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The Power Generation from Coal in Pakistan: Assessment of Physicochemical Pollutant Indicators in Indigenous Reserves in Comparison to the Foreign Coal

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Abstract

Electricity production through coal combustion is the only viable solution in minimum timing. As environmental chemists, our primary goal is to assess environmental hazards and suggest cost-effective technologies for reducing combustion pollutants. In the present study, indigenous coal samples from different mines were analyzed for their physicochemical properties and toxic metals. Five samples from foreign coal mines were also studied for comparison purposes and already in use for power generation. The sulfur content in Thar coal (0.62 %) is similar to foreign samples of Indonesian and Mozambique samples (0.35 – 1.63 %). Heating values of local coals show their potential as future fuel. The principal component analysis was applied to the data. It interprets that the concentration of toxic metals in indigenous and foreign samples is lower than the reported Greece samples. The concentration of metals in Badin coal samples is similar to foreign studied samples and is lower than the reported China, South Africa and other local samples. Copper (77.64 mg/kg), Zinc (63.23 mg/kg), Chromium (75.80 mg/kg), Mercury (0.22 mg/kg) and Manganese (119.07 mg/kg) are found to be high in Thar and Lead (49.41 mg/kg) in Lakhra. Balochistan is elevated in the concentration of nickel (52.63 mg/kg). It becomes obligatory for the policy makers of the country to suggest legislation for the use of appropriate Clean Coal Technologies (CCT) in the use of the coal in power sector.

Keywords: Indigenous coal, Power generation, Trace metals, Pollutants, CCT, Clean coal technologies.

Introduction

Pakistan is facing an acute crisis of electricity since the last decade. The total shortfall of the country is about 5000 Megawatts (MW). The electricity generation capacity of the country is heavily dependent on either natural gas or imported furnace oil. Pakistan is rapidly depleting natural gas reserves. On the other hand, the price of furnace oil is uncertain in the international market.

Coal is the most plentiful fossil fuel in the world. The global reserves of all types of coal are estimated to be 990 billion tons. The International Energy Agency (IEA) report indicates that the global usage of Coal for the power generation is about 42 % [1].

Energy can be extracted from coal through its combustion process. It involves three basic steps. Firstly, release of volatile compounds, followed by burning of these compounds and finally burning of the char. The sequence of these steps very much dependent on specific combustion condition. The nature of volatiles and composition of the char depends upon the nature of raw coal and its geological origin and chemical composition.

Pakistan has huge coal deposits. According to the BP Statistical Review of World Energy June 2013, the proven reserves of the country are 2070 million tonnes. However, after the exploration of the Thar coalfield, Sindh province, Pakistan, coal scientist estimated 175 billion tonnes resources of good quality lignite in the field [2]. The country first coal fired power plant was established in 1995 of 150 MW electricity generation capacity at near the Lakhra coalfield. It has the largest proven reserves of the country. However, due to a poor maintenance of the plant, its capacity has been now dropped to few MW. Some concrete steps have now been taken at the government level to overcome the country worst energy crises [3]. Various agreements and a Memorandum of Understanding (MoU) are signed for the establishment of electricity generating power plants based on imported and indigenous coal [4]. The use of coal for energy production has documented environmental concerns [5]. Therefore, assessments of physicochemical properties of the probable fuel are imperative, keeping in view the possible environmental hazards to nearby population in the future. This study will also be helpful to manage the emissions and to propose an appropriate Clean Coal Technology (CCT) for the power generation plant.

The objectives of the study are two-fold: the primary object of the study is to analyse the coal samples of four local mines for toxicity assessment. The composition parameters (proximate, ultimate, calorific value and trace metal) of indigenous samples are used for this purpose. The secondary object of the study was the comparison of local coal samples with the foreign coal samples, already being used in the different places of the world. The comparison was made to evaluate the potential utilization of indigenous coal reserves for the environment friendly utilization in the power generation. Trace elements and quality of coal, lead us to evaluate their environmental and health impacts during its usage as a fuel. The other aspect of the present work is to interpret the concentration of toxic metals in studied samples with the reported work by applying chemometric techniques. The outcome of this study will not limit to local consumption, but also useful in a regional environment.

