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A Study on the Presence of Heavy Metals (As, Cd, Fe, Pb, Cr) and Their Spatial Distribution in Groundwater near Barapukuria Coal Field

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Abstract

Bangladesh carries the groundwater table very close to its surface and the table fluctuates with recharge and discharge conditions. This study aims to investigate the impacts of heavy metals in groundwater acquired from the shallow aquifer near a coal-bearing formation at Barapukuria, Dinajpur, Bangladesh. All the groundwater samples of the testing zones are tested using the ICP-OES. These samples are found contaminated for drinking, being rich in cadmium (more than 20 ppb) exceeding safe limits set by BSTI (5 ppb) and WHO (3 ppb). While arsenic touches the safe limit, chromium and lead are way below the range. The HEI and HPI indices of these zones are much higher and cadmium here is the prime contaminant. The hydrological structure being complex, the spatial distribution of the heavy metals is also predicted. Iron and cadmium levels decline at specific intervals while the rest are spread out alike over the region. This research will help further parley on the groundwater assessment of the study area and the impacts of underground coal mining.

Keywords: groundwater, heavy metals, heavy metal pollution index, heavy metal evaluation index, spatial distribution.

1. Introduction

Groundwater, infiltrated by sediments and rock cracks, is crucial for biodiversity, replenishing streams, rivers, and lakes through rainfall, despite being largely invisible. Even today, almost 30 times more water is stored underground than water in rivers, wetlands, lakes, or reservoirs [1]. Half of the world's irrigation and drinking water comes from groundwater, but continued mismanagement and mining operations beneath the surface of the planet threaten to seriously harm it. The increment of heavy metals in the environment is mostly released from rocks [2]. There are other sources such as the breakdown of rocks, mid-ocean ridges, and volcanic activity. The human activities that are responsible for the release of heavy metals in the environment consist of mining, smelting, coal burning, disposal of wastewater, etc.

Activities related to coal mining and burning are crucial for emerging nations like Bangladesh and China to advance economically, and recent decades have seen a sharp increase in the production and burning of fuel coal [3]. A variety of environmental problems are connected to the extraction, processing, conversion, and burning of coal [4]. Due to the production of many forms of mining wastes and their leftovers from burning coal, these wastes might contaminate groundwater by entering soils [5]. Groundwater pollution as a result of coal mine operations is a serious issue everywhere in the world [6]. Different hazardous heavy metals from coal waste, including Cd, Cu, Cr, Pb, Ni, As, and Zn, may leak into water bodies and provide the greatest long-term risks to groundwater [7].

Barapukuria is the first major coal field in Bangladesh that is in shallow mineable depth. The Barapukuria coal deposit is overlain by a 100 to 200-meter-thick poorly consolidated active groundwater aquifer. The coal seams happen to take place at depths between 118 meters to 506 meters under the surface. Water inrush into a mine is explicable jeopardy in case the adjacent environment hosts an aquifer system. There were occurrences of water inrush in the Barapukuria underground coal mine back in 1998 [8]. Bangladesh's aquifer water table is dropping due to withdrawals outpacing recharge, with mining operations causing contamination of groundwater sources. Poor water quality has global attention for human well-being. Due to their high persistence, toxicity, and carcinogenicity in people and other ecosystems, heavy metal contamination has become a global problem [9].

Research from coal basins throughout the world as in Bokaro coal basin and Korba coal basin in India, the Thrace coal basin and Mugla coal basin in Turkey, Linhuan coal basin in China have claimed that heavy metals heavily contaminate the groundwater of these basins. Several initiatives have been made to address the groundwater contamination caused by Bangladesh's coal mining and burning operations. Halim et al. (2013) demonstrated the movement and effects of higher heavy metals like Pb, As, and Mn in the water of the BCB region [10]. Bhuiyan et al. (2010) claimed that the proximal half of the basin has increased levels of heavy metals in water bodies [11]. D. A. Chowdhury and M. Jamaluddin Ahmed (2019) researched physicochemical characteristics of groundwater

quality from the Chittagong area, south-eastern Bangladesh using the standard method for physicochemical parameters as prescribed by APHA [12]. The major target was to evaluate the groundwater contamination of that area in terms of the presence of various heavy metals. They found out that most of the groundwater samples were within the safe limits of BSTI guidelines, except some were contaminated with arsenic, cadmium, and lead.
The overall aim and objective of this study are to identify the presence and measure the concentrations of certain heavy metals such as arsenic, cadmium, chromium, iron, and lead in the groundwater samples collected from the shallow aquifer through hand tube wells (HTW) from Barapukuria Coal Field (BCF) zone. Another major objective is to examine the spatial distribution of these heavy metals near the BCF zone. Being the only commercially operated coal field in this country, this research will surely catch the attention of the importance of coal and gas extraction without endangering the environment in this region. This research is significantly connected to human life and wildlife. Achieving the goal of this study through these objectives will support the security of groundwater quality. Since this sort of work has not been done yet in this region, this especially will create awareness in society.

