AN INVESTIGATION OF HEAVY AND TRACE ELEMENTS IN COAL DEPOSITS OF MAKARWAL PAKISTAN AND THEIR POSSIBLE IMPACTS ON SURROUNDING WATER-CASE STUDY

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Abstract

This research present, the coal samples from different locations of Makarwal (Pakistan) were investigated for the existence of trace and heavy metals. Coal samples used under investigation were: Makarwal section-A (Mk-A), Makarwal section-B (Mk-B), Kurd section C (Kr-c), Makarwal section D (Mk-D) and Gula-khel section E (Gk-E). It was observed that, these coalfields contain arsenic (As) ranging from 3.0–16.34mg/kg, cadmium (Cd) 2.23–4.61 mg/kg, Co 22.1–29.3 mg/kg, Cr 113.1–288.2 mg/kg, Cu 8.53–23.2 mg/kg, Fe 1133–10813 mg/kg, Ni 44.35–148.8 mg/kg, Pb 18.49–46.25 mg/kg and Zn 25–242 mg/kg. Moreover, the Mk-D coal samples had the high concentration of Ni, Cr, As, and Cd. However, the coal samples from Mk-C had higher contents of Cu and Pb. Whereas, Fe was abundant in Mk-B coal samples. These results can be used as a helping tool for the prediction of environmental risks associated with coal mining in Makarwal area. Further, the water samples of Makarwal area were also analyzed and found to be contaminated with heavy metal pollutants.

KEYWORDS: *Water contamination, environmental issues, heavy metals and coal energy.*

INTRODUCTION

Coal is a combustible black or brownish black rock formed from the transformation of organic matter over millions of years at a high temperature and pressure under anaerobic conditions on the edge of the sedimentary basin (Rahmani and Flores, 1985).

Coal is complex mixture organic and inorganic compounds. Organic compounds in coal is in the range of 70 to 90 percent due to plant remains after decay of microbial degradation. In coal, some inorganic elements occur in parts per million which does not import serious environmental hazards on combustion (Finkelman and Brown, 1991). Exceeds of these concentration limits have some hazardous effect on environment. However, affinity of trace elements in coal for the organic compounds in the minerals highly effect the calorific value of coal (Finkelman *et al.,* 2002; Wang, 2005).

Pakistan is a growing economy and to meet the new challenges and to overcome the energy crisis in the country, coal exploration and utilization is vital. Currently, Pakistan is facing severe energy crisis. To meet the challenges of energy crises, the exploration and utilization of coal in Pakistan is essential. Coal on the other hand is not a clean source of energy and may contain a different ranges of heavy and toxic metals usually present in traces (parts per million). Some of these metals such as arsenic, cadmium or selenium are considered to be carcinogenic when surpass a permitted limit in coal (Finkelman and Brown, 1991). These metals have adverse impact on the life of both human health and plant growth during chemical combustion and mining operations (Finkelman *et al.,* 2002; Wang, 2005). To meet the energy crisis and management of the country, coal is use as alternative solid fuel, the only limitation of coal is because of it not consideration as green source of energy due to the trace elements as interpreted in this research work.

Makarwal coalfield has about 22 million tons of coal and they mainly exist in the Paleocene Hangue formation in the Shinghar and Surghar Ranges which constitute the northernmost part of the Trans Indus Mountains (Warwick *et al.,* 1995). The Makarwal coalfield is in the Mianwali district of the Punjab as shown in figure 1. The thickness of coal seams differs from less than 0.5 meter to about 1.9 meters. Coal is generally volatile B bituminous with a high calorific value from 93000-13200 BTU /lb (Warwick and others, 1990).

In this research study, Makarwal coal fields were selected to analyze the concentration of heavy metals such as Zn, Co, Cd, As, Fe, Ni and Pb which are the by-product of coal combustion. These heavy and trace

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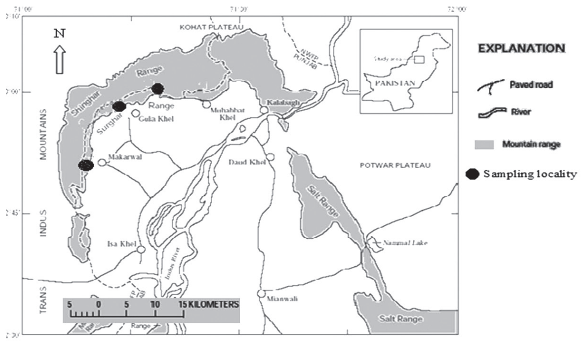

Fig. 1: Location map of Makarwal coal sampling localities (Adopted from Danichick USGS).

elements can be volatilized during coal combustion, which may be inhaled, adsorbed on the harvested corps taken up by animals and also causing severe environmental issues.