Materials and Methods

Some local and foreign coal samples were selected for their characterization. Indigenous coals were collected from a local coal miner and foreign coals were obtained from importers. After the sample has been collected, it is sealed in a container to prevent moisture loss or gain. The samples were air dried [6] and ground into fine powder of particle size 60 mesh [7]. Safety goggles, ear protection, a laboratory coat,and an air-filter mask was be used at all times during sample handling and preparation. The results are an average value of five representative samples of each coalfield. The sample profile comprises of nine coal samples, four from local origins, including Lakhra, Thar, Badin, Balochistan and the remaining five from foreign countries which are Australia (The Callide basin and The Meandu coal mine), Indonesia (South Sumatra Basin), Mozambique (Moatize coal mine) (samples are representative of their respective coal mines). Australian and Indonesian samples are already in use for the power generation and the boiler industries in their own countries. Mozambique has plans to utilize its Moatize coal resources for the power generation [8]. The results are an average value of five representative samples of each coalfield. The Proximate analysis was determined [9]. The coal samples were analyzed for calorific value [10] by Isoperibol bomb calorimeter (Parr 6300, USA) and sulfur content [11] by SC-32 LECO sulfur determinator. Ultimate analysis, including total Carbon (C), Hydrogen (H), Nitrogen (N) and Oxygen (O) were calculated from the described formula [12]. The analysis of heavy metals was performed by atomic absorption spectrophotometer (Perkin Elmer 3300) [13,14,15]. The detection limit for Mercury and Arsenic is 2ng/g and for other metals is 0.1µg/g respectively. The Statistical analysis was applied using the software Minitab and XLSTAT.

Results and Discussion

Proximate and ultimate analysis

Table 1. shows proximate, ultimate and calorific values of coal samples. The low moisture content is observed in Australian (II) and Mozambique coal sample reflecting their higher heating value. Moisture plays an important role in

coal quality and handling characteristics. High moisture in coal takes more time of heating and has a lower calorific value. With the demerits of high moisture content there is also a beneficial aspect. A mixture of low-rank coal, water, and a chemical stabilizing agent is given the name Coal water fuel (CWF). It can use as in place of oil in boilers and for other applications in industries. Approximately, all the local samples have high moisture content which can be used as a CWF. The highest ash content is observed in Thar and Badin and lowest in the Indonesian 1 sample. The ash content in other local and foreign samples is comparable. The ash is a waste product, formed in the process of coal combustion. The environmental concern is not only limited to emission, but also the disposal of waste produced due to presence of toxic metals in the ash.

Samples Lakhra, Badin and Balochistan show the highest level of sulfur content in contrast, low- intermediate concentration is found in other local and foreign samples. The sulfur is termed as one of the criteria pollutants due to releases of the SO_x during combustion and it causes various health and environmental hazards. Therefore, in any future power plant in the country, there must be the usage of low sulfur coal. Lower cost method of reducing the sulfur from coal and make it environmentally friendly is washing of coal before combustion. It reduces up to 50% of sulfur, associated with pyrite. Flue Gas Desulfurization (FGD) and Electro Static precipitators (ESP) are used to remove high sulfur content in coal [16].

Australian II and Mozambique samples show the lowest, whereas Lakhra, Thar, Badin and Indonesian 1 have higher volatile matter. High volatiles in coal assess the ignition and combustion properties. It assists the ignition and reactivity of coal towards combustion [17]. Being the high volatile matter in local coals makes them more suitable for the underground gasification process. However, in the combustion process; due to heavy releases of gases there should be an effective CCT to minimize gas emission.

Fixed carbon is the solid fuel left in the furnace after the volatile matter is distilled off. The percentage of fixed carbon in indigenous and foreign samples ranges 24.7-42.07% and 34.8-55.17% respectively. Thar in local samples and Australian I in foreign samples comprise of lowest fixed carbon. The highest percentage of fixed carbon (61.63 %) is obtained in Balochistan coal in the local samples. Mozambique coal shows the highest percentage of fixed carbon (61.40 %) in the foreign samples. The quantity of fixed carbon present in coal decides the ranking of coal. The higher the fixed carbon and heating values, higher will be the rank of coal [18]. The same pattern is also observed in the local and foreign samples (Table 1).

