
Major and some trace elements concentrations of Miocene-aged Alpu coals, Eskişehir, Turkey

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Abstract: Elemental contents, organic matter, and relation with Miocene-aged coal samples were analyzed in Alpu, Eskişehir, Turkey. The coal samples were identified as upper and lower seams in the m2 series. This series includes the following from the bottom to the top: claystone–marl, coal lower seam, sandstone, siltstone–claystone, upper coal seam, and claystone–sandstone–gravelstone. A total of 17 coal samples (eight and nine from the upper and the lower seam) were collected from five boreholes. Proximate and element analyses of the samples were performed and evaluated. In an air-dried basis, the upper and the lower seam have low average moisture contents (about 8% and 6%), average moderate ash yields (about 31% and 28%), and high total average calorific values (about 3215 and 2934 kcal/kg). The major elements found in Miocene-aged coal in this area are Na, Mg, Al, Si, P, K, Ca, Ti, Mn, Fe, and S. Compared with the average world coal content, major elemental contents of the two seams have highly enriched levels. Some trace elemental contents (e.g., V, Cr, Ni, Cu, Zn, Rb, Sr, Ba, Pb, Zr, and As) of the studied coal samples indicate similarity between the two coal seams, which are highly depleted with respect to the world average. The Al and Si contents of both coal seams have positive correlations with the ash yield, whereas those of Mg, Na, Ca, and Fe have negative correlations. The concentrations of Ni and Ba in these seams are positively correlated with the ash yield, and that of Sr and Zr have negative correlations.

Keywords: Coals, Proximate Analysis, Major and Trace Elements, Miocene, Alpu, Turkey

1. Introduction

Coal is a sedimentary rock associated with a complex heterogeneous mixture of organic and inorganic matter. Organic matter in coal comprises preserved or permineralized ancient plants or their fragments [1, 2, 3, 4, 5]. Each coal deposit has individual plant precursors, as well as specific regional, depositional, and paleoenvironmental conditions. However, these deposits can cause enrichment or depletion of different elements, phases, or minerals [6]. Most elements have mineral matter affinity in coal, but certain elements are associated with organic matter. Elements having inorganic affinity become affected by combustion, but are mostly concentrated in ash [7, 8]. This study aims to analyze the major and trace element concentrations of Miocene-aged coal samples in Alpu, Eskişehir, Turkey. Previous work has analyzed the geological setting, coal geology, and economical potential of this area [9, 10, 11]. In the Alpu coals, average total

moisture, ash, fixed carbon contents, and total calorific values in the upper coal seam are higher than those in the lower seam. However, the average value of volatile matter content in the lower coal seam is higher than in the upper seam. Al and Si contents in both coal seams have positive correlations with ash yield. These elements are generally associated with inorganic components. The ash yields in the lower coal seams have positive correlations with K, P, Ti, and S. These elements are dominantly and inorganically associated. However, these contents in the upper coal seam have negative correlations with the ash yield. Ash yields of the coal samples in upper and lower seams have negative correlations with Mg, Na, Ca, and Fe. The concentrations of Ni and Ba in coal samples from both seams are positively correlated with the ash yield. These elements have inorganic affinity. The ash yields in coal samples from the upper seam have positive correlation with Zn, Cu, Cr, and V, suggesting a mineral matter affinity. Zn is mainly associated with the inorganic components in coal [12, 13,

14]. The organic associations of Zn have also been determined in low-ranked coals [15, 13, 16]. Cu and Cr in coal have organic affinity [17, 13, 8]. In addition, V in coal is associated with both inorganic and organic matter [17, 13]. The concentrations of Sr and Zr in both coal seams are negatively correlated with the ash yield, showing the association of these elements with organic matter. Zr is commonly associated with organic matter [17, 13, 8]. Sr is mainly found together with mineral matter, and is associated with organic matter in low-ranked coals [17, 13, 18, 8].