METHODOLOGY

Field Observation

The field studies involved personal visits to the coal-fields and collection of 18 representative coal samples from Makarwal coal field. The eighteen (18) samples were collected from Makarwal section-A (MkA), Makarwal section-B (MkB,), Kurd section C (Kr-c), Makarwal section D (Mk-D) and Gulakhel section E (Gk-E). Additionally, samples were also collected from surface and groundwater. The samples were brought into the laboratory for the analysis of heavy and trace elements.

Laboratory investigations

Each coal sample was placed in furnace for drying and then pulverized to a particle size of 200 meshes in ball mill. Conning and quartering method was used for the selection of representative portion of individual sample. Fixed amount of sample was for further processing and characterization. Heavy metals such as (Fe, Pb, Zn, Cu, Cd, Ni etc) were analyzed through spectrometer of Perkin Elmer atomic absorption. Auqa-Regia and mixture of acid was prepared to digest the prepared sample.

RESULTS AND DISCUSSION

Both heavy and traces elements concentration in the various sections of Makarwal coalfields were determined through spectrometer of Perkin Elmer-700 for atomic absorption. The data are given in the table 1.

Rajappa, 2010 reported that in bituminous coal arsenic is organically bounded as pyrite mineral. Swaine (1990) also stated that arsenic occurs in coal as arsenical pyrite and arsenopyrite (AsFeS). Arsenic enter in human body by drinking through contaminated water (Johnson and Bretzler, 2015). According to WHO, (2008) the permissible level of As in drinking water is 0.01 mg/l. Fig. 2, show the amount of As present in the samples of coal analyzed. The As concentration in section-A was found in the range 0.75 to 8.46 mg/kg (3.29 mg/kg), in section-B it ranges from 2.18 to 38.49 mg/kg (15.93 mg/kg), while in section C, D and E arsenic ranges from 1.36 to 5.23 mg/kg (3.0 mg/kg), 13.29 to 19.5 mg/kg (16.34 mg/kg), and 2.3 to 3.8 mg/kg (3.1 mg/kg), respectively. This shows that on the average section-D has highest content of arsenic as compared to other section of the study area as shown in the table 1 and figure 2.

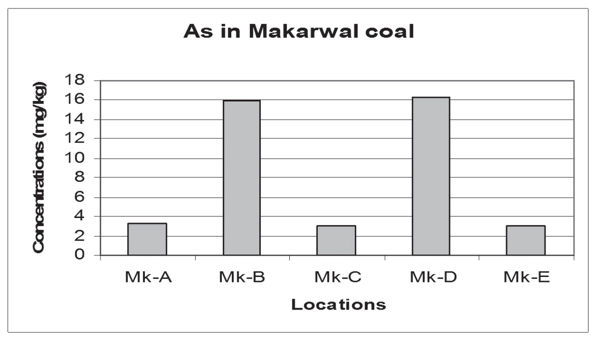

Fig. 2: Concentration of Arsenic (As) in different coal fields of Makarwal.

Arsenic concentration in water samples of Makarwal area are given in table 1. The results show that the concentration of As was at maximum level in the drinking water as prescribed by the USEPA and WHO (2008).

Cadmium (Cd) mode of existence in coal is inorganically bound (Gluskoter, 1973). Cd is released to the atmosphere during coal combustion in the form of fly ash or bottom ash (Davidson *et al.,* 1994). WHO, (2008) has reported permissible limit for Cd as 0.003 mg/l. Whereas, Cd concentrations shown in table 1 and figure 3, are above the limit as reported by the USEPA and WHO drinking standards. Fig. 3, shows that Cd concentration in section A was in the range 2.9-5.95 mg/kg (4.1 mg/kg), in section-B it ranged from 2.0 to 5.35 mg/kg (3.31 mg/kg), while in section C, D and E cadmium ranges from 2.15 to 7.05 mg/kg (4.39 mg/

kg), 1.35 to 7.0 mg/kg (4.61 mg/kg), and 2.0 to 2.45 mg/kg (2.23 mg/kg) respectively. Thus, this shows that on the average, section-C has the highest contents of Cd as compared to other sections as shown in the table 1 and figure 3.

