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# Soil pollution and elemental sources along Barapukuria coal mine, Bangladesh: Implications for eco-environmental and health risk assessment



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# ARTICLE INFO

Keywords: Toxic element Soil contamination Pollution indices Health risk APCS-MLR model

# ABSTRACT

For the first time, different pollution indices and a receptor model have been used to quantify ecoenvironmental and health risk assessments as well as identify the sources of potentially toxic elements in soil along the Barapukuria Coal Mine (BCM). Individual indices include enrichment and contamination factors showing the soil samples are moderately to highly contaminated by arsenic, cobalt, chromium, copper, lead, and zinc and heavily contaminated by sulfur. According to the geo-accumulation index, there is significant pollution with arsenic (1.24  $\pm$  0.38), lead (1.49  $\pm$  0.58), cobalt (1.49  $\pm$  0.58), and sulfur (1.63  $\pm$  0.38). Modified hazard quotient and ecological risk factor values also suggest low to moderate environmental risk hazards from the same elements. The nemerow pollution index, pollution load index, nemerow risk index, ecological risk index, and toxic risk index of soil range from 1.65 to 3.03, 0.82-1.23, 11-26, 77-165, and 6.82-11.76 suggest low toxic risk and moderate pollution, among other synergistic indices. Health risk assessment indicates that iron poses lower cancer risk for children than adults, while both face unacceptable cancer risks from inhaling chromium, cobalt, or arsenic. Principal component and phylogenetic cluster analysis extracted by the multiple linear regression with the absolute principal component score (APCS-MLR) model refer to the fact that manganese, iron, titanium, and nickel have originated from geogenic sources, while coal mine effluents enrich elements like arsenic, chromium, zinc, lead, uranium, sulfur, thorium, and zinc and phosphorous sourced from agriculture. In addition, geogenic and anthropogenic sources, including mine and agriculture activities, could potentially pollute the soil and ecosystem. The findings are crucial for regional and national planners in devising strategies to mitigate potentially toxic element pollution in soil along coal mine areas.

## 1. Introduction

Coal mining, a longstanding process driven by the global demand for industry and electricity, has played a key role as the most abundant fossil fuel. The escalating need for energy, coupled with rapid economic growth, has fueled the expansion of underground

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https://doi.org/10.1016/j.heliyon.2024.e32620

Received 21 April 2024; Received in revised form 26 May 2024; Accepted 6 June 2024

Available online 7 June 2024

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coal mining operations worldwide, notably impacting the environment [1–5]. In this context, along the Barapukuria Coal Mine (BCM) area, coal mining practices pose exceptional environmental risks, among the most hazardous globally [6]. Many eco-environmental threats extend to the surrounding ecosystem along the BCM [7–11]. The extraction of coal through underground mining involves significant activities, including the generation of waste rock and plant residues, similar to open-pit mining. These operations contribute to environmental pollution by releasing heavy and non-heavy toxic metal compounds into the air and water [8,10,12]. The contamination of water sources, a consequence of elevated toxic and heavy metals (HM) concentrations, further extends the environmental impact, affecting areas beyond the immediate vicinity of the mine [13]. Coal mining processes, including mine runoff, are recognized as substantial contributors to soil and water pollution [5,14].

Soil pollution along coal mining areas is a significant environmental concern due to the various pollutants associated with coal extraction and processing [15]. The impact of coal mining on soil quality can be extensive, affecting both the surface and subsurface layers. Common soil pollutants in coal mining areas include HM, metalloids, acidity, and organic contaminants. Coal and associated rocks often contain HM, and mining and coal processing activities can release these metals into the soil. HM can persist in the soil, leading to long-term contamination. They pose risks to both ecosystems and human health integrity (Feng et al., 2020; [16]). The discharged water from underground coal mines used for agriculture, like in the BCM, can be a viable solution for both water management and agricultural needs (Fig. 1). However, using effluent from coal mines raises concerns about increased HM content, which can adversely affect crops and the environment. Coal mining processes often bring HM to the surface or into the water due to the nature of the surrounding geological materials.

The eco-environmental and health risks associated with soil in underground coal mine areas are influenced by various factors, including mining methods used, the geology of the area, the release of contaminants during mining activities, and treatment facilities [3,6,16,17]. Soil contamination in and around coal mine areas poses risks due to the presence of various pollutants. The concentrations of contaminants and exposure pathways affect the environment. Elevated levels of HM in the soil can increase health risks to humans through something direct, such as breathing in dust or consuming contaminated food and water [18,19]. Soil contamination can also lead to the leaching of pollutants into groundwater, affecting drinking water sources [20,21]. The geological formation of the Pleistocene and the Gondwana deposit, saturated with groundwater, are crucial considerations in understanding the impact of mining on the water-bearing sedimentary formation. Soil contamination can negatively affect the diversity and abundance of plant and animal species by disrupting ecological balance. Subsidence, the sinking of the land surface, can occur in areas above underground mines, leading to habitat disruption [7,22].