2. Geological Setting

The basement rocks of the study area comprise Triassic-aged metamorphic rocks and Jurassic–Cretaceous ophiolite. These units are unconformably overlain by Miocene-aged sedimentary rocks named m1, m2, and m3 sequences. The m1 series comprises red, brown, gray, yellowish gray, and greenish gray siltstone, sandstone, and semi-rounded gravelstone. The m2 unit mainly includes green gravelstone, dark green, and gray sandstone, siltstone, marl, and coal-bearing claystone. Very thin limestone bands are locally observed in marl and claystone. The coal samples are deposited within this series. The m3 series, which is characterized by limestone and gravelstone, conformably overlies the m2 series and is unconformably covered by Quaternary alluvium (Figure 1).

The occurrences of Alpu coal in the Anatolids are found within the Miocene-aged m2 series. Two coal seam levels exist in each borehole, namely, the lower and the upper seam. The total thickness of the coal seams ranges from 1.2 m to 14.7 m for the lower seam, and 0.6 m to 15 m for the upper seam (Figure 2).

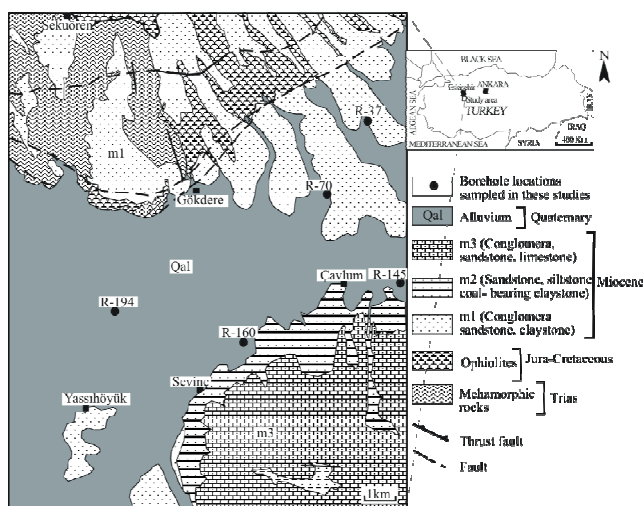


Figure 1. Geological map of the Alpu basin and locations of the boreholes sampled in this study.

3. Methods

Seventeen core samples (eight and nine coal samples

from the upper and the lower seam) from five boreholes (R-37, R-70, R-145, R-160, and R-194) were collected from the Alpu field (Fig. 2). The boreholes were drilled by the General Directorate of Mineral Research and Exploration (MTA). Proximate analysis was performed on the coal samples. Atomic absorption spectrometry (AAS) and X-ray fluorescence (XRF) were performed using standard techniques at the MTA Laboratory. For proximate analysis, the calorific values were obtained using Leco AC 500 device with the standard set to ASTM 5865. Total moisture, volatile matter, and ash contents were measured using Leco TGA-701 device. Total sulfur content was determined using Leco-SC144DR device. AAS was used to analyze the major elements. V, Cr, Ni, Cu, Zn, Rb, Sr, Ba, Pb, Zr, and As were analyzed by ARL 9800 series XRF.