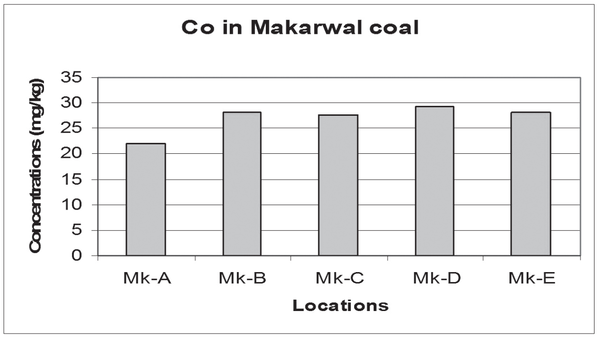

Fig. 3: Concentration of Cadmium (Cd)) in different coal fields of Makarwal.

Cobalt occurs in an inorganic fraction in coal (Finkelman *et al.,* 1993). It is important for humans, plants and animals because it is responsible for the synthesis of vitamin B_{12} (Krishnan, 1996). Excessive accumulation mainly occurs in the liver, kidneys, and bones. In drinking water the maximum tolerance limits for cobalt is not available. Fig. 4, depicts that, the Co concentration in section-A was found in the range 13.9-28.35 mg/kg (22.1 mg/kg), in section-B it ranged from 14.8 to 40.1 mg/kg (28.26 mg/kg), while in section C, D and E arsenic ranges from 9.1 to 40.45 mg/kg (27.66 mg/kg), 22.7 to 44.85 mg/kg (29.3 mg/kg), and 23.75 to 37.25 mg/kg (28.25 mg/kg) respectively. This shows that on the average, section-D has the highest contents of cobalt as compared to other sections as shown in the table 1 and figure 4. However, concentrations of cobalt in water samples of Makarwal area was 0.065 as shown in table 2, whereas, its contamination limit is not available in the literature.

There are in consistent evidences in the literature on Cr mode of occurrence in coal. Some authors reported that Cr is organically bound in coal, whereas others found that Cr is either an intermediate product of mineralization or inorganically bound or both (Frinkelman, 1991). The USEPA (2004) and WHO (2008) suggested a tolerance limit of Cr as 0.1 mg/l and 0.05 mg/l respectively, in drinking water table 2. Fig. 5, shows that the Cr concentration in section-A was found in the

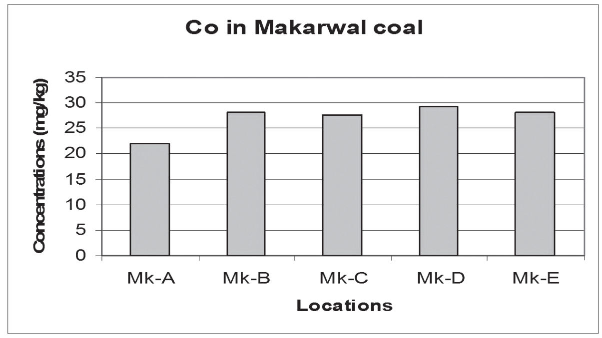

Fig. 4: Concentration of cobalt (Co) in different coal fields of Makarwal.

range 108.45-329.1 mg/kg (183.75 mg/kg), in section-B it ranged from 106.75 to 270.1 mg/kg (189.48 mg/kg), while in section C, D and E arsenic ranges from 13.7 to 631.5 mg/kg (286.7 mg/kg), 102.75 to 478.45 mg/kg (288.2 mg/kg), and 70.1 to 191.0 mg/kg (113.12 mg/kg), respectively. Therefore, this shows that on the average section-D have the highest contents of chromium as compared to other sections of the study area as shown in the table 1 and figure 5.