In the BCM region, the coal mine piles contain a combination of shale, sandy shale, mudstone, and coal. Notably, black shale is identified as a significant source of HM such as arsenic, chromium, copper, cobalt, manganese, nickel, lead, zinc, and non-metallic



Fig. 1. Location map of the study area showing sampling sites along BCM.

elements like sulfur, phosphorus, and so on [8,9,11,12,23]. This environmental concern is compounded through the eradication of native soils and the development of new soils, as acknowledged by FAO [24]. Understanding the potential harm to human health and the ecosystem necessitates a comprehensive investigation into the transport of potentially toxic elements through soil in the continental environment. HM, a prevalent environmental pollutant, can be transmitted to animals and humans through various exposure pathways, including direct contact and the food chain [25,26]. Given the hazardous effects of certain metals on soil, it is imperative to assess the extent of HM pollution in the vicinity of the BCM.

A variety of individual and synergistic indices are used to evaluate pollution, environmental, and health risk assessments of the area. Individual indices have been widely used in several studies to assess the impact of a single pollutant, while a new approach, synergistic indices, consider multiple pollutants and their combined effects, providing a more comprehensive understanding of overall environmental risk [27–30]. The APCS-MLR model has been used for quantitative analyses of probable sources of individual elements in soil [27,28,31,32]. According to Zhang et al. [33], the APCS-MLR model is a useful method for classifying samples and identifying the origins of HM. The model has recently been used for pollution source apportionment in the sediment since it does not rely on past source signatures [32,34].

Effluent water from underground BCM used for irrigation raises ecological and health risks due to the presence of contaminants [8, 9,12]. The simultaneous mine effluent water, fertilizers, and pesticides used for irrigation can lead to complex soil pollution issues, affecting the elemental composition of the soil. However, research on pollution and subsequent ecological and health risks in the vicinity of BCM and combustion have negative effects on the water, soil, and plants in the surrounding area [8,10,23]. A few studies have been carried out to evaluate ecological risk and identify the potentially toxic elements in soil using a receptor model along the BCM. However, there is still a need for an integrated evaluation of pollution indices, source apportionment and source-specific risk of HM in the area. This research aims to determine the elemental composition of soil along the BCM, evaluate ecological and health risks using different individual and synergistic pollution indices, and identify the sources of elements using statistics and a receptor model. The research helps evaluate specific risks, identify the sources of elements, and take mitigation measures depending on the characteristics of the contaminants, the local environment, and the specific agricultural practices in the area.

# 2. Study area

The study area is located in the northwestern part of Bangladesh coordinates ranging between latitude  $88^{\circ}57'$  E to  $88^{\circ}59'$  E and longitude  $25^{\circ}31'$  N to  $25^{\circ}35'$  N (Fig. 1). In 1985, the Geological Survey of Bangladesh drilled seven coal-exploring boreholes. Based on the core study, Jiangsu Coal Geology Company began developing an underground coal mine in June 1996. Commercial coal production began in September 2005 after Barapukuria Coal Mining Company Limited was established. The coalfield covers an approximate area of 6.49  $Km^2$  and exhibits a total thickness of 74.14 m, hosting six groups of coal-bearing seams. Tectonically, the Barapukuria basin falls on the Rangpur saddle of the Indian Platform. Rangpur Saddle is connected to the Indian Shield and Rajmahal Hill to the west, while the Shillong Massif to the east. A set of faults crossed by the NW-SE and N–S directions, as well as NW-SW trending faults, formed smaller elevated ridges (horsts) and subsiding basins (graben and half-graben) that separate the region [35]. The region is separated into smaller elevated ridges and sunk basins, as seen by the regional gravity data [36,37]. The plain terrain in the Recent Alluvium and remnants of Pleistocene Barind Clay cover the Barapukuria basin [36]. The region under consideration falls within the alluvial plain, primarily influenced by the confluence of the Ghirnai and Khorkhori rivers. It includes the Barind Tract of the Pleistocene, the Old Himalayan Floodplain, and the Tista Floodplain of the Recent Age [38].

# 3. Methodology

## 3.1. Sample preparation and analysis

A comprehensive soil sampling process was conducted, resulting in the acquisition of a significant number of samples for analysis. Soil samples were collected systematically and scientifically around the BCM where the mine wastewater was used for irrigation (Fig. 1). To create a representative and homogeneous composite sample, duplicate soil samples were methodically selected and carefully stored in airtight plastic bags, each appropriately labelled for identification. Initially, the collected samples were dried overnight in an oven at 60–70 °C and carefully crushed into powder. Visible roots and shell fragments were methodically removed while making powder. To remove moisture content, the powdered material (<60  $\mu$ m) was dried in the oven overnight again. Following the preparation steps, the powdered material was transferred to aluminum rings (30 mm) and shaped into pellets using two tungsten carbide pellets. The elemental composition of these pellets were determined using energy-dispersive X-ray fluorescence spectrometry in Cartesian geometry (EDXL 300, Rigaku, Japan) at Shinshu University, Japan. The specific elements analyzed included aluminum (Al), arsenic (As), calcium (Ca), chlorine (Cl), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), nickel (Ni), vitrium (V), phosphorus (P), potassium (K), silicon (Si), strontium (Sr), sulfur (S), thorium (Th), titanium (Ti), uranium (U), and zinc (Zn), providing a comprehensive overview of the soil composition of the study area. The relative standard deviation (<3 %) was determined by using standard reference samples before the soil samples were analyzed. Analytical precession was measured for each of the eight samples by performing multiple analyses, obtaining a result of  $\pm 2.8$  %.

#### 3.2. Environmental and ecological risk indices

Ecological and environmental risks of soil have been assessed using both individual and synergistic indices (Supplementary

 Table 1

 Statistical summary of elemental composition of soil along BCM.

