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THE DISTRIBUTION OF THE TRACE ELEMENT CONTENTS IN LIGNITE AND ASH FROM DRAMA LIGNITE DEPOSIT, USING MULTIVARIATE STATISTICAL ANALYSIS

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ABSTRACT

Multivariate statistical analysis was used on existing geochemical data of the Drama lignite deposit, eastern Macedonia, Greece. Factor analysis with varimax rotation technique was applied to study the distribution of major, trace and rare earth elements in the lignite and 850°C lignitic ash, to find a small set of factors that could explain most of the geochemical variability. The study showed that major elements Al, Na, K, contained in the lignite samples, presented high correlation with most of the trace and rare earth elements. In 850°C lignitic ashes major and trace elements present different redistribution. Only Al remained correlated with the trace elements Co, Cr, Rb, Ta, Th, Ti, Sc and rare earths related with inorganic matter in the lignite beds. Trace elements Fe, Mo, U, V, W, and Lu were associated with organic matter of lignite and had also been affected by the depositional environment.

ΣΥΝΟΨΗ

Η πολυμεταβλητή στατιστική ανάλυση χρησιμοποιήθηκε στην επεξεργασία και ερμηνεία βιβλιογραφικών γεωχημικών δεδομένων από το λιγνίτη του κοιτάσματος Δράμας. Η παραγοντική ανάλυση με την τεχνική της περιστροφής των αξόνων εφαρμόστηκε για τη μελέτη των κύριων στοιχείων, ιχνοστοιχείων και σπάνιων γαιών που περιέχονται τόσο στο λιγνίτη όσο και στις λιγνιτικές τέφρες, ώστε να ευρεθεί ένα μικρό σύνολο παραγόντων που μπορεί να εξηγήσει το μεγαλύτερο μέρος της γεωχημικής ποικιλότητας. Η μελέτη δείχνει ότι τα κύρια στοιχεία Al, Na, K που ανιχνεύτηκαν στα δείγματα λιγνίτη παρουσιάζουν υψηλή συσχέτιση με τα περισσότερα ιχνοστοιχεία και τις σπάνιες γαίες. Στις λιγνιτικές τέφρες των 850 oC τα στοιχεία (κύρια και ιχνοστοιχεία) παρουσιάζουν διαφορετική κατανομή. Μόνο το Al παραμένει πολύ συσχετισμένο με τα ιχνοστοιχεία Co, Cr, Rb, Ta, Th, Ti, Sc και σπάνιες γαίες που συνδέονται με την ανόργανη ύλη των λιγνιτικών στρωμάτων. Τα ιχνοστοιχεία Fe, Mo, U, V, W, Lu συνδέονται με την οργανική ύλη του λιγνίτη και επηρεάζονται επίσης από το περιβάλλον απόθεσής του.

KEY WORDS: lignite, trace elements, lignite ash, Drama, multivariate analysis, factor analysis

1. INTRODUCTION

Factor analysis, a multivariate statistical technique, is useful for evaluating the correlation among several variables and consequently is useful for interpreting geochemical data in the lignite samples relating them to specific processes. In the present study, factor analysis was applied to geochemistry data of major, trace and rare earth elements of the Drama lignite deposit to evaluate the dominant process controlling the distribution of elements in the lignite samples and 850 °C lignitic ashes. The chemistry and mineralogy of lignite in Drama deposit have been investigated by Filippidis et al., (1996), Foscolos et al., (1998) and Georgakopoulos (2000). The aim of this study was to point out the correlation between the 34 elements contents by the use of multivariate systems applying the method of factor analysis.

