoxicol Environ Saf 141:75–84
- Linn M, Peng W, Gui H (2016) Heavy metals in deep groundwater within coal mining area, northern Anhui province, China: concentration, relationship, and source apportionment. Arab J Geosci 9:319. https://doi.org/10.1007/s12517-016-2354-5
- Liu CW, Lin KH, Kuo YM (2003) Application of factor analysis in the assessment of groundwater quality in a Blackfoot disease area in Taiwan. Sci Total Environ 313:77–89
- McCullough CD, Lund MA (2006) Opportunities for sustainable mining pit lakes in Australia. Mine Water Environ 25:220–226
- Mohan SV, Nithila P, Reddy SJ (1996) Estimation of heavy metal in drinking water and development of heavy metal pollution index. J Environ Sci Health A 31:283–289
- Otchere FA, Veiga MM, Hinton JJ, Farias RA, Hamaguchi R (2004) Transforming open mining pits into fish farms: moving towards sustainability. Nat Resource Forum 28:216–223
- Park JD, Zheng W (2012) Human exposure and health effects of inorganic and elemental mercury. J Prev Med Public Health 45(6):344– 352

- Petritz KM, Gammons CH, Nordwick S (2009) Evaluation of the potential for beneficial use of contaminated water in a flooded mine shaft in Butte, Montana. Mine Water Environ 28:264–273
- Prasad B, Bose JM (2001) Evaluation of the heavy metal pollution index for surface and spring water near a limestone mining area of the lower Himalyas. Environ Geol 41:183–188
- Raymond J, Therrien R (2008) Low-temperature geothermal potential of the flooded Gaspe' Mines, Que'bec, Canada. Geothermics 37:189– 210
- Renz A, Rühaak W, Schätzl P, Diersch H-JG (2009) Numerical modeling of geothermal use of mine water: challenges and examples. Mine Water Environ 28:2–14
- Sakurovs R, French D, Grigore M (2007) Quantification of mineral matter in commercial cokes and their parent coals. Int J Coal Geol 72: 81–88
- Shrestha S, Kazama F (2007) Assessment of surface water quality using multivariate statistical techniques: a case study of the Fuji river basin, Japan. Environ Model Softw 22:464–475
- Sultana S, Biswas PK, Rahman A, Sultana S, Zaman MN (2016) Risk factor assessment of coal mine drainage water on surrounding agricultural soil: a case study at Barapukuria in Bangladesh. J Geosci Environ Protec 4:7–17
- Temmerman WM, Durham PJ, Vaughan DJ (1993) The electronic structures of the pyrite-type disulphides (MS₂, where M = Mn, Fe, Co, Ni, Cu, Zn) and the bulk properties of pyrite from local density approximation (LDA) band structure calculations. Phys Chem Miner 20(4):248–254
- Tiwari AK, Singh PK, Mahato MK (2017) Assessment of metal contamination in the mine water of the west Bokaro coalfield, India. Mine Water Environ 36:532–541
- USEPA (1989) Risk assessment guidance for superfund volume I human health evaluation manual (part A)
- USEPA (2004) Risk assessment guidance for superfund volume I: human health evaluation manual (part E). http://www.epa.gov/oswer/ riskassessment/ragse/pdf/introduction.pdf
- USEPA (2009) National primary and secondary drinking water standards. (http://www.epa.gov/safewater/consumer/pdf/mcl.pdf)
- USEPA (US Environmental Protection Agency) (2001) Baseline human health risk assessment Vasquez Boulevard and I-70 superfund site, Denver CO. http://www.epa.gov/region8/superfund/sites/VB-170-Risk.pdf. Accessed 01/20/2011
- Vega M, Pardo R, Barrado E, Deban L (1998) Assessment of seasonal and polluting effects on the quality of river waterby exploratory data analysis. Water Res 32:3581–3592
- Wang J, Sharma A, Tomita A (2003) Determination of the modes of occurrence of trace elements in coal by leaching coal and coal ashes. Ener Fuels 17:29–37
- Wiggering H (1993) Sulfide oxidation—an environmental problem within colliery spoil dumps. Environ Geol 22:99–105
- Williams DJ, Bigham JM, Cravotta CA, Traina SJ, Anderson JE, Lyon G (2002) Assessing mine drainage pH from the color and spectral reflectance of chemical precipitates. Appl Geochem 17:1273–1286
- Wongsasuluk P, Chotpantarat S, Siriwong W, Robson M (2014) Heavy metal contamination and human health risk assessment in drinking water from shallow groundwater wells in an agricultural area in Ubon Ratchathani province, Thailand. Environ Geochem Health 36(1):169–182
- Wright IA, Belmer N, Davies PJ (2017) Coal mine water pollution and ecological impairment of one of Australia's most 'protected' high conservation-value Rivers. Water Air Soil Pollut 228:90. https://doi. org/10.1007/s11270-017-3278-8
- Wright IA, Paciuszkiewicz K, Belmer N (2018) Increased water pollution after closure of Australia's longest operating underground coal mine: a 13-month study of mine drainage, water chemistry and river ecology. Water Air Soil Pollut 229:55. https://doi.org/10.1007/ s11270-018-3718-0

- Wu B, Zhao DY, Jia HY, Zhang Y, Zhang XX, Cheng SP (2009) Preliminary risk assessment of trace metal pollution in surface water from Yangtze River in Nanjing Section, China. Bull. Environ Contam Toxicol 82:405–409
- Yang M, Fei Y, Ju YMZ, Li H (2012) Health risk assessment of groundwater pollution—a case study of typical city in North China Plain. J Earth Sci 23(3):335–348
- Zakir HM, Arafat Y, Islam MM (2017) Assessment of metallic pollution along with geochemical baseline of soils at Barapukuria open coal mine area in Dinajpur, Bangladesh. Asian J Water Environ Pollut 14(4):77–88
- Zakir HM, Islam MM, Arafat MY, Sharmin S (2013) Hydrogeochemistry and quality assessment of waters of an open coal mine area in a developing country: a case study from Barapukuria, Bangladesh. Int J Geosci Res 1(1):20–44
- Zhao F, Cong Z, Sun H, Ren D (2007) The geochemistry of rare earth elements (REE) in acid mine drainage from the Sitai coal mine, Shanxi Province, North China. Int J Coal Geol 70:184–192

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