Sample ID	Ma	A1	Ci .	D	тс	v	Ca	ті	Fo	Cl	Mn	V	Cr	Co	Ni	Cu	7n	Ac	Sr.	Db	Th	II
Sample ID	IVIG	AI	31	r	15	ĸ	Ga	11	re	G	IVIII	v	GI	CU	INI	Gu	211	пз	51	FD	111	0
	In %									mg/kg												
MIN	0.92	6.47	34.20	0.06	0.07	1.43	0.22	0.44	1.33	58	152	85	74	63	29	41	55	2	48	29	28	3
MAX	1.42	10.20	40.40	0.09	0.55	1.93	0.49	0.64	2.93	181	672	128	128	111	79	78	131	6	86	39	38	8
AVG	1.25	8.28	37.91	0.07	0.20	1.73	0.31	0.55	1.95	98	284	101	99	80	44	56	86	3	63	33	33	6
SD	0.13	1.06	2.09	0.01	0.13	0.16	0.07	0.05	0.48	43	155	12	13	16	14	11	20	1	12	3	3	1
CV (%)	10.6	12.8	5.5	13.1	67.5	9.4	22.5	9.0	24.9	43.2	54.5	12.2	13.1	19.8	32.0	19.4	22.9	33.0	19.6	8.4	7.8	19.2
ASV	1.3	8.04	30.1	0.07	0.14	2.88	2.21	0.46	4.67	180	775	60	92	10	10	28	67	4.5	-	10	12	3.7
BGV	2.09	8.13	27.72	0.11	0.03	2.59	3.63	0.44	5	130	950	135	100	25	75	55	70	1.8	375	13	7.2	1.8
SQGs																						
TEC													26		18	16	123	6		31		
PEC													90		36	197	315	17		91		
SEC													110		75	110	820	33		250		

ASV: Average shale volume [63].

BV: Background value [64].

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SQGs (mg/kg): sediment quality guidelines [42].

TEC: threshold effect concentration; PEC: probable effect concentration; SEC: severe effect concentration.

Table 1). Individual indices, including enrichment factor  $(E_F)$ , contamination factor  $(C_f^i)$ , geo-accumulation index  $(I_{geo})$ , modified

hazard quotient (*mHQ*), and ecological risk factor ( $E_r^i$ ) have been employed to assess the influence of a single pollutant ([39–42]; Müller, 1981; [43]), while synergistic indices, including ecological risk index (ERI), toxic risk index (TRI), pollution load index (PLI), nemerow risk index (NRI), and nemerow pollution index (NPI), are used for considering multiple pollutants and their combined effects ([41,42,44–46]; Ustaoğlu et al., 2022; [47]). The formulas that were used to compute these indices and classify the level of contamination are shown in Supplementary Table 1.

#### 3.3. Health risk assessment

Hazard quotients (HQ) for both oral intake and dermal absorption pathways are commonly used to assess the possible noncarcinogenic effects of HM exposure on human health. The equations used for HQ are given below:

$$HQ_{\text{ingestion}} = \frac{ADD_{\text{ingestion}}}{R_f D_{\text{ingestion}}}$$
$$HQ_{\text{dermal}} = \frac{ADD_{\text{dermal}}}{R_f D_{\text{dermal}}}$$

 $R_f D_{\text{dermal}} = R_f D_{\text{ingestion}} \times AB_{sg}$ 

where  $R_f D_{\text{ingestion}}$  and  $R_f D_{\text{dermal}}$  are single-component reference doses (g/kg/day). The following equation can be used to anticipate the overall probable non-carcinogenic risk (NCR) based on the hazard index (HI).

$$HI = \sum_{i=1}^{n} HQ_{ingestion} + HQ_{dermal}$$

Only if the HQ and HI results are more than 1 (one) the non-carcinogenic health concerns be taken into account carcinogenic risk, on the other hand, has no unit, and is the cumulative likelihood of developing a single cancer for a lifetime due to carcinogenic exposure [48-52]. The carcinogenic risk (*CR*) for each component is calculated using the following equation:

$$CR = ADD \times CSF$$

## 3.4. APCS-MLR model

The APCS-MLR model has been used to identify possible sources and percentages of the contribution of each elemental that would contribute to the total pollution. In this model, absolute principal component scores (APCS) are combined with multiple linear regression (MLR) [27,28,31,33,53]. APCS has been used as an independent variable for MLR analysis [53,54]. MLR could potentially applied to determine the relevant sources that are adding to each metal's concentration ( $C_i$ ) [54,55]:

$$C_j = b_{jo} + \sum_{h=1}^n r_{hj} \times APCS_h$$

where n is the number of sources,  $r_{hj}$  denotes the multiple regression coefficients of source *h* concerning metal *j*, and  $b_{jo}$  denotes a multiple regression constant for metal j;  $APCS_{hj}$  is the scaled value of the rotation *h* for the sample under consideration, and  $r_{hj} \times APCS_{hj}$  is the source has contribution to  $C_j$ . The APCS-MLR model has negative contributions throughout the computation phase. This compromises the accuracy of source apportionment by making it difficult to interpret and analyze the various pollution sources' contributions. To avoid this problem, all contribution rates have been calculated as absolute numbers ([55]; Liu et al., 2020).

