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Abstract

Production of coal is of utmost importance for energy generation while being crucial for the economic welfare of the nation. Mining of this precious natural resource however results in a hazardous aftermath owing to the built up accumulation of heavy metals that are non-biodegradable and damage the environment. The current study evaluates the impact of metal contamination on water from underground and open-cast mines in two different seasons by pollution indices approach. pH analysis placed all samples under study in the acidic scale with an average range of 2.4-3.65. Samples from open-cast were noted to have more of total dissolved solids compared to the underground mine samples. The concentration of metals in the samples were seen to be in the order Cu < Pb < Cr < Zn <Fe from low to high. The leaching of heavy metals due to the mining operation has led to the contamination of the water bodies as depicted by HPI and HEI values. Higher metal accumulation is noted in the pre-monsoon compared to the post-monsoon season, with samples from underground mines showing slightly lower metal contamination compared to that of the open-cast mines. A feasible and effective technology needs to be integrated into conventional coal mining methods in order to contain and prevent the detrimental leaching of metals contaminating the environment.

Keywords: Metal contamination, Coal mines, Underground, Open-cast, Water pollution indices

Introduction

Coal is the most abundant and essential fossil fuel in India. With increase in demographic pressure, growing economy and a need for improving quality of life, the demand for energy in India is also rising. Mining not only helps in fulfilling the increasing energy demand of scores of industries, but also provides an important opportunity for the economic development of the country (Chaulya and Chakraborty, 1995). Almost 45% of the total energy consumption in India is met by coal, which makes it indispensable in the

welfare of our economy (Energy Statistics, 2017). In India, coal is mined and extracted via two main methods namely, underground mining and open-cast mining. However, coal extraction in India is mostly done by open-cast mining method and it constitutes 93.26% of the total coal production in the country (Coal Directory of India, 2016-17). The process of extracting coal by open-cast mining has increased over the years, as this method incurs less expenditure with minimum wastage (Ghose and Majee, 2001).

Choice of mining method depends on various factors such as geological conditions (type, depth, size and quality of deposit), technological development and degree of mechanisation. Besides these, other factors need to be taken into consideration, such as production cost along, selling price, environmental and social aspects, all of which are also significant in the entire process of mining. All of these factors determine the differences between resources and reserves of coal (Zehirov, 2017). Open-cast method is usually decided for areas with shallow coal seams, while underground mining method is the approach selected to access deeper coal seams (Mukherjee and Pahari, 2019). Both of the mining methods drastically affect the mined area and its surrounding environment with huge amounts of water being discharged onto the surface during the process of mining, in order to facilitate the mining operation. These liquid mine discharges usually contain high concentrations of TDS, TSS and heavy metals which contaminate the surface and ground water of the area (Dhar, 1993; Tiwary, 2001). Coal extraction is an intricate process which generates hazardous toxicants and heavy metals due to their virtue of being highly solubility in nature. Metals such as Fe, Cu, Mn, Co, Ni, Pb, Zn and other mineral dusts are released in the soil and water bodies creating an acidic environment (Chandra and Jain, 2013). The chief cause of coal mine pollution is leaching of acidic mine tailings or run-offs known as 'Acid Mine Drainage' (AMD) discharged into the surroundings. The sulfide ores present in the mines get readily oxidised in presence of air and water to form sulphate-rich acidic drainage resulting in leaching of metals and other metalloids. This causes increased accumulation of inorganic matter and soil acidity while reducing the organic matter content of the natural environment. The discharged effluents from coal mines pick up metals, combined with other mine wastes and leftovers that pollutes the soil and adjacent water bodies thus causing a massive alteration in the dynamics of associated life forms (Johnson and Hallberg, 2005; Zhou et al., 2007). The increase of heavy metals in the environment is said to have a dynamic effect on all living organisms, threatening the food chain, indigenous flora, fauna and native soil microbiota (Yao et al., 2012). In India, a majority of the coal mining districts have been assessed and declared as critically polluted areas by MoEF (CSE, 2012).

Contamination of water bodies in and around coal mines with heavy metals raises an alarm that calls for the need to assess the water quality standard. This can be evaluated using the indexing approach such as the heavy metal pollution index (HPI) and heavy metal evaluation index (HEI), that takes into consideration the overall quality of water with respect to weighted arithmetic quality mean and heavy metal concentrations compared to its respective acceptable standards (Edet and Offiong, 2002). Metal pollution

indices, HPI can be categorized into three main classes *viz* low (< 300), medium (300-600) and high level (> 600); HEI values are also placed in three categories *viz* low (< 150), medium (150-300) and high (> 300), respectively (Bhuiyan *et al.*, 2010; Mahato *et al.*, 2017). In the current study, water samples from both underground and open-cast coal mines have been considered in order to assess the impact of heavy metal contamination on the overall quality of water.