2. Study area

Geological settings

The Barapukuria Coal Field was invented in 1985 by the Geological Survey of Bangladesh (GSB). It is located in the Parbatipur thana of the district Dinajpur. The coalfield has an area of 5.25 square kilometers. The Himalayan Foredeep to the north, the Shillong Shield/Platform to the east, and the Indian Peninsular Shield to the west are the boundaries of the Barapukuria coal mine (BCM) Basin, which is situated in Bangladesh's Dinajpur Shield. The Precambrian crystalline (granitic) basement lies unevenly beneath the coal-bearing sedimentary strata of the basin. The basin is stretched in a north-south orientation, measuring roughly 4.5 kilometers in length and 1.5 kilometers in width. A similar half-graben asymmetric basin atop the Precambrian foundation has been found in the important coal fields of West Bengal and Bihar in Central India [13].

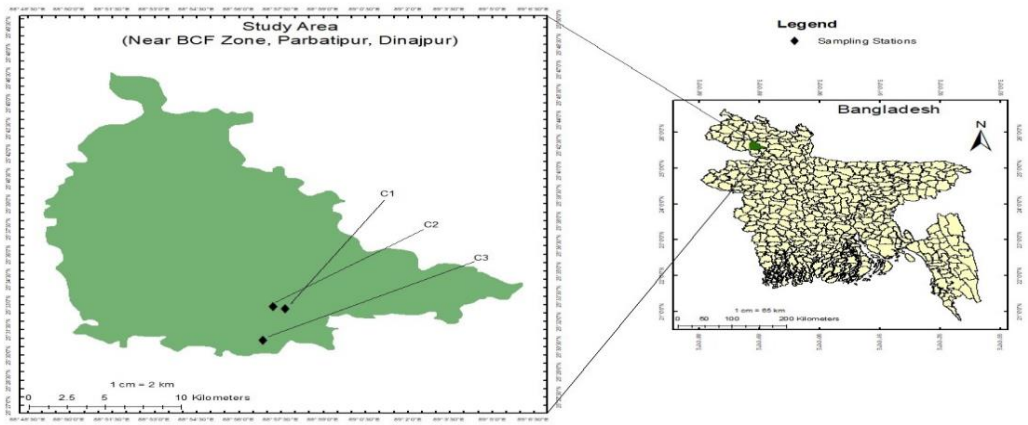


Fig. 1. Location map of the study area and the sampling stations generated using ArcGIS v10.7.1

Hydrogeological settings

It should be noted that, given the hydro-geologic context of the BCM, this mine is part of a far more complicated hydrogeological situation than other areas in Bangladesh. The main design restrictions, in reality, for the BCM are the large thickness of the seam (average 36 m), the presence of enormous Gondwana sandstones, and the unconsolidated Dupi Tila formation. The strata can be classified into four units Basement complex, Gondwana group, Dupi Tila Formation, and Barind Residuum Clay.
Table 1 showcases the hydro-stratigraphic sequences of BCF zone. The Barind Clay Residuum has an infiltration rate of roughly 1.5 mm/day. The upper Dupi Tila (UDT) with a depth that ranges from 102 to 136 m, is the main groundwater reservoir in the mining area. The lower Dupi Tila (LDT) is made primarily of grayish-white weathered residual clay and clayey silt. The depth of the floor varies from 115 to 118 meters. The coal seam VI separates the top and lower portions of the aquifer system that is represented by the Gondwana sandstones. The bottom portion thickens from the basin's northwest and southeast toward the center [15].