The quantity of carbon, hydrogen and oxygen are very vital to evaluate the characteristics of coal for gasification, coking and liquification purposes. In the studied samples Thar has the lowest value of C (33.29%) followed by the Badin (41.8%). Balochistan, Australian II, Indonesian 1 and Mozambique range 61-64% of carbon. It is evident that the samples containing higher carbon contents are more heating values. This causes an emission of CO_2 in the atmosphere at a significant level [18]. The hydrogen content is almost the same in all the samples. The H/C ratio in the coal has significance in coal conversion processes such as liquefaction or gasification. The high oxygen content is found in Australian I, Thar, Indonesian 2 and Indonesian 1 in a range of 20.4-23.43%. The higher oxygen content reduces the heat production ability of coal during spontaneous combustion. The studied samples contain higher oxygen and hence lower heating values. Nitrogen is responsible for the NO_x production during coal combustion, which is one of the major pollutants to the environment. The SO_x and NO_x are called secondary particulate matter, because after the formation, they are associated with $PM_{2.5}$ [19]. These oxides, upon addition to water in the atmosphere, a result, in the precipitation of acid, sometimes also called acid rain. Acid rain is often prevalent downwind from coal burning power plants, indicating the connection between acid formation and airborne emissions caused by coal-fired power plants. The NO_x is a serious threat to China due to its increase during the last two decades. China, Germany and Japan are already using SCR (Selective Catalytic Reduction) to decrease NO_x emission [16].

Table 1. **Proximate, Ultimate analysis and Calorific value of the Local and Foreign samples (On air dried basis). Results are an average value of five representative samples of each coalfield.**

Samples	Proximate Analysis				Ultimate Analysis					Calorific value
	Moisture %	***Ash*** %	***Volatile Matter*** %	***Fixed Carbon*** %	***C*** %	***H*** %	***N*** %	***O*** %	***S*** %	**kJ/kg**
Lakhra	8.60 ±0.90	15.30 ±0.80	41.60 ±1.0	34.50 ±2.0	55.74 ±0.60	5.38 ±0.01	1.60 ±0.05	15.83 ±0.09	6.15 ±0.02	20212.90 ±120.0
Thar	7.57 ±0.10	38.70 ±0.20	42.10 ±3.0	11.63 ±0.20	33.29 ±0.10	5.39 ±0.05	1.59 ±0.02	20.41 ±0.05	0.62 ±0.03	13644.02 ±110.0
Badin	7.20 ±0.20	31.10 ±0.40	37.00 ±1.5	24.70 ±0.10	41.80 ±0.20	5.31 ±0.01	1.66 ±0.09	15.51 ±0.06	4.62 ±0.09	17204.61 ±150.0
Balochistan	9.43 ±0.30	8.80 ±0.10	39.70 ±1.4	42.07 ±2.50	61.63 ±1.00	5.36 ±0.09	1.62 ±0.01	16.44 ±0.01	6.15 ±1.0	24078.92 ±120.0
Australian I (Brown)	11.30 ±0.50	9.30 ±0.70	44.60 ±1.6	34.80 ±1.50	58.74 ±0.50	5.42 ±0.08	1.56 ±0.03	22.67 ±0.09	2.37 ±0.05	21417.90 ±130.0
Australian II (Black)	4.23 ±0.10	15.60 ±0.20	28.60 ±2.0	51.57 ±0.10	61.14 ±1.50	5.07 ±0.01	1.76 ±0.07	15.2 ±0.01	1.22 ±0.04	27191.82 ±200.0
Indonesian 1 (Black)	10.23 ±0.15	4.57 ±0.10	41.00 ±1.5	44.23 ±0.30	64.90 ±2.00	5.38 ±0.02	1.61 ±0.08	21.91 ±0.02	1.63 ±0.07	26865.46 ±160.0
Indonesian 2 (Brown)	10.77 ±0.20	16.03 ±0.05	37.90 ±1.9	35.27 ±0.70	53.21 ±0.70	5.32 ±0.09	1.65 ±0.05	23.43 ±0.06	0.35 ±0.02	21175.22 ±115.0
Mozambique	4.47 ±0.40	15.36 ±0.20	25.00 ±1.0	55.17 ±0.10	61.40 ±1.00	4.93 ±0.01	1.80 ±0.06	16.06 ±0.01	0.45 ±0.02	27564.19 ±190.0