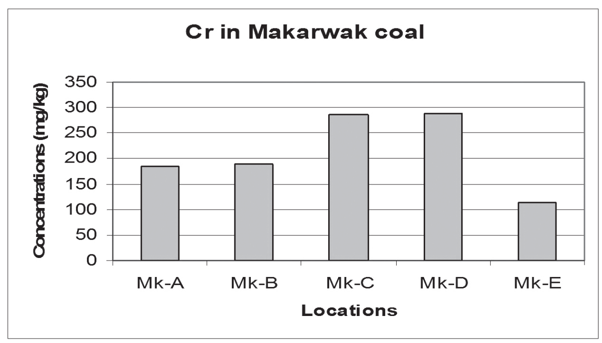

Fig. 5: Concentration of Chromium (Cr) in different coal fields of Makarwal

Concentrations of Cr in water sample of Makarwal area are given in the table 2. The sample results show that the Cr concentration was above the maximum contaminant level of the drinking water as prescribed by the USEPA and WHO.

Copper (Cu) is a necessary element in humans for normal biological activities (Siddiqui, 2008). It occurs in the form of minerals like Chalcocite (Cu_2S), Covellite (CuS), Chalcopyrite (CuFeS2), Malachite ($Cu_2CO_3(OH)_2$ and Azurite $2[Cu_3(OH)_2(CO_3)_2]$ (Sadiqui, 2008). According to WHO (2008), the highest guideline limit for Cu in water is 2 mg/l. Concentration of copper in water samples of Makarwal area are given in the table 2. The Table shows that the Cu concentration is below the maximum contaminant level of the drinking water as prescribed by the USEPA and WHO.

The Cu concentration in section-A, shown in figure 6, were found in the range 5.9 to 13.15 mg/kg (8.53 mg/kg), in section-B it ranges from 11.7 to 16.8 mg/kg (13.6 mg/kg), while in section C, D and E copper ranges from 8.45 to 33.9 mg/kg (mean 23.2 mg/kg), 8.15 to 42.0 mg/kg (mean 22.68 mg/kg), and 9.8 to 11.9 mg/kg (mean 11.03 mg/kg) respectively. Thus, this shows that on the average section-C had the highest contents of Cu as compared to other section of the study area as shown in the table 1 and figure 6.

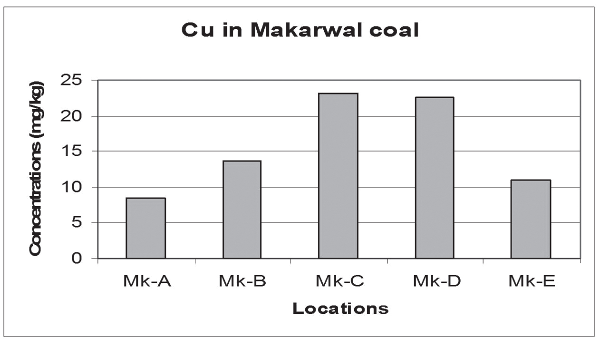

Fig. 6: Concentration of Copper (Cu) in different coal fields of Makarwal

Fe distribution is heavily regulated in mammals. The Fe concentration in section-A, shown in Fig. 7, was found in the range 2495-8760 mg/kg (4609 mg/kg), in section-B it ranges from 920 to 23515 mg/kg (10813 mg/kg), while in section C, D and E iron ranges from 860 to 19140 mg/kg (6880 mg/kg), 849 to 14200 mg/kg (5818 mg/kg), and 876 to 1469 mg/kg (1133 mg/kg) respectively. This shows that on the average section-B had the highest content of Fe as compared to other section of the study area as shown in the table 1 and figure 7.

Nickel (Ni) occurs in coal in both organic and inorganic mode (Finkelman *et al.,* 1991). Ni dominates in the organic fraction of coal, whereas traces are found in silicate and sulfides (Swaine, 1990). Similarly, Ni is also associated with the FeS minerals in coal in the range of 7-800 mg/kg (White *et al.,* 1989).