2. STUDY AREA

In the central plain area of the Drama basin, western of Philippi peat-lignite deposit (Melidonis 1969 ; Christanis 1983), drilling investigation executed by IGME discovered a new lignite deposit which covers an area over 90 Km2 with 1.4 *109 tons of geological lignite reserves (Figure 1). The lignite deposit consisted mainly by

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three lignite seams, which are separated by medium to fine grained clastic sediments as silts and sandy silts (Goodrarzi et al., 1990). The maximum thickness of the three lignite seams occurs in the center of the lignite deposit and exceeds 160 meters. The average moisture content is 59.4 %, the average ash content is 16% and the gross calorific value is 1.015 Kcal/Kg (as received basis). The mineralogy of low temperature ash consists of quartz and gypsum in all samples and feldspars, clay minerals and pyrite (Foscolos et.al. 1998). Most of the elements such as As, Cl, Co, Cr, Cu, Mn, Mo and Sb have relatively high concentrations in these lignites compared to trace elements content of international lignites (Foscolos et al., 1989).

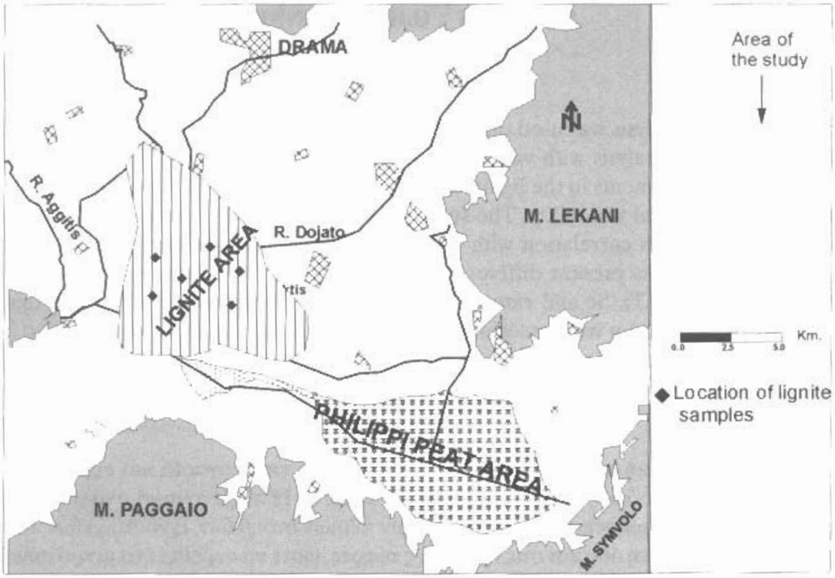

Fig. 1. Location of the study iv Drama area, Eastern Greece.

In a previous study Foscolos et al. (1998) compared the concentration of 39 elements (major, trace and rare earths) analysed on 34 samples, with the element crustal abundances (Table 1). The mean concentration of the trace elements, Sb, As, Br, Cs, I, Mo, Se, W and U were found enriched, having an enrichment factor 2.7 to 82.7. The major elements Al, Br, Co, Cu, Na showed concentration values, which were less than the Earth's crust values. The enrichment of elements in the 850oC lignite ashes were also high as it showed an enrichment factor from 40.86 to 92.5.

3.PROCEDURE USED

Statistical method of multivariate factor analysis is used in data reduction, by identifying a small number of factors which explain most of the variance observed in a much larger number of manifest variables without losing much information (Davis, 1986). This is accomplished by diagonalization of the correlation matrix of the data, which transform the original variables into uncorrelated (orthogonal) ones (weighed linear combinations of the original variables) called Principal Component (David et al., 1997). The data were standardized and arranged in correlation coefficient matrix with normal distribution in all variables. The calculated eigenvalues quantified the contribution of a factor to the total variance. Their percentages were computed. The contribution of a factor is significant when the eigenvalue is greater than one. The factor loadings were calculated by the Varimax rotation technique (Kaiser 1958) in such way that they were enclosed to +1, 0, -1, representing positive contribution, zero and negative contribution. This technique has the effect of producing a set of uncorrelated factors in which each variable has high loadings on some factors and near-zero loadings on the others.