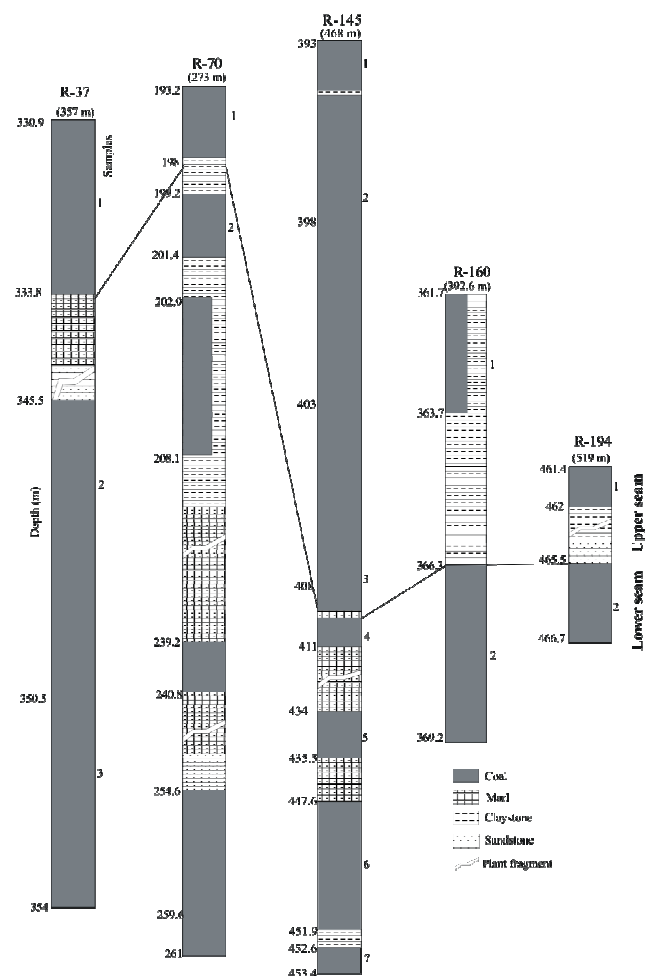


Figure 2. Sample locations studied in this study.

4. Results And Discussion

4.1. Proximate Analyses

Table 1 lists the results of proximate analyses of the Alpu coal samples in an air-dried basis. The total moisture contents of these coal samples range from 4.24% to 12.38% in the upper seam and 3.12% to 11% in the lower seam. The volatile matter values of the coal samples from the

upper and the lower seam range from 17.40% to 60.17%, and from 12.14% to 75.36%, respectively. The ash contents in the upper and the lower coal seam range from 22.04% to 37.14%, and from 19.85% and 43.12%, respectively. The fixed carbon contents range from 9.66% to 33.09% in the upper coal seam and 1.47% to 38.62% in the lower coal seam. The total sulfur contents in upper and lower coal seams range from 1.26% to 7.44% and from 0.42% to

3.05%, respectively. The total calorific values range from 1640 kcal/kg to 4617 kcal/kg in the upper coal seam, and from 983 kcal/kg to 5531 kcal/kg in the lower coal seam. Average total moisture, ash, fixed carbon contents, and total calorific values in the upper coal seam are higher than those in the lower seam. However, the average value of volatile matter content in the lower coal seam is higher than in the upper seam.

Table 1. Proximate analysis results of Alpu coals.

Upper Seam			Lower seam	
Proximate analyses (air-dried basis)				
Sample no	N=8	Average	N=9	Average
Total moisture (%)	4.24 - 12.38	8.13	3.12 - 11.00	6.14
Volatile matter (%)	17.40 - 60.17	40.2	12.14 - 75.36	46.27
Ash (%)	22.04 - 37.14	30.8	19.85 - 43.12	28.4
Fixed carbon (%)	9.66 - 33.09	20.9	1.47 - 38.62	19.2
Total sulphur (%)	1.26 - 7.44	2.6	0.42 - 3.05	1.9
Gross cal. Val. (kcal/kg)	1640 - 4617	3215	983 - 5531	2334

4.2. Geochemical Analyses

4.2.1. Major Elements

Table 2 lists the major elemental compositions of the Miocene-aged Alpu coal samples. Geochemical analyses indicate that the average values of the major elemental concentrations show similarity between the seams. The contents of Na, Mg, P, K, Ca, Ti, Mn, and S in the coal samples of the upper and the lower seam have nearly similar values. However, the average values of Al and Fe content are higher, and that of Si is lower in the upper seam

compared with that in the lower seam.

Table 2 also compares the major elements in the coal samples from the upper and the lower seam with the average world coal contents [19], in which differences in concentrations are observed. The general trends are similar, although a clear enrichment is shown in both seams with respect to the average world coal contents (Figures 3A and 3B). High major element concentrations in the Alpu coal samples show that these coals are probably low in rank.