Table 2. **Trace metal concentration of the local and foreign coals (All concentrations are on air dried basis, bdl - below detection limit). Results are an average value of five representative samples of each coalfield.**

Samples	As mg/kg	Cr mg/kg	Cu mg/kg	Hg mg/kg	Mn mg/kg	Ni mg/kg	Pb mg/kg	Zn mg/kg
Lakhra	0.03±0.01	21.32±0.2	33.20±0.3	0.07±0.02	29.24±0.3	18.20±0.7	49.41±1.2	19.26±1.0
Thar	0.42±0.02	75.80±1.6	77.64±1.1	0.22±0.05	119.07±0.7	52.63±0.5	10.14±0.1	63.23±2.0
Badin	0.08±0.01	19.62±0.3	34.42±0.4	0.05±0.01	68.91±0.3	33.62±0.1	1.87±0.01	15.70±0.5
Balochistan	bdl	18.62±0.2	18.23±0.3	bdl	29.70±0.4	100.60±1.5	1.35±0.03	35.42±0.1
Australian I brown	0.42±0.03	26.75±0.2	4.94±0.4	bdl	29.70±0.1	9.70±0.1	3.03±0.05	7.75±0.09
Australian II Black	0.47±0.02	12.90±0.4	8.93±0.4	0.01±0.001	57.94±0.9	16.61±0.5	10.01±0.1	15.69±0.05
Indonesian 1 (Black)	bdl	8.17±0.4	2.19±0.2	bdl	12.92±0.2	3.30±0.07	0.27±0.01	6.08±0.03
Indonesian 2 (Brown)	bdl	26.34±0.3	8.94±0.4	bdl	51.25±0.15	16.12±0.08	0.75±0.01	18.52±0.05
Mozambique	bdl	11.34±0.3	16.61±0.4	0.02±0.005	42.62±0.1	20.72±0.02	3.80±0.02	38.15±0.08
World coal [28]	5.0	10.0	15.0	0.012	50.0	15.0	25.0	50.0
Chinese coal [35, 36]	5.0	12.0	13.0	0.15	77.0	7.0	13.0	35.0
US averages [37]	24.0	05.0	16.0	0.17	43.0	14.0	11.0	53.0

Trace metals

Table 2 represents the results of metals in local and foreign samples on an air dried basis and also the reported values of World [28], Chinese [35,36] and US [37] coal average.

Arsenic (As)

Arsenic is termed as a prime environmental sensitive element. It is an everlasting poison in the body, even at trace level exposure [20]. It causes the cancer of different body parts and skin lesions and affects mental health, pulmonary function and immune functions [21]. Among the entire studied local and foreign samples the detectable concentration of arsenic is only in Thar, Badin and Australian samples. Arsenic contents in the samples are lower than the World coal average [28], US [37] and Chinese coal [35, 36]. Coal mining and combustion cause the leaching of arsenic resulting in contamination of soil, air and water, which can raise the health hazards [22]. It can also damage the catalyst used in SCR technology for the removal of NO_x level [23]. The proper handling of coal is required even in lower concentrations of arsenic.

Chromium (Cr)

As shown in Table 2, all the samples are in a higher concentration of chromium than World coal average [28] except Indonesian 1. The Thar sample is enriched with chromium content. The biologically important forms of Cr are Cr (III) and Cr (VI) and according to their toxicity characteristics both the forms are distinguishable. Cr (VI) is termed as carcinogenic whereas, Cr (III) is essential for the adult body in the range of 50-200 mg/day. It is required in the process of insulin metabolism [24]. The abundance of Cr in coal is in the form of trivalent, but during combustion the oxygen environment converts half of the Cr in the hexavalent form. It is water soluble and its presence in the fly ash increases its toxicity due to leaching in a nearby water system and soil.