In the present work 34 lignite samples and 850oC lignitic ashes from the data of Foscolos et al., (1998) were used separately. For the calculations 34 variables were used for each sample. Statistical package SPSS 7.5 for Windows (1992) was used to carry out the calculations.

Table 1: The enrichment ratio of elements concentration in the lignite and 850°C lignitic ashes. (Data from Foscolos et al. 1998)

Element	Crustal average ppm	Factors ratio in the lignite samples (ppm)			Factors ratio in 850C lignite ash (ppm)		
		Min	Max	Mean	Min	Max	Mean
Al	81300.0	0.10	0.8	0.4	0.40	1.7	1.17
Sb	0.2	8.50	47.9	21.7	17.00	136.5	69.89
As	1.8	12.22	76.7	39.8			
Ba	425.0	0.04	1.2	0.7	0.93	4.5	2.32
Br	2.5	6.28	18.9	11.2			
Ca	36300	0.63	5.6	1.2	1.43	23.0	4.65
Ce	60.0	0.15	0.9	0.4	0.54	2.2	1.32
Cl	130	0.02	2.9	1.1			
Co	25	0.10	0.5	0.2	0.04	1.0	0.69
Cr	100	0.20	1.0	0.5	0.50	2.4	1.59
Cs	3	0.97	4.5	2.7	2.07	17.0	8.77
Cu	55	0.12	0.8	0.3	0.37	2.0	1.08
Dy	3.0	0.30	1.2	0.7	0.07	3.2	2.15
Eu	1.2	0.08	0.8	0.3	0.42	1.6	1.02
Fe	50000.0	0.00	0.7	0.4	0.39	1.8	1.16
Ga	15.0	0.22	1.0	0.6			
Hf	3.0	0.10	0.8	0.4	0.57	9.2	1.51
Ho	1.2	0.33	1.9	0.8	1.00	5.9	2.74
I	0.5	5.60	13.0	9.1			
K	25900.0	0.00	0.4	0.2	0.24	6.1	0.73
La	30.0	0.02	0.9	0.3	0.08	2.0	1.00
Lu	0.5	0.20	1.0	0.6	0.20	3.0	1.90
Mg	20900.0	0.00	0.4	0.2	0.44	1.4	0.91
Mn	950.0	0.03	0.7	0.1	0.18	1.3	0.40
Mo	1.5	5.67	50.3	25.8	11.67	214.9	92.51
Na	28300.0	0.00	0.1	0.1	0.06	0.3	0.21
Nd	28.0	0.03	1.0	0.4	0.25	2.0	1.31
Rb	90.0	0.11	1.1	0.5	0.58	2.2	1.42
Sm	6.0	0.13	0.7	0.4	0.45	1.9	1.18
Sc	22.0	0.10	3.3	0.3	0.30	1.1	0.78
Se	0.1	36.00	188.6	82.1			
Ta	2.0	0.10	0.5	0.2	0.30	1.0	0.72
Th	7.2	0.19	1.9	0.8	0.92	4.1	2.56
Ti	4400.0	0.09	0.6	0.3	0.08	1.4	1.00
U	1.8	7.44	43.3	24.5	15.17	173.9	88.57
V	135.0	0.37	1.9	0.8	0.86	5.5	2.82
W	1.5	2.87	19.3	11.5	5.87	103.2	40.86
Yb	3.4	0.15	0.6	0.3	0.41	1.8	1.04
Zn	70.0	0.08	1.7	0.6	0.59	4.7	1.87

4.RESULTS AND DISCUSSION

Table 2 shows the mean, standard deviation, minimum and maximum values obtained from the descriptive analysis applied to determined variables. After the descriptive analysis of the variables, the construction of the correlation matrix proceeded between chemical parameters of the lignite and 850°C lignitic ashes.