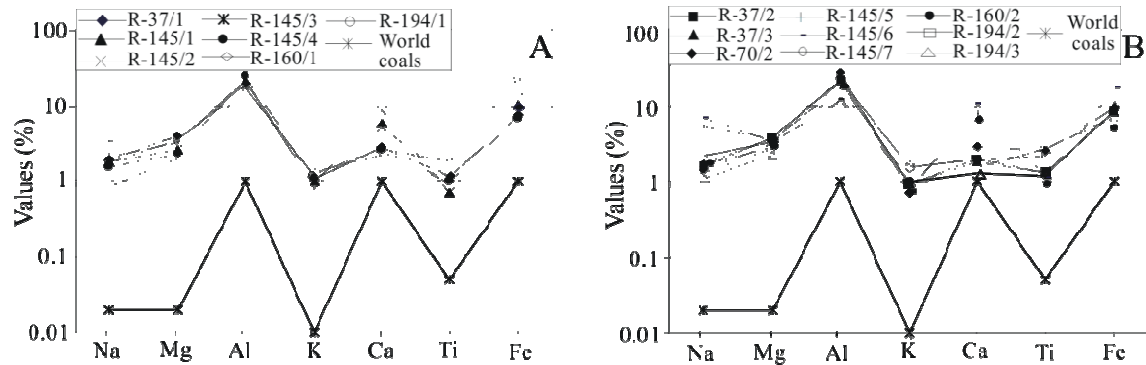


Figure 3. Diagrams of major element contents for Alpu coals and world average (A: Upper seam, B: Lower seam).

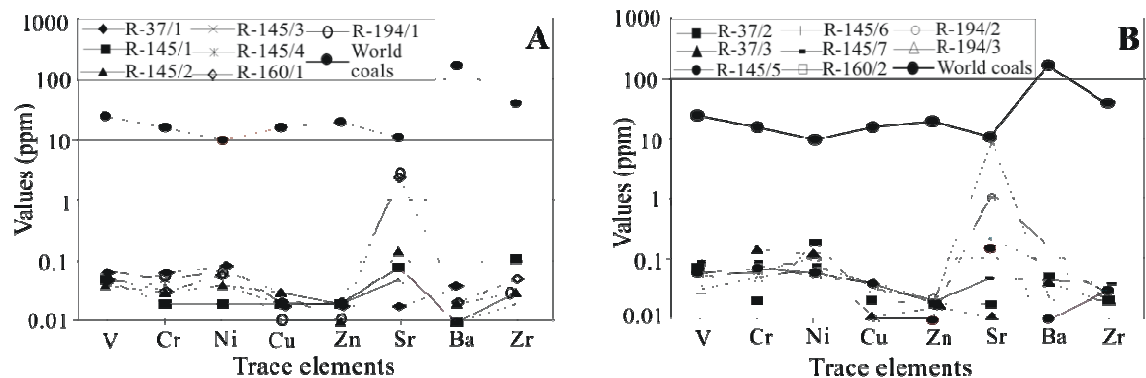


Figure 4. Diagrams of trace element contents for Alpu coals and world average (A: Upper seam, B: Lower seam).

Table 2. Major element contents of Alpu coals and world coals average.

	Sample	Major Elements (%)										
	No	Na	Mg	Al	Si	P	K	Ca	Ti	Mn	Fe	S
Upper Seam	R-37/1	2.04	4.08	20.70	57.60	0.10	1.12	2.96	1.22	0.10	7.86	2.23
	R-145/1	1.94	2.81	23.43	43.96	0.11	1.10	6.48	0.70	<0.1	10.90	8.51
	R-145/2	3.79	4.01	21.08	41.04	0.22	1.00	9.59	0.70	<0.1	8.92	9.56
	R-145/3	2.23	3.39	22.90	49.08	0.11	0.85	5.51	1.10	<0.1	8.70	6.08
	R-145/4	1.53	2.34	25.79	56.87	0.1	1.12	2.65	1.00	<0.1	6.93	1.64
	R-160/1	0.92	2.25	21.48	53.69	0.1	1.43	2.45	1.02	<0.1	14.32	2.33
	R-194/1	1.76	3.52	17.29	44.92	0.12	1.20	2.69	1.86	0.10	23.30	3.22
	Average	2.03	3.20	21.81	49.59	0.12	1.17	4.62	1.08	0.03	11.56	4.79
Lower Seam	R-37/2	1.63	3.78	22.14	58.20	0.10	0.92	2.04	1.33	<0.1	8.27	1.63
	R-37/3	1.72	3.75	21.99	58.99	0.10	0.91	1.82	1.32	<0.1	8.31	1.07
	R-70/2	1.43	3.17	27.43	49.65	0.10	0.72	2.97	2.50	<0.1	9.73	2.24
	R-145/5	5.56	3.93	18.43	42.31	0.11	0.87	8.40	1.20	<0.1	9.50	9.60
	R-145/6	7.35	3.62	13.73	26.44	0.11	0.77	11.10	1.10	<0.1	18.76	17.90
	R-145/7	2.14	3.37	21.14	56.47	0.10	1.53	1.94	2.10	<0.1	9.50	1.66
	R-160/2	1.97	3.39	22.28	45.76	0.11	0.98	8.74	0.98	<0.1	6.12	6.96
	R-194/2	1.32	2.85	22.57	59.16	0.15	1.50	1.52	2.74	<0.1	7.01	1.19
	R-194/3	1.13	2.26	12.22	69.23	0.20	1.00	1.34	1.23	0.20	9.76	1.40
	Average	2.69	3.34	20.21	51.80	0.12	1.02	4.43	1.61	0.02	9.66	4.85
World coals*		0.02	0.02	1.00	-	-	0.01	1.00	0.05	0.05	1.00	-