Copper (Cu)

The exposure of Cu beyond permissible limits causes the gastrointestinal distress and can damage the liver or kidney. All the local and Mozambique coal samples are found to be higher in copper concentration. The Cu content is highest in the Thar sample. Global emission of Cu from coal combustion lies in the range of $2.3\text{-}8.0\text{x}10^6$ kg/year, which is higher than all the anthropogenic combustion sources like oil, wood, etc. [25]. According to the [26], Cu is considered as prime metal for environmental concern. The major association of Cu is with pyrite in the form of chalcopyrite ($CuFeS_2$).

Lead (Pb)

Among all the local and foreign samples the Lakhra coal is predominantly high in Pb concentration (49.41mg/kg). This concentration is even more than World coal average [28]. Lead is listed in metal which shows the higher enrichment factor in fly ash and liberated into the atmosphere during combustion [27]. According to the [28], the Pb can be associated with minerals and sulfides. Irrespective of the organic or inorganic forms of Pb, the higher amount can cause adverse effects on all the spheres of environment. Conventional efficient particulate control devices, such as ESP or fabric filters can be used to reduce the Pb emission into the atmosphere.

Manganese (Mn)

The Thar, Badin, Australian II and Indonesian 2 are rich with Mn as compare to World coal average [28]. All the above samples have lower concentrations than the Chinese [35,36] average values (77.0mg/kg) except Thar (119.07mg/kg). Manganese is the essential element under the allowable limits, but toxic when exposed in higher concentrations. The Mn toxicity varies with route and period of exposure, whether it is ingested or inhaled. The inhaling of Mn has profound effects on the brain [29]. From the combustion waste, Mn can leach into a water system near coal power plants which can cause its elevated concentrations in all the geo-spheres. Prior steps like washing and cleaning of coal before combustion can decrease the Mn content [26].

Mercury (Hg)

Lakhra, Thar, and Badin show the presence of Hg metal in the range of 0.05-0.22mg/kg. 0.02 and 0.01mg/kg is detected in Mozambique and

Australian I samples respectively (Table 2). Although the concentration of Hg in the samples is higher than World coal average [28], but it is lower than the Chinese [35, 36] and US coals [37]. Mercury is classified in metals which are volatile in nature and emitted to an atmosphere during combustion. And due to high residential time in the atmosphere, Hg and As may cause serious health problems associated with nervous, digestive and immune systems, and on lungs, kidneys, skin and eyes [30]. The liberated Hg contaminates land, water, aquatic life and ultimately to human beings in elevated concentration causes toxic effects to their health [31].

Nickel (Ni)

Only the Australian I and Indonesian 1 have the lowest concentration of Ni with respect to World coal [28], the US [37] and Chinese coal averages [35,36]. The highest concentration is observed in Balochistan (100mg/kg). As, Be, Cd, Cr, Ni, Se, Mn and Pb are the elements termed as non-mercury hazardous air pollutants. They release into the atmosphere with the aerosols that contain the size lesser than 2.5 micrometers ($PM_{2.5}$). These fine matters possess its own hazardous to human and environment and after the addition of these metals elevate its hazardous nature to a higher extent [32]. According to the [33], approximately one million deaths occurred around the US, caused by the elevated amount of Ni in the $PM_{2.5}$ along with other metals and sulfates. These can be controlled by using ESP and after further treatment, it can be disposed. It is inherited in the coal during the coalification process.

Zinc (Zn)

The higher concentration of Zn is found in Thar than the other studied and World coal average [28]. Zinc is characterized as an element of primary environmental concern [26]. Zinc is the important metal for the body, but too much exposure causes serious effects to health. Coal combustion contributes 5-20% of global emissions of Cu, Sn, TI and Zn [25]. Zinc is mostly associated with sphalerite and pyrite. The presence of Zn along with particulate matter $PM_{2.5}$ in the atmosphere increases its adverse effects.

Principal component analysis (PCA)

In the present study the PCA is applied on the trace metals (Hg, Zn, Pb, Cu, Ni, As and Cr) present in the studied local and foreign coal samples. To make the interpretation more elaborate, PCA is also applied to the previously reported values of coal from China [38], South Africa (SA) [39] and the Elassona basin, Central Greece [40] (Table 3).