Table 2: Statistical data of trace elements in air-dried lignite samples and 850°C lignite ash (Geochemical data obtained from Foscolos et al. 1998)

element	air-dried lignite				850° C lignite ash			
	Mean	Minimum	Maximum	Std. Deviation	Mean	Minimum	Maximum	Std. Deviation
Al	30583.0	8450.0	63700.0	14012.6	95513.0	32800.0	137000.0	29243.07
Sb	4.0	1.7	9.6	1.7	14.0	3.4	27.0	6.6
As	72.0	22.0	138.0	27.0				
Ba	291.0	15.1	493.8	106.6	985.0	396.7	1908.0	358.7
Br	28.0	15.7	47.2	6.1				
Ca	43562.0	22700.0	203000.0	36828.1	168928.	51910.0	836000.0	153419.0
Ce	25.0	8.8	55.7	12.1	79 0	32.2	129.0	22.9
Cl	137.0	2.3	371.0	82.5				
Co	6.0	2.5	12.4	2.3	17.0	0.9	25.0	4.9
Cr	50.0	19.6	95.4	19.2	159.0	50.2	240.0	38.3
Cs	8.0	2.9	13.6	2.9	26.0	6.2	51.0	8.6
Cu	18.0	6.8	44.6	9.2	60.0	20.3	110.0	24.1
Dy	2.0	0.9	3.62	0.7	6.0	0.2	10.0	1.8
Eu	0.0	0.1	0.9	0.2	1.0	0.5	2.0	0.4
Fe	18158.0	17.9	32600.0	6333.0	57955.0	19620.0	92200.0	16542.5
Ga	9.0	3.3	15.1	2.9				
Hf	1.0	0.3	2.4	0.6	5.0	1.7	28.0	4.3
Ho	1.0	0.4	2.3	0.4	3.0	1.2	7.0	1.3
I	5.0	2.8	6.5	0.9				
K	4480.0	5.4	10800.0	2361.3	19007.0	6170.0	158000	26117.4
La	10.0	0.5	26.1	6.7	30.0	2.5	59	15.2
Lu	0.0	0.1	0.5	0.1	1.0	0.1	1.5	0.4
Mg	5205.0	5.5	7520.0	978.0	18958.0	9160.0	29910	4555.0
Mn	119.0	30.3	621.0	97.7	382.0	168.2	1273	229.8
Mo	39.0	8.5	75.4	15.1	139.0	17.5	322	69.6
Na	1830.0	1.3	3150.0	751.0	6067.0	1640.0	9260	1733.3
Nd	12.0	0.8	27.0	5.5	37.0	7.0	57	10.7
Rb	41.0	10.2	100.0	20.1	128.0	51.9	202	36.9
Sc	7.0	2.1	72.3	11.7	17.0	6.5	24	4.7
Se	4.0	1.8	9.4	1.8				
Sm	2.0	0.8	4.4	1.0	7.0	2.7	11.6	2.0
Ta	1.0	0.2	0.9	0.2	1.0	0.6	2	0.4
Th	6.0	1.4	13.9	3.0	19.0	6.6	29.5	6.1
Ti	1428.0	390.0	2760.0	641.7	4382.0	346.0	6280	1457.7
U	44.0	13.5	78.0	17.0	159.0	27.3	313	79.7
V	112.0	49.9	252.0	42.7	381.0	116.2	748	152.3
W	17.0	4.3	29.0	5.5	61.0	8.8	155	28.9
Yb	1.0	0.5	2.1	0.4	4.0	1.4	6.1	1.0
Zn	40.0	5.4	120.0	24.2	131.0	41.6	331	59.9

In the lignite samples, significant correlation between the major elements (Al-K: 0.93), (Al-Na: 0.76), (Na-K: 0.85) can be recognized. These elements were correlated with most of the trace elements and rare earth's. It is noted that Mg and Ca presented low correlation's with all the other elements.