Table 1. Hydro-stratigraphic sequences of Barapukuria Coal Field Zone [14]

Age	Lithologic Units	Hydro-stratigraphic Units	Average Thickness (meters)
Pleistocene	Barind Clay Residium	Aquiclude	10
Pliocene	Upper Dupi Tila	Aquifer	104
	Lower Dupi Tila	Aquiclude	80
Permian	Gondwana	Aquifer	156
	Sandstone of Seam VI Roof	Aquifer	140
	Sandstone of Seam VI Floor	Aquifer	67
Archean	Basement complex	Relatively aquiclude	53
	Upper section Lower section	Aquiclude	31

3. Methodology

Several field investigations have been conducted to collect groundwater samples and assess the quality and contamination level of the water bodies around the BCF. The tested groundwater samples were collected through HTWs from the shallow aquifer that has a depth of almost 10 to 30 meters across the BCF zone. The samples were collected during the dry season, the pre-monsoon timeline of the year. The collected samples were cautiously sealed in plastic bottles with proper labeling. Samples were gathered in sterilized plastic bottles with an amber hue. Bottles were thoroughly rinsed many times using the same water before collecting. The samples were carefully sealed for further analysis.

According to Fig 1, the zone was subdivided into three sampling locations namely, C_1, C_2, and C_3. Table 2 depicts a precis of the sampling stations. C_1 was taken as the reference station and located inside the Barapukuria Coal Mine. C_2 is located 500 meters away and C_3 is located 5 kilometers away from the reference station. The Latitudes and Longitudes of the locations were acquired using the Global Positioning System (GPS) to create a location map of the study area.

Table 2: Location coordinates and distances of the sampling stations.

Sampling Stations	Coordinates		Locations	Distance (km)
	Longitude	Latitude		
C_1	88°57'39.85"E	25°32'46.57"N	Barapukuria Coal Mine	0
C_2	88°57'14.93"E	25°32'54.32"N	Phulbari Road	0.5
C_3	88°56'52.9"E	25°30'54.1"N	Malipara	5

The level of profanity can be measured by relying on some parameters namely Heavy Metal Pollution Index (HPI) and Heavy Metal Evaluation Index (HEI) etc. HPI is a mensuration of assessing the degree of heavy metal pollution in a specific property, usually in the water bodies or environment. Heavy metals can be defined as metallic elements with a high atomic weight with high density that have impacts on living organisms even at a lower concentration. Chromium, cadmium, arsenic, iron, lead, and mercury can be considered examples of heavy metals. The HPI offers a numerical evaluation of heavy metal pollution and aids in judging the level of contamination in a specific location [16]. The rating system, which is a random number between zero and one, can be evaluated by making values that are inversely proportional to the recommended standard for the relevant parameter [17] depending on how important a particular quality consideration is in comparison to other considerations. To calculate the HPI, unit weightage (W_i), a number that was inversely proportionate to the suggested standard (S_i) for the associated parameter [18]. The HPI model is given by

$$HPI = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i} \quad (1.1)$$

Where Q_i is the i^{th} parameter sub-index, W_i is the unit weightage i^{th} parameter and n is the number of parameters that are considered in the experiment. The unit weightage can be found using the following formula

$$W_i = \frac{K}{S_i} \quad (1.2)$$

Where K is the proportionality constant and S_i is the standard permissible limit of the i^{th} parameter. The sub-index can be calculated using the formula

$$Q_i = \sum_{i=1}^{n} \frac{M_i}{S_i} \times 100 \quad (1.3)$$

Where M_i is the calculated value of the heavy metal of i^{th} parameter. The critical pollution index of HPI for drinking water is approximately 0 to 100.

The HEI is a method used to assess the possible dangers to the environment and human health posed by the presence of heavy metals in soil, sediments, and water. It takes into account the level of heavy metals present as well as their capacity for bioaccumulation and toxicity in both plants and animals [19]. Larger scores indicate a larger potential danger of heavy metal contamination; the HEI score goes from 0 to 1, meaning that an HEI value below 1.0 is certified as "Fit" and an HEI value beyond 1.0 is classed as "Unfit" for home use [20]. It is used to interpret the pollution level and is expressed as

$$HEI = \sum_{i=1}^{n} \frac{M_i}{S_i} \quad (1.4)$$

The measurements of the concentrations of the detected heavy metals in the samples were conducted at the Advanced Analytical Chemistry and Research Lab (ACRL) in the Training Institute for Chemical Industries (TICI) at Polash, Narsingdi. The instrument inductively coupled plasma optical emission spectrometry (ICP-OES) and other laboratory apparatus were used. The ambient temperature at the time of testing was 22°C and the relative humidity was 55%. ICP-OES is a technique in which plasma and a spectrometer can be used to analyze the elemental content of (mainly water-dissolved) materials.

4. Results and discussions

Table 3 represents the overall result of the conducted study consisting of the concentrations of the selected heavy metals at distinct sampling stations. The concentrations are measured at parts per billion (ppb).