Materials and methods

Study sites and sample collection

Two types of coal mines, underground and open-cast were targeted for the collection of samples in order to assess the impact of metal contamination on water quality. Underground mines are in Khliehriat, Jaintia hills district of Meghalaya and open-cast mines are located in Ledo, Tinsukia district of Assam, India (Figure 1). Four water samples each, were collected from the two mine sites – MW1, MW2, MW3 & MW4 represent underground mines whereas AW1, AW2, AW3 & AW4 represent open-cast mines. Samples were collected in sterilized sample bottles by immersing the bottles about 10 cm below the water surface and brought to the laboratory for analysis. The samples were collected in two seasons, pre-monsoon and post-monsoon seasons.



Figure 1: Underground and open-cast methods of coal mining in Meghalaya and Assam

Determination of physicochemical parameters

Three main parameters were considered for this analysis – pH, Total dissolved solids (TDS) and Electrical conductivity (EC). The pH of the samples were recorded by means of DIC μ pH meter (GOLD 533, Digital Instrumental Corp) calibrated using standard buffers, washing the pH probe between measurements using sterilized deionized water (Rayment and Higginson, 1992). TDS of samples was measured using pre-calibrated EcoTestr TDS (Eutech instruments). For EC, DiST® 4 EC Tester (HANNA Instruments) was used by standardisation of probe using 0.01M KCl solution and conductance recorded.

Estimation of Heavy metals

Five heavy metals were analyzed in the water samples namely iron (Fe), zinc (Zn), copper (Cu), lead (Pb) and chromium (Cr). The presence of metals in the samples was carried out following the protocol of acid digestion. Accordingly the samples were filtered through Whatman filter paper No.42 and the pH adjusted to pH<2 with HNO₃. The sample (500 ml) was heated allowing evaporation and finally concentrated to a residual volume of 50 ml. The concentrate was filtered and subjected to metal estimation using *ICP-OES* (Thermo Scientific iCAP 7600). The concentration of metals was expressed in parts per million (ppm) using a standard formula (Radulescu *et al.*, 2014).

Determination of water pollution indices

Two main pollution indices were considered in the study namely Heavy metal pollution index (HPI) and Heavy metal evaluation index (HEI). The indices are a measure of total quality affected by the presence of different heavy metals in the water samples. For determination of HPI, two equations are given:

Eq. 1
$$Qi = \sum_{i=1}^{n} \frac{\{Mi(-)Ii\}}{(Si-Ii)} \ge 100$$
; Eq. 2 $HPI = \frac{\sum_{i=1}^{n} Wi Qi}{\sum_{i=1}^{n} Wi}$

For sub index Qi: Mi - monitored value for the heavy metal; Ii - ideal desirable value; Si - standard value of the i^{th} parameter.

For **HPI**: *Wi* - unit weightage of *i*th parameter, a value inversely proportional to Si of the metal (Prasad *et al.* 2014).

For determination of HEI, $HEI = \sum_{i=1}^{n} \frac{Hc}{Hmac}$

 H_c - monitored value; H_{mac} - maximum admissible concentration (MAC) of the *i*th parameter (Edet and Offiong, 2002).

Results and Discussion

Physicochemical profile of the samples

Three main parameters were taken into consideration for physicochemical profiling of the water samples *viz* pH, total dissolved solids and electrical conductivity.

Underground coal mine samples											
MW1 MW2 MW3 MW4											
Parameters	PRE	POS	PRE	POS	PRE	POS	PRE	POS			
pН	3.63	2.63	3.24	2.41	3.57	2.51	3.65	2.54			
TDS	561	246	990	401	710	358	607	315			
EC	807	702	1597	978	1568	786	1046	780			
			Open-cast o	coal mine sa	amples						
	AV	W1	Open-cast of	coal mine sa V2	amples AV	W3	AV	V4			
Parameters	AV PRE	V1 POS	Open-cast of AV	coal mine sa V2 POS	amples AV PRE	W 3 POS	AV PRE	V4 POS			
Parameters pH	AV PRE 3.21	V1 POS 2.7	Open-cast of AV PRE 3.11	version of the second mine second mine second mine second	amples AV PRE 3.13	W3 POS 2.9	AV PRE 3.16	V4 POS 2.9			
Parameters pH TDS	AV PRE 3.21 1860	V1 POS 2.7 1662	Open-cast of AV PRE 3.11 2360	coal mine sa V2 POS 2.54 2126	AND PRE 3.13 2040	W3 POS 2.9 1984	AV PRE 3.16 2240	V4 POS 2.9 1864			
Parameters pH TDS EC	AV PRE 3.21 1860 3780	V1 POS 2.7 1662 3340	Open-cast of AV PRE 3.11 2360 4482	v2 POS 2.54 2126 3961	amples A PRE 3.13 2040 4422	W3 POS 2.9 1984 3501	AV PRE 3.16 2240 4284	V4 POS 2.9 1864 3263			