# 4. Results and discussion

## 4.1. Elemental composition of soil

The elemental composition of soil samples reveal that, the ranges and average concentration of major elements are (0.92-1.42 %), 1.25 % for Mg; (6.47-10.20), 8.28 % for Al; (34.20-40.40 %), 37.91 % for Si; (0.06-0.09 %), 0.07 % for P; (0.07-0.55 %), 0.20 % for S; (1.43-1.93 %), 1.73 % for K; (0.22-0.49 %), 0.31 % for Ca; (0.44-0.64 %), 0.55 % for Ti; (0.02-0.07 %), 0.03 % for Mn; and (1.33-2.93 %), 1.95 % for Fe (Table 1). However, the range and average concentration of minor/trace elements are (58-181 mg/kg), 98 mg/kg for Cl; (152-672 mg/kg), 284 mg/kg for Mn; (85-128 mg/kg), 101 mg/kg for V; (74-128 mg/kg), and 99 mg/kg for Cr; (63-111 mg/kg), 80 mg/kg for Co; (29-79 mg/kg), 44 mg/kg for Ni; (41-78 mg/kg), 56 mg/kg for Cu; (55-131 mg/kg), 86 mg/kg for Zn, (2-6 mg/kg), 3 mg/kg for As, (48-86 mg/kg), and 63 mg/kg for Sr, (29-39 mg/kg), 33 mg/kg for Pb, (28-38 mg/kg), 33 mg/kg for Th; and (3-8 mg/kg), 6 mg/kg for U (Table 1). Besides, the coefficient of variance (CV) ranges from 8.42 % to 54.28 %, and higher values were shown in U (19.2 %), Cu (19.4 %), Sr (19.6 %), Co (19.8 %), Zn (22.9 %), Fe (24.9 %), Ni (32.0 %), As (33 %), Cl (43.2 \%), Mn (54.3 %), and S (67.5 %). The mean concentrations of Zn, Pb, V, Cu, U, Ni, Co, and Th are 20.10 %, 39.39 %, 40.59 %, 41.07 %, 62.16 %, 72.27 %, 87.50 %, and 175 %, higher than the average shale volume (Table 1). The presence of a higher CV for elements in soil along a coal mine area can be attributed to various factors related to both natural geological processes and anthropogenic activities

associated with coal mining. The underlying geological formations in the vicinity of a coal mine area can be naturally diverse, resulting in variations in the concentration of HM [10,56]. The coal seam has a rich source of humic acid, which absorbs a significant amount of metals like As, Cr, Co, Cu, Mn, Pb, Th, U, and Zn, as well as nonmetals like S, Cl, and P [57,58]. The exposure of rocks and soils during mining activities can accelerate weathering processes in aerobic conditions, leading to the release of HM into the soil [59]. Sulfur is commonly found in organic matter, such as plant material. When plants accumulate in swampy environments and undergo partial decay, sulfur-containing organic compounds become incorporated into the peat, which later transforms into coal [60]. In swampy environments, variations in water salinity can lead to the accumulation of chlorine in the plant material, which is later preserved in coal. The paleoenvironmental conditions during the accumulation of plant material in swampy and brackish environments can influence the concentrations of sulfur, phosphorus, and chlorine in the precursor peat and subsequently in the coal [61,62]. During paddy production, certain elements tend to accumulate or become enriched in the soil, particularly as a result of agricultural practices, crop residues, and nutrient inputs. Phosphorus, K, Zn, and S are essential nutrients that are often supplied to paddy fields through fertilizers.

The amounts of a few HM in the soil samples from the BCM region of this study are compared to those in other coal mines and coalfired power plants across the world in Table 2 [9,10,23]. The average concentrations of Mn, Fe, Pb, and Zn in the present study were much lower than those found in earlier investigations in Bangladeshi coal mines. Since 2006, the effluent from the BCM-based power station and coal mine have both gone down the same drain. The power plant was shut down on July 22, 2018, though, as a result of management issues. This causes decreased levels of HM in the soil samples used in the current investigation. The average concentrations of heavy and potentially toxic elements surrounding the irrigation soils along coal mine areas, such as BCM [9,10]; Tinsukia coal mine, India [65]; Jharia and Beijing coal mine, China [17,66]; and Oltu coal mine, Turkey [67], are comparable and found to be higher than those of coal-based power plants, such as the Nicola Tesla coal-fired power plant in Siberia [68] and Santa Catarina coal-fired power plant, Brazil (Rodriguez-Irur-etagoiena et al., 2015) (Table 2).

#### 4.2. Environmental and ecological risk assessment

The average  $E_F$  of Co, Pb, As, Zn, Cu, Cr, K, and Ni in soil are 8.37, 6.78, 5.02, 3.22, 2.71, 1.80, and 1.50, respectively, whereas the enrichment values of TS, Cl, and P values are 21.01, 2.05, and 1.86, respectively (Table 3). The study shows 100 % K, Mg, P, V, Ni, and 80 % Cr soil samples have small enrichment; 33 % Cu, 27 % Al, 20 % Cl, 60 % Ti, and 60 % Zn have medium enrichment; 7 % Si, 100 % Co, 53 % As, and 100 % Pb have medium to high enrichment; and 80 % Th and 47 % U have high enrichment (Table 3). Since an element's  $E_F$  value is between 0.05 and 1.5, it indicates a natural process—that is, the crustal materials are the only source of metal—while values above 1.5 suggest that certain elements could be hazardous to the environment. Human activities such as mining, industrial processes, or waste disposal lead to high concentrations of specific elements [70,71].