Table 3 Concentrations of heavy metals at each sampling station (measured at ppb)

Heavy Metals	Sampling Stations			Safe Limit According to BSTI	Safe Limit According to WHO
	C_1	C_2	C_3		
As	57.3	53.2	54.1	50	10
Cd	37.7	24.4	25.4	5	3
Cr	35.4	33.5	33.8	50	50
Fe	93.4	63.9	59.2	300	300
Pb	50.4	48	48.6	50	10

Fig 2 demonstrates the monitored concentrations of heavy metals according to the distinct sampling locations at a semi-logarithmic plot. This figure illustrates also the standard permissible limit of heavy metals in drinkable water approved by BSTI and WHO. All three groundwater sample locations from the BCF zone have extensively crossed the safe limits of cadmium proposed by both BSTI and WHO.

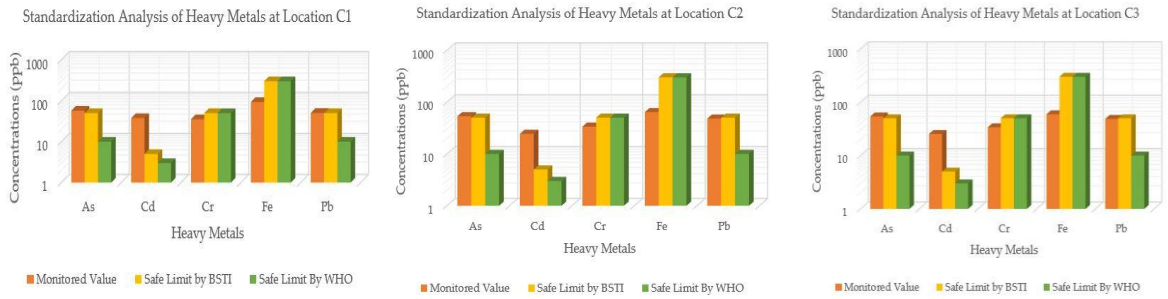


Fig. 2(a) Fig. 2(b) Fig. 2(c)

Fig. 2 (a), (b), (c) Concentrations of heavy metals at the sampling stations compared with the permissible limits of BSTI and WHO

The availability of certain heavy metals such as arsenic, chromium, and lead maintained a constant rate at almost all the samples from the BCF zone. However, it is to be noted that the concentrations of both iron and cadmium vary with the locations. Fig 3 confronts the enquired data of groundwater samples acquired from the region. The negative gradient of the concentrations of iron and cadmium in Fig 3 shows that these two heavy metals are spatially distributed with diversified concentrations. This illustration is pledged according to the locations and concentrations at the ppb level.

All the groundwater samples have excess levels of arsenic. On the contrary, all samples have iron levels below the permissible limits. Chromium and lead concentrations are at optimum levels in all three locations. BSTI and WHO has different standards for permissible limits of heavy metals in groundwater. As per Fig 3, both iron and chromium have the same permissible limits for BSTI and WHO.

But there appears a big margin of dissimilarities in the case of standardizations of arsenic, and lead. Cadmium has a slightly distinct secure flashpoint. Fig 4 depicts the overall summary of the heavy metals' concentrations along the three sampling stations in terms of the safety limits. Fig 5 is the demonstration of the HEI and HPI indices of the sampling locations of the BCF zone in a semi-logarithmic plot varying with distances. Both the values of HPI and HEI have significant discrepancies over the sampling locations.

These variances are shaped in a disciplined manner for BCF zone sampling stations. A decline is visible along the distances of the locations for both indices of HPI and HEI. C_1 being the reference location, the HEI and HPI values decline at C_2 and C_3 consequently. Over and above that, a linear forecast is predicted of these indices and is also depicted showing a downward gradient.

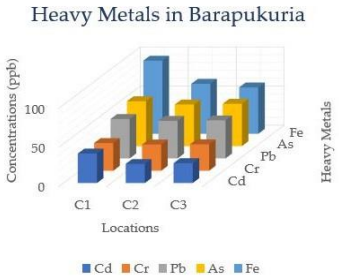


Fig 3 Concentrations of heavy metals at each sampling stations

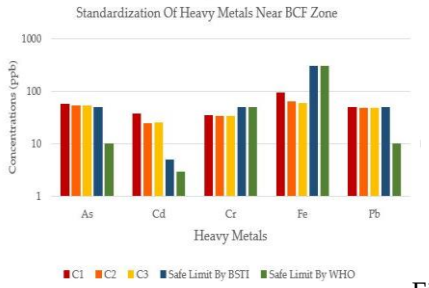


Fig. 4 Levels of Heavy Metals near BCF Zone

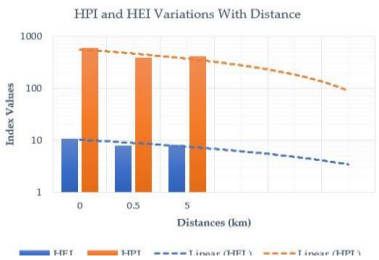


Fig. 5 Linear forecasts of HPI and HEI along the sampling locations.