Table 3. **Comparisons with average Chinese, South Africa and Greece coal.**

Trace Metals (mg/g)	**China**			**South Africa**						**Greece**
	[38]			***[39]***						***[40]***
	1	***2***	***3***	***1***	***2***	***3***	***4***	***5***	***6***	
As	3.79	5.0	3.8	3.74	2.36	2.76	2.8	2.84	2.66	32.06
Cu	17.50	13.0	18.35	13	14.3	12.7	11.6	12.8	16.4	119.09
Ni	13.7	15.0	13.71	20	16	14.83	16.8	23.5	21.0	88.43
Hg	0.163	0.1	0.19	0.22	0.178	0.21	0.1	0.16	0.23	0.34
Pb	15.1	14.0	15.55	7.44	5.92	6.38	9.96	6.61	15.34	57.12
Zn	41.4	38.0	42.18	10.1	15.5	10.8	18.7	13.30	22.0	176.44
Cr	15.4	16.0	15.35	53.8	46.5	46.5	49.2	59.8	64.6	119.33

Table 4. **Factor matrix for trace metal levels of the first two factors studied for Greece, Studied samples, China and South Africa.**

Variables	Factors	
	1	*2*
Cu	0.418	-0.042
Ni	0.408	-0.085
Zn	0.385	-0.321
Pb	0.392	0.028
Hg	0.226	0.897
As	0.363	-0.256
Cr	0.419	0.133