Table 1: Physicochemcial profile of water samples from the mines in two seasons

PRE – Pre-Monsoon; POS – Post-monsoon; BDL –below detection level; TDS – Total dissolved solids (mg/L); EC- Electrical conductivity (μS/cm).

For underground mines, the highest pH recorded was 3.65 and an average reading of pH 3.02 ± 0.19 . The average pH reading for the open-cast samples was 2.95 ± 0.08 and the highest reading being 3.21. The pH of the samples is noted to be slightly lower and more acidic during the post-monsoon than the pre-monsoon season, similar to that reported by Sahoo *et al.* (2011). The overall pH of water bodies in and around coal mines is acidic and much lower than the prescribed limit of pH 6-8 (WHO, 2011). Low pH due to coal mine overburdens has been reported in several studies (Dowarah *et al.*, 2009; Rai *et al.*, 2011). Increased acidity is attributed to the acid drainage and mine spoils leaching into the water systems in the vicinity of the mines (Cherry *et al.*, 2001). The geology of the rock composition and mineral deposits also add to the acidification of mining effluents (Dutta and Agarwal, 2002). The average total dissolved solids (TDS) of the samples for the pre and post monsoon seasons was found to be 717 ± 96.18 and 330 ± 33.04 mg/L in underground mine samples whereas 2125 ± 110.26 and 1909 ± 98.21 mg/L in the open-cast

samples, respectively. The average electrical conductivity (EC) of the underground mine samples was recorded to be 1254.5 ± 195.64 and $811.5\pm58.70 \ \mu$ S/cm, whereas that of opencast samples was 4242 ± 159.48 and $3516.25\pm156.32 \ \mu$ S/cm, for the pre and post monsoon seasons respectively (Table 1). The results corroborate with the past findings reporting EC of coal mine streams to be approximately $3630 \ \mu$ S/cm (Lyngdoh and Kayang, 2012). High conductance of water bodies surrounding coal mines has been suggested in earlier studies and this could serve as an indicator for mining activities indicating contamination (Soucek *et al.*, 2000). The TDS and EC of all samples is seen to be higher in the opencast mine as compared to the underground mine samples; also higher values are seen in the pre-monsoon than the post-monsoon season. TDS and EC are positively correlated but are however inversely related to pH. High TDS and EC level with decrease in pH indicates severe water pollution (Islam *et al.*, 2017).

Impact of metal contamination on water quality

The methods employed in coal extraction are in many regards crude and unscientific which tend to have a serious damaging impact on the environment. Coal mine sites and the adjacent areas are most affected with mine spoils containing heavy metals and other contaminants percolating into the soil and water systems. In this current study, water from underground and open-cast mines was assessed for the impact of contamination caused by heavy metals, based on water pollution indices - heavy metal pollution index (HPI) and heavy metal evaluation index (HEI). Four samples each, collected from the two types of mines for pre and post monsoon seasons, were analyzed for the presence of heavy metals.

Underground coal mine samples									
Samples	MW1		Μ	W2	М	MW3		W4	
	PRE	POS	PRE	POS	PRE	POS	PRE	POS	
Fe	47.34	36.64	93.03	56.18	95.23	56.18	78.55	52.83	
Zn	2.740	1.347	3.160	1.870	4.01	2.101	4.101	2.013	
Cu	0.002	BDL	0.003	0.001	0.004	0.001	0.004	0.002	
Pb	0.062	0.035	0.031	0.012	0.037	0.019	0.044	0.021	
Cr	0.055	0.031	0.063	0.036	0.071	0.041	0.107	0.087	
			·						
Open-cast coal mine samples									
Samples	AV	W1	AW2		AW3		AW4		
	PRE	POS	PRE	POS	PRE	POS	PRE	POS	
Fe	132.2	80.02	113.1	68.12	96.70	45.30	145.3	95.45	
7	2 400	1.007	1 671	0 975	0.022	0.792	2 001	1 745	