Correlation coefficient matrix at 850 °C lignite ash showed that the ash content presented negative correlation coefficient with Mg (-0.72), Mo (-0.82), Sb (-0.68), U (-0.83), V (-0.54), and W (-0.78). These elements showed that their concentration increased with decreasing ash content and can therefore be evaluated as associated with the organic matter of the lignite. The elements Th (0.53), Ce (0.52), La (0.70), Ta (0.54), Ti (0.52)showed low to medium positive correlation with ash content and can be related with the inorganic mineral in the lignite. The major element Al presented positive correlation with the ash content (0.42) and high positive correlation with the trace elements. Major elements as Na, Ca, K, Cu, Mn are poorly correlated with trace and rare earth elements and didn't show any preference towards minerals or organic matter.

Factor matrix obtained by Principal Component Analysis was subjected to a Varimax rotation. The extracted factors with eigenvalues greater than one were considered in the interpretation following varimax rotation.

Table 3 shows eigenvalues and variance explained (%) from the first five extracted axes of elements in the lignite samples accounted for about 87% of total variance (eigenvalues greater than one) and Table 4 shows seven eigenvalues explained 87.7% of the total variance from the 850oC lignitic ashes.

Table 3. Results of the Varimax rotated analysis in lignite samples

Factor	Eigenvalues	% of	Cumulative
1	17.37	51.08	51.08
2	5.09	14.97	66.04
3	4.08	11.99	78.03
4	1.66	4.87	82.91
5	1.40	4.11	87.02

Table 4. Results of Varimax rotated analysis in 850 oC lignitic ashes

Factor	Eigenvalues	% of	Cumulative
1	12.3	36.2	36.2
2	5.4	16.0	52.2
3	3.9	11.4	63.6
4	2.5	7.4	71.0
5	2.1	6.2	77.2
6	2.0	5.8	83.0
7	1.6	4.6	87.7

Major and trace elements in lignite samples

Table 5 summarizes the results obtained from rotated factor analysis loadings of variables in lignite samples. It shows that 78% of the total variance is attributable to the first three factors and only 9% of the total variance in the two minor factors.

Table 5: Varimax rotated factor matrix of elements in the lignite samples

Variable	VF1	VF2	VF3	VF4	VF5
Al	**0.94**	0.04	0.30	0.05	0.00
Sb	0.10	**0.92**	0.06	-	0.08
Ba	0.43	0.66	0.29	0.10	-0.16
Ca	-0.22	-0.25	-0.35	0.04	0.06
Ce	**0.94**	0.01	0.25	**0.60**	0.03
Co	**0.91**	0.17	0.20	0.08	0.07
Cr	**0.87**	0.36	0.22	0.14	0.15
Cs	**0.75**	0.35	0.05	-	0.07
Cu	0.43	0.27	**0.79**	0.10	-0.01
Dy	**0.91**	0.25	0.28	0.00	-0.01
Eu	**0.94**	0.05	0.19	0.02	0.00
Fe	0.56	0.38	0.29	0.01	0.11
Ga	**0.78**	0.12	0.47	-	-0.04
Hf	**0.85**	-0.08	-0.13	0.33	-0.19
Ho	0.54	0.03	-0.22	-	-0.44
K	**0.85**	-0.10	0.06	0.17	0.32
La	**0.95**	-0.20	0.19	-	0.03
Lu	0.37	0.65	0.42	0.22	0.15
Mg	0.23	-0.11	-0.12	-	**0.90**
Mn	0.30	0.24	**0.85**	-	0.04
Mo	-0.12	**0.81**	-0.36	0.07	-0.04
Na	**0.80**	0.19	-0.28	0.18	0.08
Nd	**0.88**	0.14	0.30	0.09	0.05
Rb	**0.96**	-0.03	0.03	0.08	0.03
Sc	0.39	-0.20	-0.11	0.75	0.00
Sm	**0.93**	0.19	0.26	0.06	0.06
Ta	**0.94**	0.02	0.17	0.25	-0.04
Th	**0.89**	0.09	0.43	0.02	0.01
Ti	**0.94**	0.02	0.16	0.18	0.06
U	-0.17	**0.93**	0.01	-	-0.23
V	0.32	**0.84**	0.34	-	0.01
W	-0.13	0.37	**-0.70**	-	0.24
Yb	**0.88**	0.25	0.29	0.04	0.01
Zn	**0.77**	0.22	0.50	0.06	0.13