The maximum  $C_f^i$  of Co in the studied soil is 4.44; As is 3.44; Pb is 3.00; Zn is 1.87; Cu is 1.42; Cr is 1.28; Ni is 1.05; whereas S and Cl concentrations are 21.19 and 1.39, respectively (Table 3). The results indicated that 100 % of soil samples have low contamination by Mg, P, K, Ca, V, Mn, Fe, and Sr; 100 % Si, Ti, As, and Pb; 47 % Al, 27 % Cl, 53 % Cr, 7 % Ni, 53 % Cu, and 73 % Zn show moderate contamination; 47 % Co, 100 % Th, and 73 % U of the studied samples show considerable contamination. The moderately contaminated contamination factor indicates that the underground coal mining activities have led to an increased concentration of a specific element in the sampled area. Elements commonly associated with coal mining include HM (e.g., arsenic, lead, zinc, and cobalt), metalloids, and other elements present in coal seams or associated rocks. The results of  $I_{geo}$  indicated that 100 % of As; and 100 % of Pb show uncontaminated to moderately polluted; and 47 % of Co, 73 % of U, and 100 % of Th in samples show moderately polluted (Table 3; Fig. 2). The moderately polluted  $I_{geo}$  value indicates that the underground coal mining activities have led to an increased concentration of a specific element in the sampled area. The element in question could be associated with coal seams, mining processes, or waste materials [3,8].

The  $E_r^i$  of Zn in the studied soil is 0.79–1.89, As is 11.11–33.33, Cu is 3.73–7.09, Ni is 11.36–31.60, Cr is 1.48–2.56, and Pb is 11.15–15.00 (Table 3). The metals are categorized based on the ratios of their mean  $E_r^i$  values to the RI as follows: As > Ni > Pb > Cu > Cr > Zn. The results of  $E_r^i$  of 100 % of soil samples for Cr, Ni, Cu, Zn, As, and Pb showed low potential ecological risk (Er < 40). The

Table 2

An analytical comparison of result of heavy metals (mg/kg) of th	study and other coal mines an	d power plants in Bangladesh and worldwide.
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Location	Cr	Mn	Fe	Ni	Cu	Zn	Pb	Reference
Barapukuria coal mine	$99 \pm 13$	$284 \pm 155$	$19477\pm4838$	$44\pm14$	$56\pm11$	$86\pm20$	$33\pm3$	This Study
Barapukuria coal mine	$85\pm0.8$	$555\pm5$	$35488 \pm 167$	$35\pm0.3$	$33\pm0.5$	$82\pm0.7$	$26 \pm 0.2$	[23]
Barapukuria coal mine	$82\pm38$	$226\pm34$	$18874\pm8724$	$57\pm26$	$32\pm10$	$102\pm5$	ND	[10]
Barapukuria coal mine	$107\pm10$	$1048 \pm 194$	$27774 \pm 9831$	$99\pm 38$	$30\pm13$	$160\pm 56$	$189\pm10$	[9]
Tinsukia coal mine (India)	$112\pm 27$	ND	ND	$88\pm32$	ND	ND	$183\pm70$	[65]
Beijing coal mine (China)	$49\pm15$	$737\pm248$	ND	$31\pm12$	$39\pm34$	$87\pm25$	$55\pm40$	[66]
Jharia coal mine (China)	$43\pm19$	$634\pm126$	$\textbf{39,662} \pm \textbf{8226}$	$64\pm13$	$66 \pm 13.1$	$127\pm21$	$28\pm9$	[17]
Oltu coal mine (Turkey)	$136\pm17$	ND	ND	$60\pm16$	$23\pm10$	$35\pm9$	$31 \pm 12$	[67]
Santa Catarina coal-fired power plant	$12\pm10$	$310\pm170$	$24238~\pm$	$6\pm5$	$20\pm9$	$81\pm37$	$14\pm11$	[69]
(Brazil)			11780					

Table 3Obtained results of individual pollution indices of soil.

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Elements	s EF				CP				I <sub>geo</sub>				Ecological Risk Factor				mHQ			
	Min	Max	Avg	Std	Min	Max	Avg	Std	Min	Max	Avg	Std	Min	Max	Avg	Std	Min	Max	Avg	Std
S	6.51	66.64	21.01	16.34	2.75	21.19	7.59	5.12	0.87	3.82	2.09	0.84	_	-	_	_	_	_	_	-
K	1.15	2.30	1.80	0.41	0.55	0.75	0.67	0.06	-1.44	-1.00	-1.17	0.14	-	-	-	-	-	-	-	-
Mn	0.45	1.35	0.75	0.30	0.16	0.71	0.30	0.16	-3.23	-1.08	-2.49	0.70	-	-	-	-	-	-	-	-
Со	7.48	9.47	8.37	0.61	2.52	4.44	3.22	0.64	0.75	1.57	1.08	0.27	-	-	-	-	-	-	-	-
Cr	1.89	3.22	2.62	0.43	0.74	1.28	0.99	0.13	-1.02	-0.23	-0.62	0.19	1.48	2.56	1.97	0.27	1.69	2.22	1.95	0.13
Ni	1.07	2.05	1.50	0.30	0.39	1.05	0.58	0.19	-1.96	-0.51	-1.43	0.41	11.60	31.60	17.17	5.71	1.29	2.10	1.55	0.23
Cu	1.89	3.85	2.71	0.56	0.75	1.42	1.03	0.20	-1.01	-0.08	-0.57	0.27	3.73	7.09	5.19	1.00	1.61	2.21	1.87	0.18
Zn	2.17	4.56	3.22	0.69	0.79	1.87	1.22	0.28	-0.93	0.32	-0.33	0.32	0.79	1.87	1.23	0.29	0.67	1.03	0.83	0.09
As	2.20	8.74	5.02	1.69	1.11	3.33	1.89	0.62	-0.43	1.15	0.26	0.47	11.11	33.33	18.65	6.39	0.65	1.04	0.80	0.11
Pb	4.86	8.39	6.78	1.23	2.23	3.00	2.54	0.21	0.57	1.00	0.75	0.12	11.15	15.00	12.80	1.02	0.97	1.13	1.04	0.04