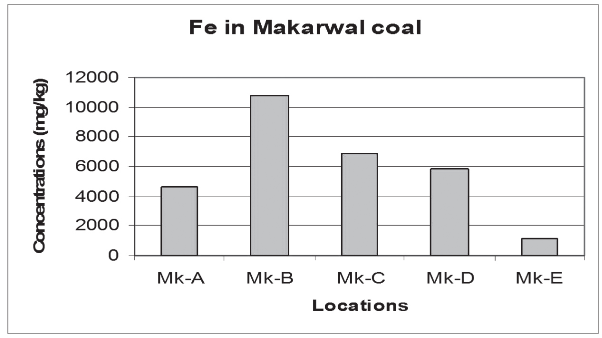

Fig. 7: Concentration of Iron (Fe) in different coal fields of Makarwal

The Ni contents in Makarwal water sample were found as 0.25 mg/l which is higher than the 0.07 mg/l recommended by the WHO (2008) for drinkable water as shown in table 2.

The Ni concentration in section-A, shown in figure 8, was found in the range 40-95 mg/kg (mean 62.0 mg/kg), in section-B it ranged from 39.9 to 47.5 mg/kg (mean 44.35 mg/kg), while in section C, D and E nickel ranges from 9.0 to 278.3 mg/kg (mean 110.95 mg/kg), 24.6 to 342.2 mg/kg (mean 148.8 mg/kg), and 18.9 to 60.45 mg/kg (mean 45.28 mg/kg) respectively. Thus, this shows that on the average section-D had the highest content of Ni as compared to other sections of the study area as shown in the table 1 and figure 8.

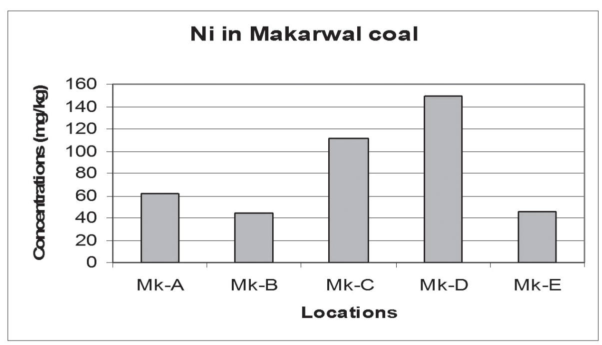

Fig. 8: Concentration of Nickel (Ni) in different coal fields of Makarwal

Concentration of nickel in water sample of Makarwal area is given in the table 2, that shows the nickel concentration is above the maximum contaminant level of the drinking water as prescribed by the WHO.

Lead (Pb) is considered to be inorganically bound elements in coal. The most abundant form of Pb in coal is galena or galena attached with pyrite (Finkelman, 1988). According to WHO, (2008) and USEPA, (2004) the maximum permissible limit for lead in drinking water is 0.01 mg/l as shown in table 2. Fig. 9, shows the Pb concentration in section-A, that was found in the range 9.0-31.15 mg/kg (18.46 mg/kg), in section-B it ranged from 23.36 to 35.5 mg/kg (28.13 mg/kg), while in section C, D and E it ranges from 19.9 to 61.85 mg/kg (46.25 mg/kg), 22.9 to 60.55 mg/kg (37.37 mg/kg), and 18.8 to 29.8 mg/kg (24.4 mg/kg) respectively. Thus, this shows that on the average section-C had the highest contents of Pb as compared to other section of the study area as shown in the Table 1 and Fig. 9.

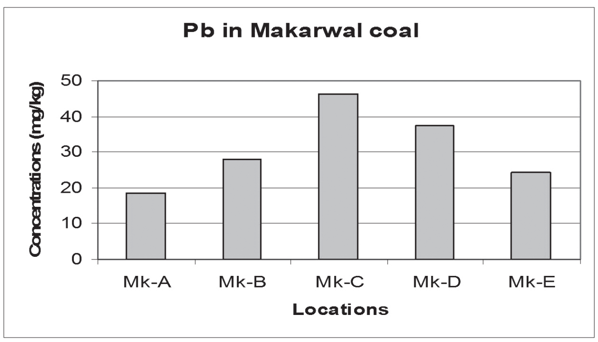

Fig. 9: Concentration of lead (Pb) in different coal fields of Makarwal

Concentration of lead in water sample of Makarwal area is given in the table 2, which shows that the lead concentration has surpassed the maximum contaminant level of the drinking water as prescribed by the USEPA and WHO.