The first rotated factor VF1, which explained 51% of the total variance, presented high positive loadings for major elements Al, Na, K, trace elements Zn, Co, Cr, Ti, Cs, Rb, Ta, Th and rare earths (*Ce, *Dy, *Eu, *La, *Nd, *Sm and *Yb). This factor can be associated with detrital constituents in lignite resulting from the surrounding rocks of Drama basin.

The second factor VF2, which explained 15 % of the total variance, presented high loading in the trace elements Sb, Mo, V and U and probably could be associated with inorganic constituents deposited syngenetically with the organic matter of lignite.

The third factor VF3 that explained 12 % of the total variance showed that W played an antagonistic role to the Cu associated to Mn.

From the graphical representation of the first two rotated factors VF1-VF2 (Figure 2), which explain 66% of total variance, it is observed that factor VF2 is characterized by high loadings in the elements Mo, U, V, and Sb. These variables are positively located closely in VF2 coordinate system. Intermediate affinity between the factors VF1 (characterized by detrital elements) and factor VF2 (characterized by organic constituents) present the elements Fe, Cu and Mn.

Plot of rotated factor VF1-VF3 (Figure 3) shows that Mn and Cu presented positive high loadings in the factor VF2 with W negative correlation in the same factor.

The other two minor factors VF4 and VF5 explained only 4.9% and 4.1% of the total variance and were characterized by high loadings of Ca and Mg respectively.

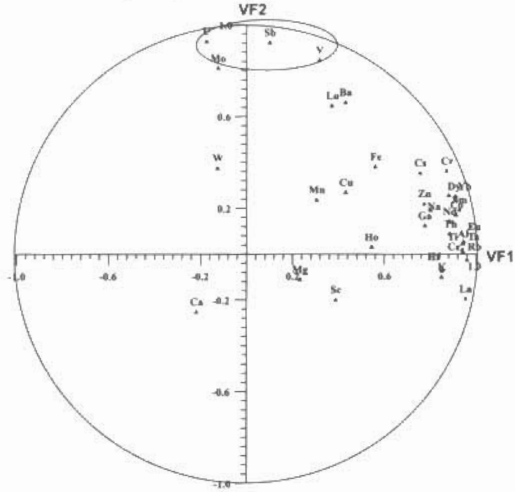

Fig. 2 Plot of the first two rotated factor weights 1-2 in lignite elements

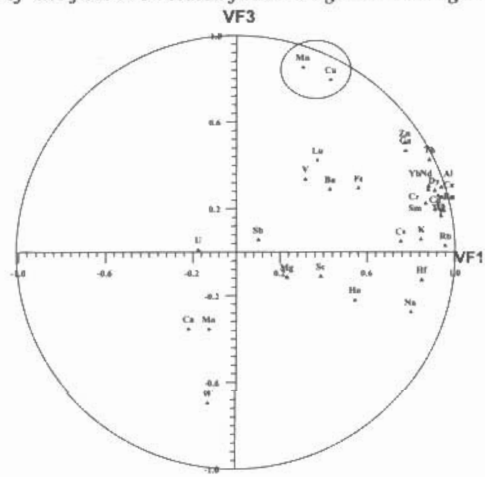

Fig. 3 Plot of the first two rotated factor weights 1-3 in lignite elements

Major and trace elements in 850 °C lignite ash

Table 6 shows that 63% of the total variance is attributable to the first three factors and the other four minor factors explained 24.4% of the total variance.