Table 2: Heavy metal concentration of water samples from the mines in two seasons

Samples	AW1		AW2		AW3		AW4	
	PRE	POS	PRE	POS	PRE	POS	PRE	POS
Fe	132.2	80.02	113.1	68.12	96.70	45.30	145.3	95.45
Zn	2.400	1.007	1.671	0.875	0.922	0.783	3.901	1.745
Cu	0.004	0.002	0.002	0.001	0.002	BDL	0.007	0.003
Pb	0.052	0.030	0.047	0.022	0.025	0.018	0.071	0.047
Cr	0.216	0.162	0.144	0.113	0.112	0.095	0.194	0.143

Heavy metals (mg/L); PRE - Pre-Monsoon; POS - Post-monsoon; BDL -below detection level

The concentration of metals in the samples were found to be in the order Cu < Pb < Cr < Zn < Fe from low to high, with Fe being highest in concentration for all the samples (Table 2). The analysis of water from coal mine area of Damodar River India reported a similar finding where Fe is said to be one of the main heavy metal contaminants found in high concentrations in water bodies of coal mine sites (Mahato et al., 2017). The overall pollution indices calculated for the samples revealed open-cast mine samples to have higher values in terms of HPI (482.62) and HEI (103.98) than that of underground mine samples with HPI (363.69) and HEI (69.17), respectively (Table 3). Metal pollution indices for each individual samples from both mine types, along with mean indices, for the two seasons were evaluated and tabulated. The mean HPI of underground mine samples was noted to be 473.73 and 253.67, and mean HEI of 84.603 and 53.729; whereas opencast samples showed mean HPI of 598.12 and 367.08 and mean HEI of 130.18 and 77.78, for pre and post monsoon season, respectively (Table 4). The pollution index values exhibited by open-cast samples were noted to exceed that of underground samples by 100 units, indicating a comparatively higher metal concentration and contamination in open-cast mines. The analysis according to seasonal variation showed pre-monsoon to have higher level of water contamination than the post-monsoon season.

	Underground coal mine samples								
Heavy Metals	<i>Mi</i> Mean value (µg/L)	<i>Si</i> Standard permissi- ble value (µg/L)	<i>Ii</i> Ideal desirable value (µg/L)	<i>Wi</i> Unit weightage	<i>Qi</i> Sub index	Wi x Qi	H_c/H_{mac}		
Fe	64497.5	1000	300	0.001	9171.071	9.171071	64.4975		
Zn	2667.75	15000	5000	0.00006	23.3225	0.0014	0.17785		
Cu	2.125	1500	50	0.0006	3.30172	0.00198	0.001417		
Pb	32.625	10	0	0.1	326.25	32.625	3.2625		
Cr	61.375	50	0	0.02	122.75	2.455	1.2275		

Table 3: Mean HPI	and HEI	calculation	of all	the samples	collected	from t	he r	nines	in
two seasons									

 Σ *Wi* = 0.12166; Σ *WiQi* = 44.247. **HPI** = 363.69. **HEI** = 69.17.

Open-cast coal mine samples								
Heavy Metals	Mi	Si	li	Wi	Qi	Wi x Qi	H_c/H_{mac}	
Fe	97023.75	1000	300	0.001	13817.68	13.81768	97.02375	
Zn	1663	15000	5000	0.00006	33.37	0.002002	0.110867	
Cu	3	1500	50	0.0006	3.241379	0.001945	0.002	
Pb	39	10	0	0.1	390	39	3.9	
Cr	147.375	50	0	0.02	294.75	5.895	2.9475	
$\Sigma Wi = 0.12166; \Sigma WiQi = 58.716.$ HPI = 482.62. HEI = 103.98.								

HPI- Heavy metal pollution index; HEI- Heavy metal evaluation index. Standards BIS IS10500: 2012, WHO (2011)



Figure 2: Categorization of water quality for samples from underground and open-cast coal mines based on heavy metal Pollution Index (HPI).