*From Valkovic (1983)

4.2.2. Trace Elements

Table 3 lists the trace element contents in the Alpu coal samples (V, Cr, Ni, Cu, Zn, Rb, Sr, Ba, Pb, Zr, and As). Concentrations of As (except R-37/1, R70-2), Rb, and Pb (except R-37/1, R-37/2, and R-37/3) in the upper and the lower seam are below the detection limit. The average values of V, Cr, Ni, Cu, Zn, Rb, Ba, Pb, and Zr in the coal samples indicate similarity between both two seams. However, the Sr content from the upper seam is lower than

that from the lower seam.

Table 3 compares the trace elements in the coal samples from the two seams with the average world coal contents [20]. The trace element contents from both seam coals are highly depleted with respect to world coal contents (Figures 4A and 4B). The coal samples have low trace element concentrations because of the absence of a volcanic source [21, 8].

Table 3. Trace element contents of Alpu coals and world coals average.

	Sample No	Trace Elements (ppm)										
		V	Cr	Ni	Cu	Zn	Rb	Sr	Ba	Pb	As	Zr
Upper Seam	R-37/1	0.07	0.07	0.09	0.02	0.02	0.01	0.02	0.05	0.01	0.01	<0.01
	R-145/1	0.05	0.02	0.02	0.02	0.02	-	0.08	0.01	-	-	0.11
	R-145/2	0.04	0.03	0.04	0.03	0.01	-	0.14	0.02	-	-	0.03
	R-145/3	0.07	0.05	0.07	0.03	0.02	-	0.08	0.01	-	-	0.02
	R-145/4	0.05	0.04	0.04	0.02	0.02	-	0.05	0.01	-	-	0.03
	R-160/1	0.05	0.04	0.06	0.02	0.02	-	2.28	0.02	-	-	0.05
	R-194/1	0.05	0.06	0.06	0.01	0.01	-	3.11	-	-	-	0.03
	Average	0.05	0.04	0.05	0.02	0.02	0.001	0.82	0.02	0.001	0.001	0.04
Lower Seam	R-37/2	0.07	0.02	0.18	0.02	0.02	0.01	0.02	0.05	0.01	-	0.02
	R-37/3	<0.01	0.14	0.12	0.01	0.01	0.01	0.01	0.04	0.01	-	0.02
	R-70/2	0.06	0.06	0.09	0.03	0.02	-	0.08	0.01	-	0.03	0.11
	R-145/5	0.06	0.07	0.06	0.04	0.01	-	0.15	0.01	-	-	0.03
	R-145/6	0.06	0.06	0.06	0.04	0.02	-	0.23	<0.01	-	-	0.03
	R-145/7	0.09	0.09	0.08	0.04	0.02	-	0.05	<0.01	-	-	0.04
	R-160/2	0.05	0.06	0.10	0.03	0.02	-	8.87	0.02	-	-	0.02
	R-194/2	0.08	0.08	0.05	0.04	0.02	-	1.17	-	-	-	0.04
	R-194/3	0.03	0.05	0.07	0.01	0.01	-	1.36	-	-	-	0.02
	Average	0.06	0.07	0.09	0.03	0.02	0.002	1.33	0.01	0.002	-	0.04
World Coals*		22	15	9	15	18	10	10	150	6.6	7.6	345

*From Ketris and Yudovich (2009)