Table 6: Varimax rotated factor matrix in 850 oC ash elements

Variable	VF1	VF2	VF3	VF4	VF5	VF6	VF7
Al	**0.87**	-0.24	0.23	0.20	0.00	0.12	-0.07
Ba	0.23	0.30	**0.72**	0.20	0.10	0.10	0.33
Ce	**0.90**	-0.04	-0.08	0.20	0.00	-0.30	-0.03
Co	**0.76**	0.27	-0.10	-0.10	-0.20	0.20	0.08
Cr	**0.88**	0.13	0.16	0.00	-0.10	0.10	-0.20
Cs	0.31	0.14	**0.83**	0.10	-0.10	-0.11	0.08
Cu	0.38	0.22	-0.09	**0.67**	-0,.90	0.21	-0.28
Dy	**0.77**	0.44	0.23	0.15	0.10	0.00	0.21
Eu	**0.81**	-0.08	-0.05	0.28	0.05	-0.10	0.29
Fe	0.24	**0.77**	0.08	0.15	-0.04	0.29	0.17
Hf	-0.07	0.27	0.04	0.15	-0.05	**0.88**	0.05
Ho	0.01	0.06	0.27	-0.22	-0.10	0.07	**0.85**
K	0.18	-0.07	-0.08	-0.05	**0.96**	0.03	-0.10
La	**0.80**	-0.29	-0.27	0.18	0.10	-0.27	0.17
Lu	0.22	**0.87**	0.13	0.21	0.04	0.09	-0.18
Mg	-0.14	0.46	**0.66**	-0.20	-0.23	-0.09	-0.06
Mn	0.13	0.21	0.43	**0.68**	0.11	0.32	0.11
Mo	-0.36	**0.76**	0.27	-0.36	-0.10	0.00	-0.01
Na	0.32	0.11	0.55	-0.07	0.10	0.49	-0.04
Nd	**0.86**	0.00	0.02	0.24	-0.13	0.01	0.05
Rb	**0.78**	-0.21	-0.11	-0.35	-0.10	0.16	-0.17
Sb	-0.12	0.53	**0.64**	0.04	-0.03	0.39	0.18
Sc	**0.92**	0.05	0.21	0.17	0.01	0.11	0.04
Ca	-0.31	-0.02	-0.06	0.00	**0,90**	-0.10	0.02
Sm	**0.93**	0.14	0.11	0.18	0.03	-0.07	0.11
Ta	**0.84**	-0.25	0.03	-0.02	-0.08	0.15	-0.23
Th	**0.90**	-0.15	0.10	0.30	0.10	0.02	-0.08
Ti	**0.84**	-0.32	0.25	-0.50	0.04	0.10	-0.21
U	-0,20	**0.74**	0.51	-0,05	-0.13	0.24	0.22
V	0.22	**0.66**	0.48	0.19	-0,01	0,40	0,20
W	-0.51	**0.62**	0.21	-0.35	-0.14	-0.12	-0.07
Yb	**0.85**	0,39	0.02	0.13	-0.04	-0.03	0.15
Zn	0.38	-0,29	-0.10	**0.72**	-0.03	-0.14	-0.27

For VF1 the elements Al, Co, Cr, Rb, Ta, Th, Ti, Sc and rare earths (*Yb, *Ce, *Dy, *Eu, *La, *Nd, and *Sm) had high loadings (>0,8). Factor VF1 accounted for 36 % of total variance in 850 oC ash contents, whereas factor VF1 in the lignite elements accounted for 51% of the total variance. The difference may reflect the redistribution of major elements Na and K in lignitic ashes. Na presented low loadings in all factors, while K presented high ones (0.96) at factor VF5 correlated with Ca.

The second factor (VF2) explained 16% of the total variance and showed high positive loadings in trace elements Fe, Mo, U, V, W, Lu. This factor is characterized as having an organophilic affinity and it is associated with lignite macerals related also to environmental effect.

The common plotting factors are projected on the plane VF1 and VF2 (Figure 4) and the contributions of the elements as well as the sign of their loadings on these factors are listed on Table 6.