Fig. 2. Geoaccumulation index of studied soil sample.

*mHQ* is a measure used in environmental risk assessment to evaluate the potential health risks associated with exposure to multiple contaminants and shows a moderate level of potential health risk associated with exposure to contaminants [72]. The *mHQ* values of Cr are 1.69–2.22, Ni is 1.29–2.10, Cu is 1.61–2.21, Zn is 0.67–1.03, As is 0.65–1.04, and Pb is 0.97–1.13 (Table 3). In this study, 93 % of the *mHQ* values of Zn and As show very low severity of contamination; 87 % of Pb have low severity of contamination; 40 % of Ni and 80 % of Cu have moderate severity of contamination; and 40 % of Cr have considerable severity of contamination.

Synergistic indices in the context of an underground coal mine area typically refer to measure that assess the combined or interactive effects of multiple contaminants on the environment or human health [73,74]. The ERI values of the soil samples range from 77 to 165 for measured metals, revealing that most of the studied samples have low ecological risk. The pollution load index estimated from CF ranges from 0.82 to 1.23, referring to 40 % of the studied samples being highly contaminated metal (PLI >1), where values below 1 (one) refer to no pollution (Fig. 3). The results of the ERI for measured metals (Cr, Ni, Cu, Zn, As, and Pb) reveal that 93 % of soil samples have low ecological risk. The results of TRI for measured metals (Cr, Ni, Cu, Zn, As, and Pb) range from 6.82 to 11.76, revealing that 93 % of the soil samples have a low toxic risk (Fig. 3). The NPI value for As and Pb shows moderate pollution; for Cr, Cu, and Zn, it shows low pollution; and for Ni, it shows a warning line of pollution. The NRI value for measured metals (Cr, Ni, Cu, Zn, As, and Pb) revealed that the soil samples are at low risk. The causes of moderate pollution in synergistic indices along an underground coal mine area can be attributed to a combination of factors related to coal mining activities and the interactions of various contaminants present in the environment. Underground coal mining involves various processes such as excavation, blasting, and transportation, which can contribute to the release of particulate matter, dust, and potentially harmful substances.

#### 4.3. Health risk assessment

In the study area soils were polluted by toxic elements, and people might be exposed to these elements through direct ingestion of foods which is grown in polluted sites, as a result, people in this area face several health problems including inhalation and dermal



Fig. 3. The results of synergistic indices used for environmental risk assessment.

exposure. Numerous techniques have been used to assess the HRA with soil samples, including the calculation of cancer risk by carcinogenic elements, the average daily dose, the non-carcinogenic risk/hazard index for all elements, and counting the daily dose of elements that are entering the human body. The results of  $HQ_{ingestion}$ , and  $HQ_{dermal}$  and HI for soil samples are shown in Suplimentary Tables 2 and 4.

### 4.4. Non-carcinogenic health risk for soil

Supplementary Table 2 provides a summary of the results of *HQ*, and *HI* of HM for soil samples from the adjacent area of the BCM. The elements included in the table are Pb, Cr, Fe, Mn, Zn, Cu, Ni, Co, and As. The values represent the HQ for ingestion, inhalation, and dermal contact, as well as the total HQ for children and adults. The table also includes the minimum, maximum, and mean values for each element and exposure pathway (Supplementary Table 2). The *HQ* values indicate the potential non-carcinogenic risk associated with the exposure to these HM in the soil. The HI values, which are the sum of the HQ values, provide an overall assessment of the non-carcinogenic health risk associated with exposure to multiple HM through various pathways. According to the estimated average HI for adults and children, the following order of HM has been observed: Zn > Co > Mn > Pb > Ni > Cr > Cu > As > Fe. Among the HM, Zn has the highest HQ value for both children and adults, followed by Co. Fe has the lowest*HQ*, and*HI*values for both children and adults. The analyses suggest that children may be more vulnerable than adults due to harmful toxic elements through the consumption of food and inhalation of soil particles. This order is based on the summation of the HQ values for each metal, representing the non-carcinogenic risk associated with the exposure to these HM in the soil of BCM.