4.3. Affinity of the Elements

Coal is a sedimentary rock and a complex heterogeneous mixture of organic matter and inorganic components. The correlation of the element contents with the ash yield of

coal may offer information for its organic or inorganic affinity [22, 23]. For coal samples with high ash contents, elements are mostly associated with inorganic components [24]. The amount of organically bound elements increases for coals with decreasing ash content, whereas the

abundance of inorganically bound elements decreases [25, 26]. Al and Si contents in both coal seams have positive correlations with ash yield ($r = 0.62$ for upper seam (Us), 0.69 for lower seam (Ls), and $r = 0.80$ for Us, 0.77 for Ls, respectively) at 95% confidence level (Figure 5). These elements are generally associated with inorganic components. Silisium has affinity with clastic minerals, and Al has aluminosilicate and feldspar affinities. Figure 5 also shows that the ash yields in the lower coal seams have positive correlations with K, P, Ti, and S ($r = 0.36, 0.35, 0.37$, and 0.63 ; at 95% confidence level). These elements are dominantly and inorganically associated. K has aluminosilicate and feldspar, S has sulfide, Ti has clay or FeTi-oxide minerals, and P has apatite affinity. However, these contents in the upper coal seam have negative correlations with the ash yield ($r = -0.32, -0.25, -0.41$, and -0.36 ; at 95% confidence level Figure 5). Yossifova et al. [5] indicated that organic matter increases the abundance of Ti and S into the bog. S and K are found in both organic

and inorganic matter. Ash yields of the coal samples in upper and lower seams have negative correlations with Mg, Na, Ca, and Fe ($r = -0.18$ for Us and -0.37 for Ls, $r = -0.26$ for Us and $r = -0.63$ for Ls, $r = -0.26$ for Us and $r = -0.72$ for Ls, and $r = -0.78$ for Us and $r = -0.36$ for Ls, respectively; at 95% confidence level, Figure 5). Na is mainly abundant in organic matter [5]. Fe is generally associated with inorganic components, but studies have indicated that this element has intermediate organic affinity in coal [27, 28, 29, 22, 23]. Mg is mainly related to clay and ferromagnesian minerals. Ca is characterized by carbonate affinity. However, studies have shown that Mg in some coal samples has organic affinity [27, 30, 31, 32, 33, 34, 35, 22]. Thus, Ca may also be found in organic matter [36]. Figure 6 shows that the concentrations of Ni and Ba in coal samples from both seams are positively correlated with the ash yield ($r = 0.32$ for Us and $r = 0.25$ for Ls, and $r = 0.52$ for Us and $r = 0.35$ for Ls, respectively; at 95% confidence level). These elements have inorganic affinity.