A similar pattern was observed by Singh and Kamal (2017) where they reported lower metal concentration in coal mine water bodies during the post-monsoon and higher in the pre-monsoon season. This could be due to the heavy rains during monsoon seasons causing strong water currents washing away most of the suspended and dissolved matter along with other debris. All water samples of pre-monsoon season were seen to fall in the medium range of pollution, while most samples of post-monsoon being in the low pollution range, according to their respective HPI index. Only two samples belonging to the open-cast mines were placed in the high pollution range (Figure 2). In the case of HEI, all the samples were noted to fall in the low pollution range with not much distinction seen between the two types of mine samples (Figure 3). These findings corroborates with the study by Bhuiyan *et al.* (2010) reporting similar HPI and HEI index values for quality analysis of water samples from coal mines. In a number of developing countries, water pollution indices have been successfully employed for determination of water quality assumed to be burdened by metal contamination (Edet and Offiong, 2002; Prasad and Mondal, 2008).



Figure 3: Categorization of water quality for samples from underground and open-cast coal mines based on Heavy metal Evaluation Index (HEI)

Underground mine samples							
Samples		Pre-Monsoor	1]	Post-Monsoor	1	
	HPI	Mean Dev	% Deviation	HPI	Mean Dev	% Deviation	
MW1	582.909	109.1	23.05	340.516	86.84	34.24	
MW2	384.383	-89.35	-18.86	176.056	-77.61	-30.60	
MW3	438.919	-34.81	-7.35	235.238	-18.43	-7.27	
MW4	488.707	14.97	3.16	262.868	9.198	3.63	
	Mean 473.73	-		Mean 253.67	-		
Samples		Pre-Monsoon	1]	Post-Monsoor	1	
	HEI	Mean Dev	% Deviation	HEI	Mean Dev	% Deviation	
MW1	54.825	-29.77	-35.20	40.849	-12.88	-23.97	
MW2	97.602	12.99	15.36	58.225	4.496	8.37	
MW3	100.62	16.01	18.93	59.040	5.311	9.88	
MW4	85.366	0.763	0.90	56.805	3.076	5.73	
	Mean 84.603	-		Mean 53.729	-		
		Open	-cast mine samp	les			
Samples		Pre-Monsoon	1]	Post-Monsoor	 l	
	HPI	Mean Dev	% Deviation	HPI	Mean Dev	% Deviation	
AW1	653.348	55.17	9.23	393.498	26.41	7.20	
AW2	566.154	-32.01	-5.35	297.658	-69.42	-18.91	
AW3	355.547	-242.6	-40.56	232.066	-135.0	-36.78	
AW4	817.662	219.5	36.70	545.099	178.0	48.50	
	Mean 598.12	-		Mean 367.08	-		
Samples		Pre-Monsoon	1]	Post-Monsoor	1	
	HEI	Mean Dev	% Deviation	HEI	Mean Dev	% Deviation	
AW1	141.882	11.70	8.99	86.328	8.542	10.98	
AW2	120.792	-9.388	-7.21	72.639	-5.147	-6.62	
AW3	101.502	-28.67	-22.03	49.052	-28.73	-36.94	
AW4	156.544	26.36	20.25	103.128	25.34	32.58	
	Mean 130.18			Mean 77.78			

HPI (Heavy metal pollution index): Low <300, Medium 300-600, High >600; HEI (Heavy metal evaluation index): Low <150, Medium 150-300, High >300 (Bhuiyan *et al.*, 2010).

Results suggest higher contamination of water bodies caused by open-cast mining of coal rather than underground mining. However, both types of mining operations are seen to cause uncontrolled leaching of these non-biodegradable heavy metals rapidly accumulating in the environment and finding their way into adjacent water and soil systems. Unscientific methods of coal mining are bound to cause detrimental impacts on water quality making it unsafe for domestic use or consumption. These contaminants continue to persist in the environment and are getting incorporated in food chain affecting biotic communities and vital ecological processes (Duarte *et al.*, 2008). This calls for the need of an innovative technology for controlling and restraining the careless leaching of mine effluents that could be integrated along with conventional mining procedures in order to reduce the harmful consequences related to mining of coal.

Conclusion

The complex and unmonitored methods of coal extraction has caused severe damage to the coal mining areas due to leaching of mine effluents into the surrounding environment. Such areas face an acute shortage of clean and safe water for consumption and domestic use. This study was taken up with the objective to assess the impact of heavy metals on the overall quality of water from underground and open-cast coal mines. The analysis was performed on metal-based water pollution indices HPI and HEI. The pollution index values revealed higher contamination in the pre-monsoon than postmonsoon season. Majority of the mine water samples fall in the medium range of pollution, with samples from underground mines showing slightly lower metal contamination in comparison to that of the open-cast mines. The degree of metal discharged from the mines could be influenced by the type of mining method employed for the extraction of coal. This study serves to provide a baseline information of the difference in the water quality impacted by metal leaching from underground and open-cast mines.

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