# 4.5. Carcinogenic health risk for soil

Table 4 provides a detailed overview of the carcinogenic risks associated with four major health-concerning HM in the soil samples collected from the Barapukuria Coal Mine area in Dinajpur. The HM analyzed include Cr, Ni, Co, and As. The table presents the carcinogenic risks for both adults and children associated with each HM, offering insights into the potential health hazards posed by these contaminants. The values are expressed in scientific notation (E-notation), which represents the number as a coefficient multiplied by 10 raised to a power. The carcinogenic risks are measured in terms of the chance of having cancer over their lifetime due to contact of toxic elements. It is crucial to note that the higher the values, the greater the potential risk. Understanding the implications of these findings requires consideration of established guidelines. According to the United States Environmental Protection Agency (USEPA) standards, values of carcinogenic risk (CR) and total carcinogenic risk (TCR) less than  $1 \times 10^{-6}$  are generally considered critical values. Values between  $1 \times 10^{-6}$  and  $1 \times 10^{-4}$  fall in the acceptable range, while values are provided for both adults and children and adults are as follows: As > Cr > Co > Ni. This indicates that As has the highest mean carcinogenic risk value for both children and adults, followed by Cr, Co, and Ni. The values in Table 4 suggest that the exposure to these HM in the soil samples, may pose a significant carcinogenic risk to both children and adults. The study raises concerns about the potential health risks associated with HM in the soil samples, may pose a significant carcinogenic risk to both children and adults.

# 4.6. Sources of potentially toxic elements in soil using multivariate statistical analysis

The APCS-MLR model provides a comprehensive approach to analyzing and identifying the sources of heavy metals in soil. It combines the strengths of Principal Component Analysis (PCA) and Multiple Linear Regression (MLR) to effectively handle complex

Heavy metals	Carcinogenic risks						
	Adults	Children					
Cr							
Min	6.26E-07	1.11E-06					
Max	1.08E-06	1.92E-06					
Mean	8.35E-07	1.48E-06					
Ni							
Min	4.91E-09	8.70E-09					
Max	1.34E-08	2.37E-08					
Mean	7.37E-09	1.31E-08					
Со							
Min	2.21E-07	1.24E-07					
Max	3.89E-07	2.19E-07					
Mean	2.82E-07	1.59E-07					
As							
Min	3.88E-05	4.18E-06					
Max	1.17E-04	1.25E-05					
Mean	6.60E-05	7.11E-06					

datasets with multiple correlated variables. The APCS-MLR model was used to locate potentially toxic elemental sources in the soil along the study area. The results of the source have been verified by the square correlation coefficients (R<sup>2</sup>) for all metals from the APCS-MLR model, with a value greater than 0.5 (Table 5). The PCs and clusters extracted by the APCS-MLR model allowed the main sources to be identified and allowed to be identified sources using this model (Figs. 4 and 5). The source profile and its contribution to the metal are shown in Figs. 4 and 5. Ni (33.52 %), Mn (32.12 %), Zn (28.37 %), and Fe (26.21 %) loadings were observed highest in the first PC, which accounted for 52.20 % of the total contribution (Table 5). The contribution rate for PC2 was 28.65 % and had the highest loading on K (31.81 %), Th (30.86 %), Al (29.84 %), V (23.96 %), and Cr (26.57 %). The largest loadings of Ca (43.06 %), U (36.74 %), S (33 %), Cl (32.20 %), and Ti (29.73 %) were found in PC3, which had a contribution rate of 9.88 %. With a contribution rate of 6.30 %, the PC4 was loaded on P (35.92 %), Zn (35.76 %), U (34.17 %), Si (31.17 %), and As (28.31 %). PC5 had a contribution rate of 2.977 % and the highest loading of Cu (54.45 %), P (36.88 %), Zn (35.28 %), and Co (25.43 %). Multivariate statistical models such as MLR or PC analyses are typically used to identify and quantify potentially toxic elements in soil samples [76,77]. By examining the statistical relationships, the model can identify sources or factors contributing to the observed patterns in the data. PC1 is considered to be the geogenic origin of preexisting rock; elements of PC2 and PC3 come from both geogenic and mining activities; and PC4 and PC5 are contributors from geogenic and agricultural activity (Fig. 5; Table 5).

Interelement correlations within the elemental composition of soil provide information about the origins and pathways of HM in the geoenvironment. According to the results of the Spearman correlation, there is a substantial positive association between Fe and Cr, Mn, and As. Fe, Ni, Zn, and Sr all have a substantial correlation with Mn. Potassium demonstrated significant negative associations with Zn and Fe when the metal concentrations were considered. Fe, Zn, and Co have a significant positive connection among them (Fig. 4). Additionally, there is a significant positive association between Cr and Mn, Ni, Sr, and Pb. Phylogenetic circular dendrogram analysis extracted using the APCS-MLR model shows five clusters. Mn and Ni made up Cluster 1 (C1); Ti, Cl, Sr, V, Th, K, and S comprised Cluster 2; U and Ca made up Cluster 3; Fe, Co, Al, As, Si, and Pb comprised Cluster 4; and Zn, P, and Cu included PC5. Considering these analyses, elements C1 and C2 would be the natural sources; C3 and C4 came from geogenic and mining sources; and C5 came from agricultural sources (Fig. 4).

The elemental composition and statistical, PC, Spearman correlation and dendrogram cluster analyses of soil refer to the fact that both natural and anthropogenic sources contribute to the elemental composition of the soil. Anthropogenic activity, including mining and agriculture, contributes to the degradation of soil quality. According to the results, Fe, Ni, K, Sr, Al, Cl, Ti, and Mn, were derived from geogenic sources, whereas Cr, Co, Cu, U, V, Zn, As, Pb, Th, and S from coal mine effluents. In addition, P, Zn, and S came from agricultural activity in the area (Fig. 6). Furthermore, individual indices show the enrichment of S, Co, As, Pb, Zn, P, and U refer to the influences of anthropogenic factors. Human activities such as mining and agriculture have significantly polluted the natural resources of the area (Fig. 6).