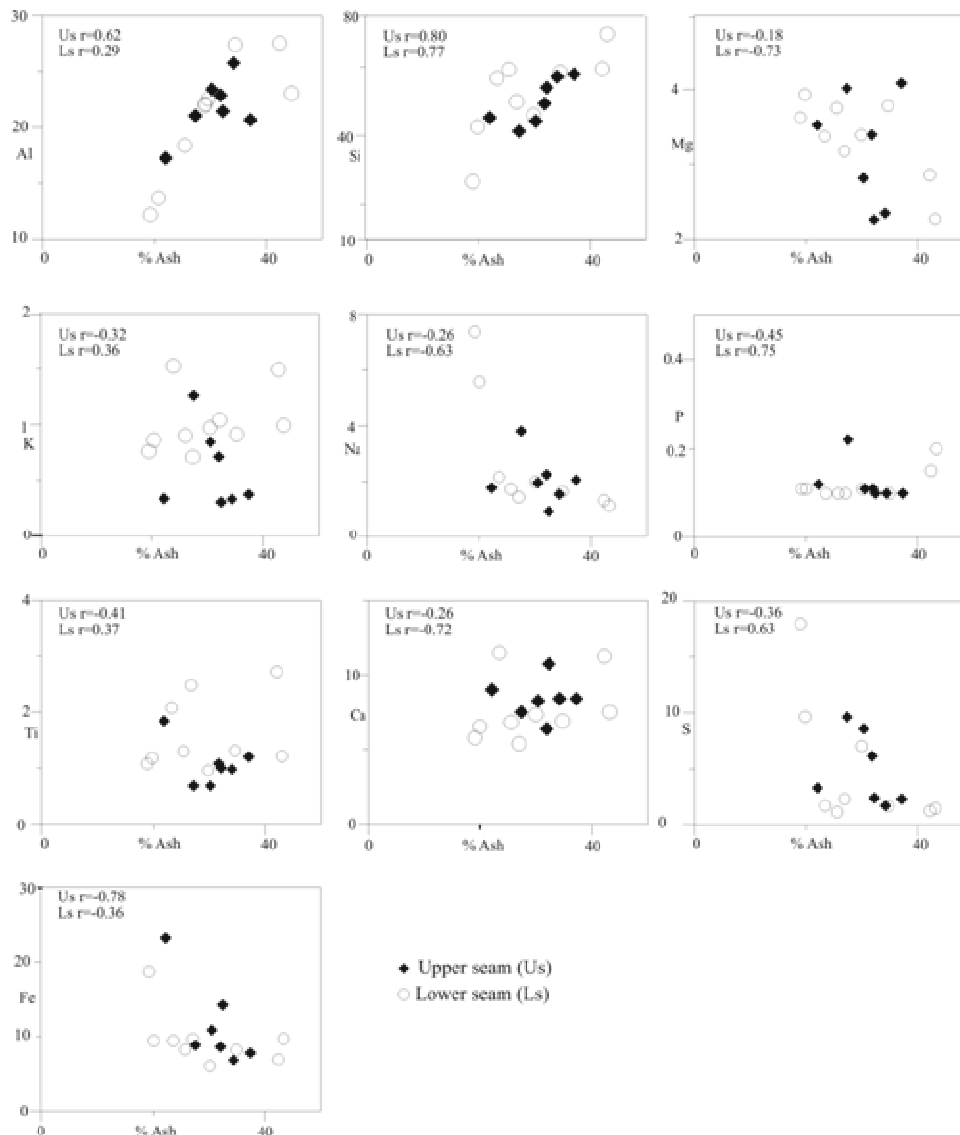


Figure 5. Relation between major element concentrations and ash yield.