The third factor (VF3) explains 11% of the total variance and showed high positive loading in the element Mg and trace elements Ba, Sb and Cs and may be analogous to factor (VF4) associated with major elements Cu, Mn and Zn (Figure 5 & 6). Factor VF5 showed moderate loadings at Ca (0.90) and K (0.96). The other two factors explained less than 10% of total variance.

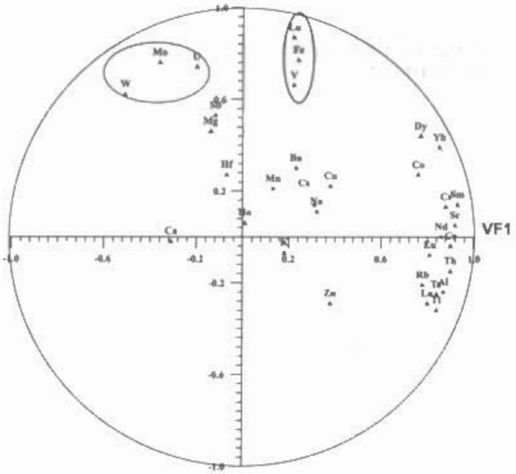

Fig. 4 Plot of the first two rotated factor weights 1-2 in 850 °C lignite ashes

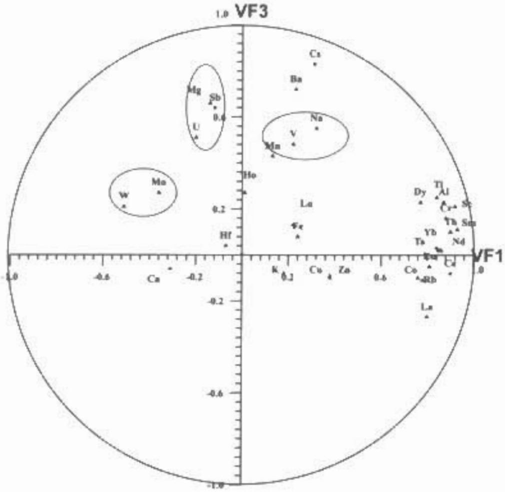

Fig. 5 Plot of the first two rotated factor weights 1-3 in 850 °C lignite ashes

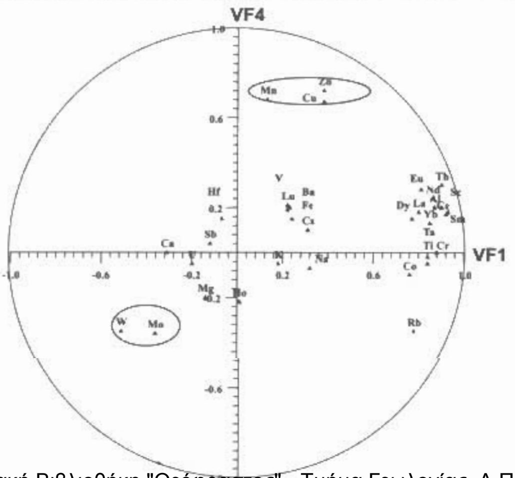

Fig. 6 Plot of first two rotated factor weights 1- 4 in 850 °C lignite ashes

5.CONCLUSIONS

Factor analysis with varimax rotation technique reduced the 34 variables in the lignite samples to five significant factors that explain 87.02% of the total variance. The first factor namely VF1 (51.O2% of the total variance), can be associated with detrital constituents in lignite resulting from the surrounding rocks of Drama basin.

From varimax rotation in the 850 °C lignitic ashes seven significant factors were extracted that explain 87.7% of total variance. Factor VF1 accounted for 36% of total variance in 850oC ashes contents, whereas factor VF1 in the lignite elements accounted for 51% of the total variance. This difference could reflect the redistribution of major elements Na and K in lignitic ashes. Na presented low loadings in all factors while K presented high loadings (0.96) at factor VF5 correlated with Ca.

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