The use of discharged water from underground coal mines for agriculture raises concerns about increased HM content, which can adversely affect both crops and the environment. Coal mining processes often bring HM to the surface or into the water due to the nature of the surrounding geological materials. Effluents from coal mines contain elevated levels of potentially toxic elements posing a significant risk to soil quality, crop health, and even human health if not properly managed. These can accumulate in the soil over time, making it toxic for plant growth and potentially contaminating the food chain if crops absorb toxic substances. The specific toxic elements and their concentrations in the soil would need to be determined through detailed soil testing and analysis in the BCM area to

Table 5 Mean contribution of sources to each metal extracted using APCS-MLR receptor model.

Parameters	R <sup>2</sup>	PC1	PC2	PC3	PC4	PC5
Al	0.92	18.86	29.84	4.23	20.45	6.97
Si	0.87	24.96	21.88	11.00	31.17	8.33
Р	0.61	19.41	20.12	9.89	35.92	36.88
S	0.86	12.13	27.53	33.00	17.55	23.98
C1	0.97	19.00	25.98	32.3	9.32	3.56
K	0.84	9.34	31.81	16.8	15.41	18.67
Ca	0.8	22.29	8.16	43.06	15.27	25.61
Ti	0.94	25.33	18.89	29.73	14.55	0.63
V	0.55	11.80	23.96	14.07	6.63	11.92
Cr	0.83	19.06	26.57	12.97	5.69	21.5
Mn	0.95	32.12	6.93	19.51	2.73	11.71
Fe	0.87	26.21	18.04	20.21	23.36	22.79
Co	0.87	23.81	20.84	22.36	22.34	25.43
Ni	0.96	33.52	2.98	11.82	10.35	2.15
Cu	0.61	21.02	18.5	3.42	22.55	54.45
Zn	0.67	28.37	3.88	1.73	35.76	35.28
As	0.72	18.03	23.14	19.74	28.31	1.25
Sr	0.94	23.92	24.11	18.31	6.69	7.6
Pb	0.81	20.61	25.00	9.7	29.32	22.36
Th	0.79	1.36	30.86	25.05	18.51	15.21
U	0.74	20.61	14.34	36.74	34.17	3.63
Proportion %		52.20	28.65	9.88	6.30	2.97
Cumulative Proportion (%)		52.20	80.85	90.73	97.03	100



Fig. 4. (a) Cluster dendrogram and (b) Correlation matrix plot of elementals.



Fig. 5. (a) Different contribution sources of each element (in %) and (b) average contribution of each source based on the receptor model.



Fig. 6. Schematic diagram for probable sources of elements of agriculture soil over the study area.

understand the extent and types of contamination present.

# 5. Conclusions

Individual and synergistic pollution indices and APCS-MLR receptor models have been used to assess the ecoenvironmental risk, health hazards, and sources of potentially toxic elements in soil along the BCM in this study. The mean concentrations of Zn, Pb, V, Cu,

U Ni, Co and Th are higher than those of ASV, and the CVs of U, Cu, Co, Zn, Fe, Ni, As, Cl, Mn, and S show higher values. The individual indices of the soil samples refer moderately contaminated and polluted by As, Co, S, Pb, Th, and U, whereas synergistic indices show low toxicity and moderate pollution. The health risk assessment reveals that inhaling Cr, Co, and As puts adults and children at intolerable risk of developing cancer. Correlation coefficients extracted by using the APCS-MLR model for every metal showed  $R^2$  values of more than 0.5, indicating the reliability of the sources of origin. The possible sources are found by the APCS-MLR model and contributed 52.20 %, 28.65 %, 9.88 %, 6.30 %, and 2.97 % of elements to the studied soil. Considered as a whole, Fe, Ni, K, Sr, Al, Cl, Ti, Th, and Mn dominated natural sources of metals, while anthropogenic sources, Cr, Co, Cu, U, V, Zn, As, Pb, Th and S, are effluents through the BCM, and P, Zn, and S are contributed by agricultural activity.

The findings of the current analysis should be useful to regional and national planners as they choose the best course of action for preventing and reducing the amount of potentially harmful material pollution in soil. A deeper comprehension of elemental exposures and their probability source identification can also aid in the development of methods for mitigation, prevention, and remediation.

## Funding

No particular funding was obtained for this research project.

## Data availability

All relevant data used in the study are described, and included in the article and supplementary file.

# CRediT authorship contribution statement

Israt Jahan: Writing – original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization. A.H.M. Selim Reza: Writing – review & editing, Validation, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. Md Masidul Haque: Writing – original draft, Visualization, Validation, Formal analysis, Conceptualization. Md Selim Reza: Formal analysis, Data curation, Conceptualization. Md Irfanul Hasan: Writing – original draft, Software, Methodology, Formal analysis, Conceptualization.

## **Declaration of Competing interest**

We declare that this manuscript is original, has not been published before, and is not currently being considered for publication elsewhere. We know of no conflict of interest associated with this publication, and there has been no significant financial support for this work that could have influenced the outcome. As corresponding authors, we confirm that the manuscript has been read and approved by all the names of the authors.

## Acknowledgements

The author(s) thank Koichi Hoyanagi, Professor Emeritus, Shinshu University, Japan for his help during the geochemical analysis of soil samples in this research.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e32620.

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