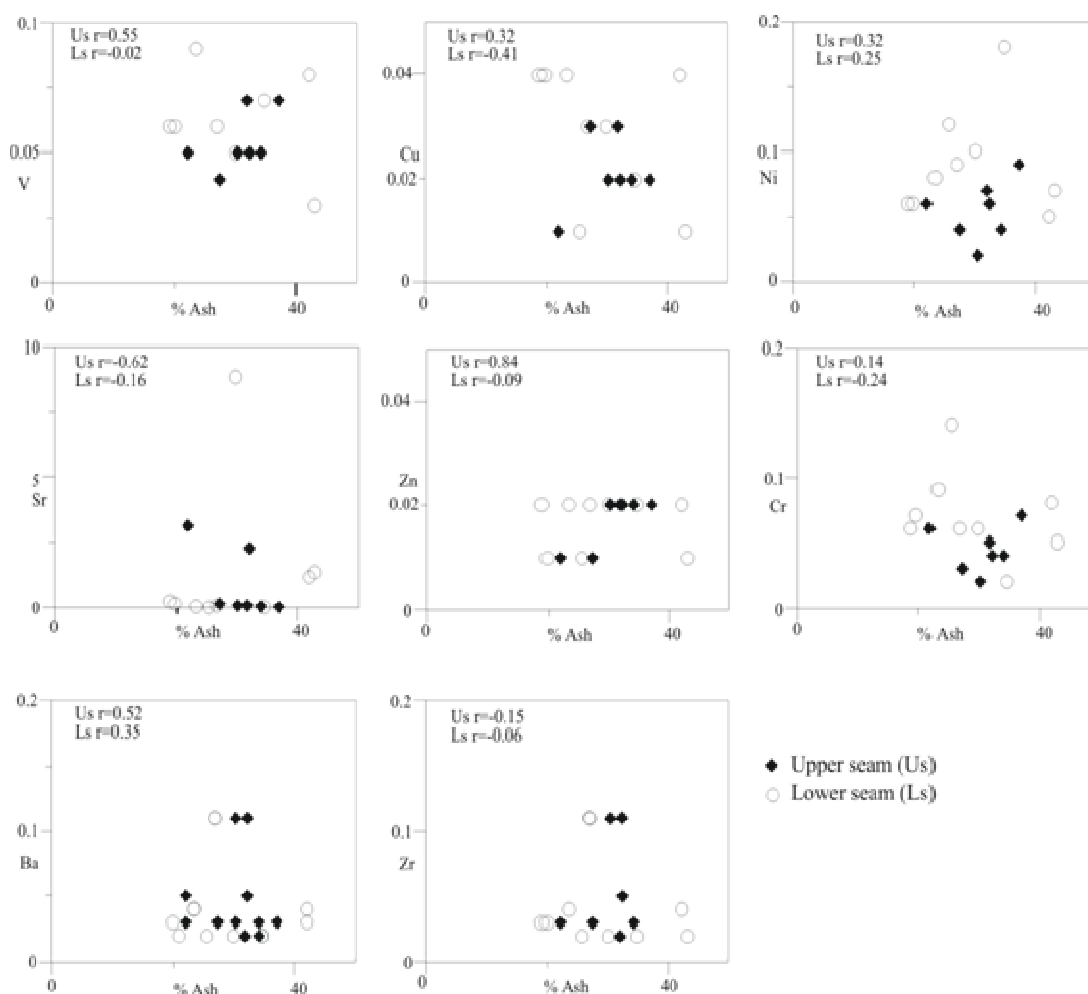


Figure 6. Relation between trace element concentrations and ash yield.

Figure 6 also shows that the ash yields in coal samples from the upper seam have positive correlation with Zn, Cu, Cr, and V ($r = 0.84, 0.32, 0.14$, and 0.55 , respectively; at 95% confidence level), suggesting a mineral matter affinity. Zn is mainly associated with the inorganic components in coal [12, 13, 14]. The organic associations of Zn have also been determined in low-ranked coals [15, 13, 16]. Cu and Cr in coal have organic affinity [17, 13, 8]. In addition, V in coal is associated with both inorganic and organic matter [17, 13]. Figure 6 further shows that the concentrations of Sr and Zr in both coal seams are negatively correlated with the ash yield ($r = -0.62$ for Us and -0.16 for Ls, and $r = -0.15$ for Us and -0.16 for Ls; at 95% confidence level), showing the association of these elements with organic matter. Zr is commonly associated with organic matter [17, 13]. The Sr is mainly found together with mineral matter (phosphate and carbonate minerals), and is associated with organic matter in low-ranked coals [17, 13, 18, 8].

5. Conclusions

Volatile matter, ash, fixed carbon, total sulfur contents, and total calorific values are higher in the upper seam than those in the lower seam on an air-dried basis. However, the

volatile matter contents are lower in the upper seam than in the lower seam. The average values of major element concentrations demonstrate similarity in both seams. However, the average Al and Fe concentrations are higher in the upper seam, and the average Si concentration is higher in the lower seam. The major elements in the coal samples from both seams differ from the world coal contents. Although a clear enrichment appears in both seams with respect to world coal contents, the general trends are similar. The Al and Si contents in both coal seams have inorganic affinity because of the positive correlations with the ash yield. The concentrations of Mg, Na, Ca, and Fe in both seams have negative correlations with the ash yield. Thus, these elements are associated with organic matter in coal. Ash yields from the lower coal seam samples have positive correlations with K, P, Ti, and S, indicating inorganic affinity. However, these elements in the upper coal seam have negative correlations with ash yield. Thus, K, P, Ti, and S contents in the upper coal seam are organically associated. The average values of V, Cr, Ni, Cu, Zn, Rb, Ba, Pb, and Zr in the coal samples show similarity for both seams. However, the Sr content in the lower coal seam is higher than that in the upper coal seam. The trace element contents of these seams are highly

depleted with respect to the world coal contents. Ni and Ba contents in both coal seams are positively correlated with ash yield, indicating that these elements have inorganic affinity. The concentrations of Sr and Zr in both coal seams are negatively correlated with the ash yield, which implies association with organic matter. The ash yield of the coal samples in the upper seam has positive correlations with Zn, Cu, Cr, and V, which indicate mineral matter affinities. Proximate and geochemical analyses imply that the Miocene-aged coal samples in Alpu are low in rank.

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