The prime heavy metal pollutants in the sampling locations regarding the HPI values in the semi-logarithmic scale are portrayed in Fig 6. This represents the most principal constituent of the groundwater pollution of the sampling locations. Surprisingly, according to the HPI values, cadmium has the highest rates at all sampling stations. Therefore, cadmium is the key pollutant for BCF zone sampling stations.

The HPI values of water samples in these locations are found greater than the normal range of 0 to 100. The HEI values at these locations also exceeded the standard range of 0 to 1. Going further from the reference point, the concentrations of these heavy metals are mitigated. This defines that the groundwater quality is seriously defiled by the BCF zone sampling stations

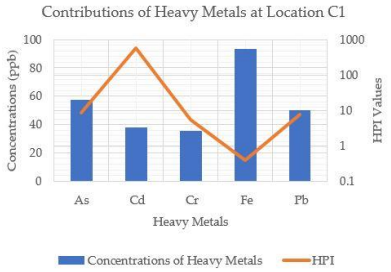


Fig. 6(a)

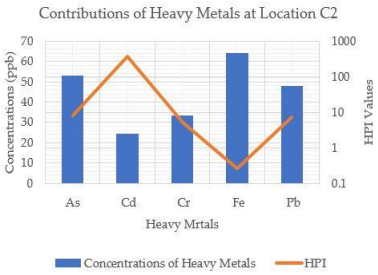


Fig. 6(b)

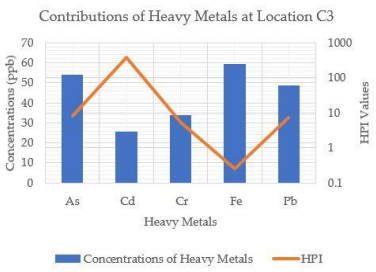


Fig. 6(c)

Fig. 6 (a), (b), (c) Major pollutants at sampling stations of BCF Zone

Table 4 sums up the overall pollution indices for the sampling stations. Fig. 7 is the visual representation of the heavy metals that are spatially distributed along the BCF zone, which depicts iron and cadmium concentrations to be minimized at a regular interval.

Table 4 HPI and HEI values for each sampling location

Sampling Stations	HPI	HEI
Barapukuria Coal Field zone		
C_1	594.86	10.7133
C_2	391.41	7.787
C_3	406.855	8

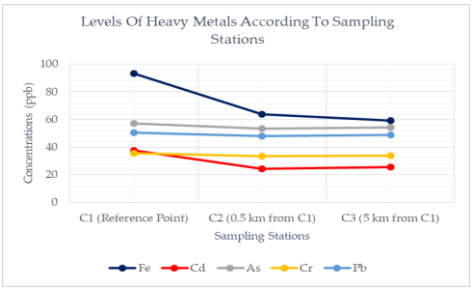


Fig. 7 Spatial distributions of heavy metals

5. Conclusions

The main objective of this study were to determine the concentration of heavy metals of the groundwater samples collected from BCF zone and their spatial distribution in that area. The notable key findings can be listed here in a nutshell:

- This research has a drawback in terms of the numbers of sampling stations and water samples for statistical analysis. Only selected heavy metals were detected, measured and analyzed.
- The groundwater samples collected from the BCF zone are highly contaminated according to the safe limits of BSTI and WHO.
- The equipment used (ICP-OES) had a particular sensitivity towards chromium and cadmium.
- The shallow aquifer is found to be cadmium-rich.
- Iron and cadmium are spatially distributed near the BCF zone.
- The concentrations of the heavy metals decrease with the distance from the center of the BCF which indicate that the coal-bearing formations have great influence on groundwater regarding heavy metal contents.
- Chromium and lead near BCF zones are well within the safe limits of BSTI and WHO.
- Arsenic is found just above the safe limit near the BCF.
- The effects of mining on groundwater aquifers should be evaluated. The hydrologic system must be protected from prejudice by planning mitigation measures.
- To maintain a good mining operation, groundwater monitoring operations must be carried out.

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