Zinc (Zn) occurs naturally in air, water and soil, but Zn concentration increases unnaturally, due to the addition of Zn through human activities. Most Zn is added during industrial activities, such as mining, coal and waste combustion and steel processing. The most abundant sources of Zn are sphalerite (ZnS) and wlutzite (Fe.ZnS) (Reimann and deCaritat, 1998).

The Zn concentration in section-A were found in the range 11.75 to 52.5 mg/kg (33.6 mg/kg), in section-B it range from 21.35 to 70.4 mg/kg (39.0 mg/kg), while in section C,D and E it ranges from 15.55 to 29.2 mg/kg (25 mg/kg), 5.0 to 53.0 mg/kg (33 mg/kg), and 24.85 to 671.5 mg/kg (242 mg/kg) respectively. This shows that on the average section-E had the highest contents of Zn as compared to other section of the study area as shown in the table 1 and figure 10.

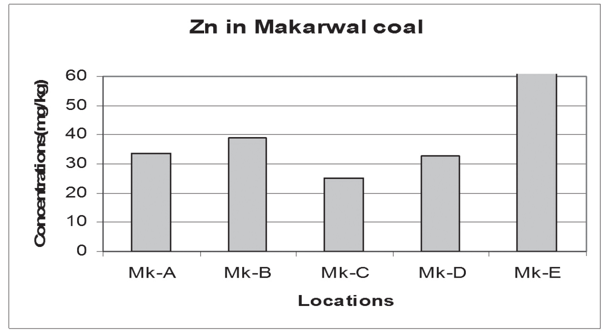

Fig. 10: Concentration of Zinc (Zn) in different coal fields of Makarwal

Concentration of zinc in water sample of Makarwal area is given in the table 2. This table shows that the zinc concentration is above the maximum contaminant level of the drinking water as prescribed by the USEPA and WHO.

Trace and heavy elements in water samples

The heavy and trace elements detected in water samples collected from hand pump, spring water and waste water discharge from coal mines are presented in table 3. The results were compared with the standard values of WHO and USEP, recommended for maximum permissible limits (WHO and USEP, 2008). The heavy and trace elements content were found less in hand pump water samples of all locations (Mk-A, Mk-B, Mk-D, Kr-C and Gk-E) than such contents in spring and waste water samples. This may be due to deep boring and distant apart of hand pumps water from coal mine sites while spring and waste water was relatively closed to the coal mines sites.

As depicted in table 3, Arsenic (As), Chromium (Cr) Nickel (Ni) and Zinc (Zn) were found within the desirable limits set by WHO and USEP in all sample of water taken from hand pump and spring water except at location Gk-E Where As (13.4 mg/kg), Cr (603mg/kg), Ni (93mg/kg) and Zn (6270 mg/kg) were above permissible limits. However, these elements were found significantly higher in waste water discharge from coal mines, which may be due to the leaching of minerals from coal mines, and indicate pollution in drinking water in future. Cadmium

Table 1: Mean concentration of heavy and trace element in various sections of Makarwal coal fields.

Elements		Mk-A	Mk-B	Mk-C	Mk-D	Mk-E	Mean Range
As (mg/kg)	Range	(0.75-8.46)	(2.18-38.49)	(1.36-5.23)	(13.29-19.5)	(2.3-3.8)	(3.0-16.34)
	Mean	3.29	15.93	3.0	16.34	3.1	
Cd(mg/kg)	Range	(2.9-5.95)	(2.0-5.35)	(2.15-7.05)	(1.35-7.0)	(2.0-2.45)	(2.23-4.61)
	Mean	4.1	3.31	4.39	4.61	2.23	
Co (mg/kg)	Range	(13.9-28.35)	(14.8-40.1)	(9.1-40.45)	(22.7-44.85)	(23.7-37.3)	(22.1-29.3)
	Mean	22.1	28.26	27.66	29.3	28.25	
Cr (mg/kg)	Range	(108.45-329.1)	(106.75-270.1)	(13.7-631.5)	(102.75-478.45)	(70.1-191)	(113.1-288.2)
	Mean	183.75	189.48	286.7	288.2	113.12	
Cu (mg/kg)	Range	8.53 2495-8760	13.6 (920-23515)	23.2 (860-19140)	22.68 (849-14200)	11.03 (876-1469)	(8.53-23.2)
Fe (mg/kg)	Mean	4609	10813	6880	5818	1133	(1133-10813)
	Range	(40-95)	(39.9-47.5)	(9.0-278.3)	(24.6-342.2)	(18.9-60.45)	
Ni (mg/kg)	Mean	62.0	44.35	110.95	148.8	45.28	(44.35-148.8)
	Range	(9.0-31.15)	(23.36-35.5)	(19.9-61.85)	(22.9-60.55)	(18.8-29.8)	
Pb (mg/kg)	Mean	18.46	28.13	46.25	37.37	24.4	(18.46-46.25)
	Range	(11.75-52.2)	(21.35-70.4)	(15.55-29.2)	(5.0-53.0)	(24.85-67.15)	
Zn (mg/kg)	Mean	33.3	39.0	25	33	242	(25-242)