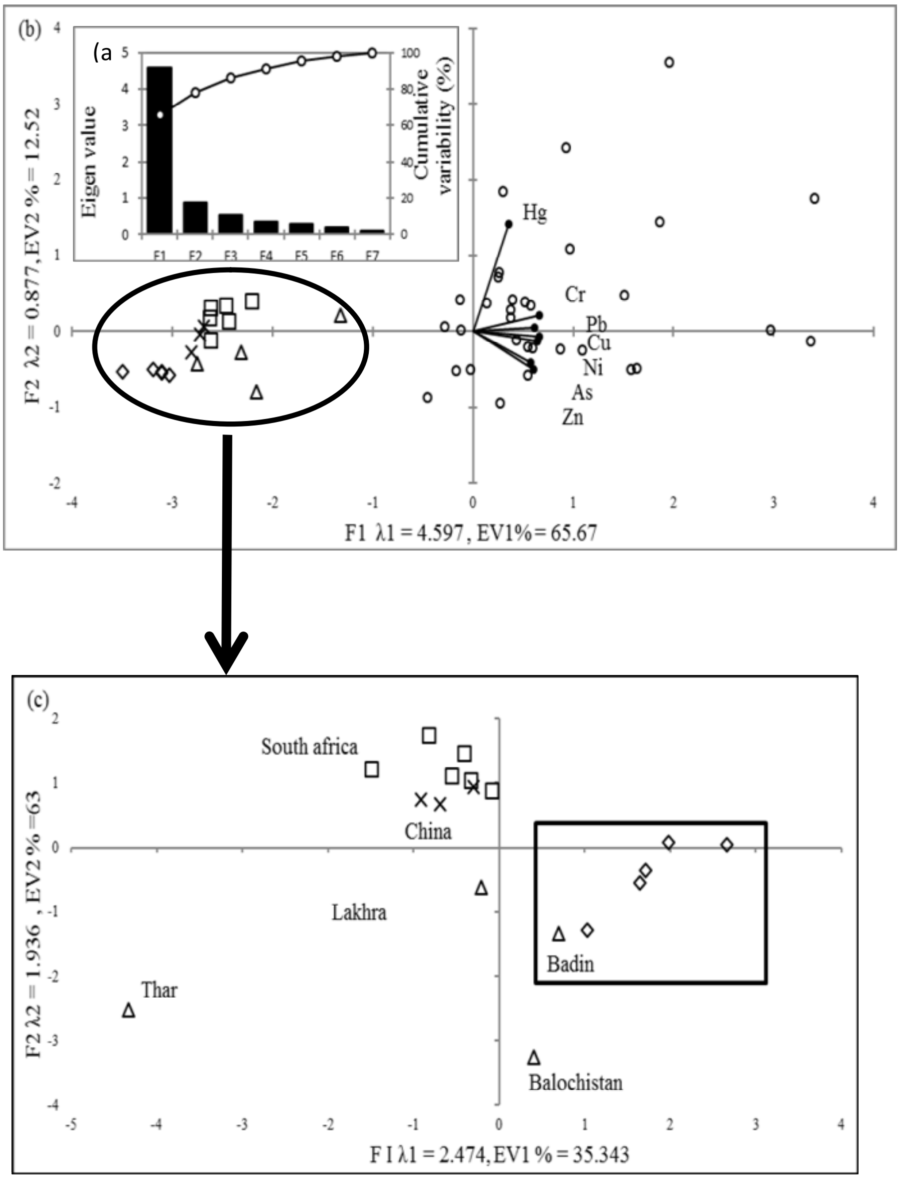

Figure 1. **Principal Component plot between F1 and F2 for Copper (Cu), Nickel (Ni), Zinc (Zn), Lead (Pb), Mercury (Hg), Chromium (Cr) and Arsenic (As) in studied samples and reported values. (a) Scree plot (b) Biplot (c) Score plot between studied, China and South Africa. λ1and λ2 are Eigenvalues and EV1 and EV2 explain Percent Variance. ∆ Local, ◊ Foreign, × China, □ South Africa and o Greece**

The Eigen value of the first factor (F1) of the PCA is greater than 1 and the second factor (F2) comprises of 0.87. The first two factors had chosen due to the % cumulative of 78.19 as shown in (Fig.1a) [34]. The PCA described the 65.67% variability for F1 and it strongly correlated with Cu, Ni, Zn, As, Pb and Cr (Table 4). With the increasing value of F1 in the biplot in Fig 1 (b), there would increase in concentration or higher enrichment of metals in locations. As shown in the (Fig. 1b), China [38], South Africa [39], all studied samples and some samples of Elassona Basin,Greece [40] are separated towards the left side of the plot showing the lower concentration of Cu, Ni, Zn, As, Cr and Pb. 12.52% variability found in F2 with a predominant function of Hg (Table 4). It is apparent from the score and the loading plot that the Hg concentration is higher in a fraction of the samples of Elassona basin, Greece [40]. The applied PCA interprets that local samples have a lower level of toxic metals compare to reported Greece mine [40]. Although local samples are grouped with China [38] and South Africa [39], but it differs with China and South Africa within the limits of these metals (Table 3). Due to this fact PCA is further applied within the concentration range of these samples. The sub grouping of samples is demonstrated in (Fig.1c). The concentration of metals in the foreign samples is lower than others and Badin grouping with foreign shows its similar behavior towards the toxicity of metals. Other regions are split due to higher concentrations of metals. Thar and Lakhra are separated because of the higher concentration of Cu, Zn, Cr, Hg and Pb respectively. Isolation of Balochistan is due to Ni. The Elevated concentration of arsenic results in separation of China and South Africa.

Conclusion

The exploitation of coal reserves is not limited to their combustion for electricity generation, but extracurricular steps should be taken to make their safe usage and environmental friendly. Indigenous coals form different mines have a tendency to be the future fuel in Pakistan owing to their ranking. Sulfur content is elevated in local samples. Thar has the sulfur content which is comparable to foreign samples. The comparison of local with foreign and reported coals evaluates that Thar has the highest concentration of Cr, Cu, Mn, Hg and Zn. But Hg content is comparable to reported values of South Africa and China. Lakhra and Balochistan are enriched in Pb and Ni content respectively. The foreign coal samples of Australia and Indonesia have lower concentrations of metals and these coals are used for electricity generation and other energy purposes. Badin resembles the foreign samples in toxic metal contents. This property of Badin coal can be exploited to overcome the energy crisis in Pakistan. The other local samples can also be used for power generation and other combustion purposes along with the technologies to lesser the dirtiness degree of coal and its hazardous impacts on the environment.

For the exploitation of indigenous coal reserves in coal-based power plants, the extracurricular steps should be taken to make their environmental friendly utilization. The policy makers should ensure the use of the Clean Coal Technologies (CCT) in the upcoming projects of power generation to minimize the hazardous impacts on the environment and human life.

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