Table 2: Comparison of Water Quality of the Study Areas with the International Standards.

Metals	Makarwal water mg.l	WHO Standard mg/l	USEPA Standard mg/l
As	0.012	0.01	0.05
Cd	0.131	0.003	0.01
Co	0.065	ND	ND
Cr	0.11	0.05	0.05
Cu	0.592	2	1.3
Fe	5	2	0.3
Ni	0.25	0.07	ND
Pb	0.173	0.01	0.01
Zn	11.90	3	5

was found below the detection limit in hand pump and spring water samples of all locations, whereas, in waste water discharge samples it was found higher than the permissible level. Copper (2460 mg/kg) (4520 mg/kg) and Nickel (92mg/kg) (93mg/kg) were found higher at locations Kr-C and Gk-E than the desirable limits. Lead (Pb) was found within the permissible level set by WHO and USEP as shown in table 3, in hand pump and spring water samples of all locations except location Kr-C (15.30 mg/kg) but was found high in water discharge from coal mining sites as shown Table 3.

In context to the parameters studied, the water collected from hand pumps of nearby villages was safe for domestic uses. However, due to proximity of the mine,

Table 3: Heavy and trace elements present in water samples collected from mines sites as well as from hand pump and spring water near coal mining site of Makarwal region (ppb)

Trace elements	Hand pump water					Spring water near coal mining sites					Waste water leached out from the mines					EPA standard	WHO Standard
	MkA	MkB	MkD	Krc	GkE	MkA	MkB	MkD	Krc	GkE	MkA	MkB	MkD	Krc	GkE	-	-
As	BDL	1.3	BDL	3.0	24	7.8	9.2	8.0	8.1	13.4	159	128	174	214	241	5	10
Cd	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	5.6	131	520	125	825	169	10	3
Co	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	432	832	1120	1100	9550	8632	ND	ND
Cr	25.6	BDL	8.0	40	45	53	46	90	BDL	603	1891	1250	7500	6500	4000	50	50
Cu	782	650	BDL	830	BDL	BDL	850	3820	2460	4520	6530	12700	4490	6760	19210	1300	2000
Fe	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Ni	BDL	BDL	BDL	BDL	BDL	58	65	32	92	93	15000	13000	25300	45200	53000	ND	70
Pb	BDL	BDL	BDL	BDL	1.30	BDL	9.50	BDL	15.30	6.46	1320	1080	2370	2950	9180	10	10
Zn	1430	130	890	183	234	925	1423	102	1042	6270	1542	8450	6325	4350	7325	500	3000

characteristics of the underground water are likely to be affected in near future.

CONCLUSION

Continuous mining and combustion of coal disperse toxic elements into the environment and thus pollute the surface and ground-water resources of the area. This study has satisfied the objectives by analyzing Makarwal coal deposits for heavy and trace metals. Thus, proper pollution control arrangements are required to be made, since these coals are expected to give rise to huge amount of toxic pollutants during the burning, because the ash content of the coals is very high. The results for heavy and trace elements can be concluded that Makarwal coal deposits are a relatively cleaner area. Maximum metals materials show no possibility of adverse effects on the local environment. These metals, however, must be monitored thoroughly to ensure that they stay at harmless levels. This study suggests that currently, the coal fields have no deleterious effects; however, these elements may be leached into local water bodies and when ingested, may influence the public health and surrounding environment in future.

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