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ENVIRONMENTAL ASPECTS IN RELATION TO THE LIGNITE COMBUSTION AT THE POWER STATION OF AMYNTAION, MACEDONIA, GREECE

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ABSTRACT

Daily lignite samples together with the corresponding flyash and bottom ash were collected from power plants of Amyntaion and analysed for their trace elements concentrations. Some possible impacts on the environment related to the lignite combustion are discussed.

Leaching experiment proved in some extent that trace elements are more readily leached from flyash than from lignite or shale and thus pointed out the need for further investigation of the suitability of the waste disposal site.

ΠΕΡΙΛΗΨΗ

Ο κύριος σκοπός της παρούσης εργασίας είναι να μελετήσει την ανακατανομή των ιχνοστοιχείων κατά την καύση του λιγνίτη της Πτολεμαίδας. Ημερήσια δείγματα λιγνίτη με την αντίστοιχη ιπτάμενη τέφρα και τέφρα δαπέδου συλλέγησαν από την γραμμή παραγωγής του ΑΗΣ Αμυνταίου. Η ανάλυση τους με μεθόδους XRF κατέδειξε το περιεχόμενο τους σε ιχνοστοιχεία και μια πρώτη περιβαλλοντική εκτίμηση μπόρεσε να καταγραφεί. Πειράματα απόπλυσης επιβεβαίωσαν ότι τα ιχνοστοιχεία πιο εύκολα μεταπίπτουν σε υδατοδιαλυτές μορφές από τη στερεά κατάσταση όταν βρίσκονται στην ιπτάμενη τέφρα παρά στο λιγνίτη, θεμελιώνοντας έτσι την ανάγκη για περεταίρω έρευνα της καταλληλότητας του χώρου απόθεσης της ιπτάμενης τέφρας στην περιοχή μπροστά στον κίνδυνο μόλυνσης του υδροφόρου ορίζοντα. Τέλος το περιεχόμενο σε ιχνοστοιχεία και κύρια της ιπτάμενης τεφρας και της τέφρας δαπέδου και του λιγνίτη μπόρεσε να καταγραφεί σε ημερήσια βάση.

INTRODUCTION

Ptolemais basin constitutes the principal coal fields of Greece. More than 50% of the total power production of Greece is generated by more than 10 power stations operating in the area.

During lignite combustion the mineral matter of lignite undergoes a series of physical and chemical changes (e.g., MITCHELL & GLUSKOTER 1976, HARRIS et al.1979, FILIPPIDIS et al. 1991,1992,1994, KOSTAKIS et al. 1991, UNSWORTH et al. 1991, FILIPPIDIS & GEORGAKOPOULOS 1992, KASSOLI-FOURNARAKI et al. 1992).

Combustion of lignite for power generation causes redistribution of trace elements from the lignite in to bottom ash, flyash and stack gases (e.g. DAVISON et al. 1974, NATUSCH et al. 1974, COLES et al. 1979,

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FOSCOLOS et al. 1989, GEORGAKOPOULOS et al. 1994, THOMAS 1992).

Unfortunately, this result in a variety of troublesome environmental consequences and very often undesirable health problems that are associated with the operation of coal-fired power plants occur (e.g. PAPASTEFANOU & CHARALAMBOUS 1979, THORNTON 1983, GAY & DAVIS 1987, GREY et al. 1988, MANOLOPOULOU 1990, GEORGAKOPOULOS et al. 1992a, GUTHRIE 1992, KASSOLI-FOURNARAKI et al. 1993).

Extended lignite deposits are found in Ptolemais basin in northern Greece, and more than ten pulverised-lignite-fired power plants operate in the greater area. Samples of lignite from a power station were collected together with the corresponding flyash and bottom ash and trace and major elements concentrations were determined. Leaching experiments were also performed for three trace elements (Cu, Ni, Cr)

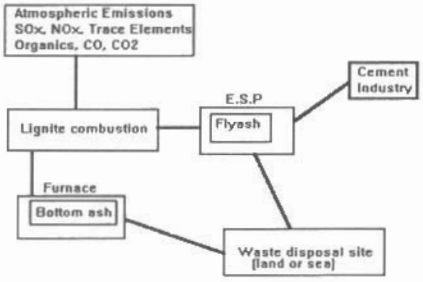

Fig. 1: Schematic diagram of pathways of trace contaminants during lignite combustion.

ENVIRONMENTAL ASPECTS

Coal has special problems associated with mining, cleaning, transportation, utilisation and waste disposal cycle. All the distinct steps of coal utilisation mentioned above could generate mobilisation of trace elements. As a result, primary exposures might contribute to possible health effects.

The potentially hazardous elements in coal resource development are classified in five categories of: i. greatest concern (As, B, Cd, Pb, Hg, Mo and Se), ii. moderate concern, iii. minor concern, iv. radioactive elements, v. concern but with negligible concentrations.

Some of the health implications concerning the combustion and disposal of coal are: respiratory irritation, chronic disease, cancer, decreased resistance to infection, silicosis, tuberculosis, cardiovascular disease, bronchitis, nephropathies and subclinical CNS disorders (e.g. KAGEY & WIXSON 1983, GAY & DAVIS 1987, FEDER et al. 1991, GUTHRIE & MOSSMAN 1993).

The amounts of pollutants entering the environment as the result of coal combustion increase with the increasing coal utilisation. Electric power generation requires large tonnage, therefore trace elements present in coal in very low levels are of environmental importance. Distribution of trace elements during the combustion of coal is given in figure 1. Among the others, radioactivity, air pollution and coal by-products (flyash and bottom ash) must be considered as a first priority when environmental evaluation is attempted.

It is well known in Greece that the city of Ptolemais faces huge pollution problems at present. The degradation of the area is due mainly to open-cast mines and power stations operating around the city of Ptolemais. One of the major problems that concerns the local authorities and can easily be seen, is the respirable dust spreaded by winds from stock piles. This of course, affects mainly the mine workers but dust transported in urban areas (due to transportation with vehicles or conveyors from coal

mines to power plants) could possibly have adverse health effects for other parts of the local population.

Another major problem which needs further study, concerns the accumulation of trace elements, emitted from the more than ten power stations operating in the area. It is well known to the residents of Ptolemais the presence of black dust into their houses, emitted from the stacks of the power plants.

Finally the flyash waste disposal cycle and the flyash leaching properties together with the suitability of the disposal site need appropriate investigation, something which has not practised yet for Ptolemais and it has been proved that it is of prime environmental importance.

COLLECTION OF SAMPLES

Amyntaio Power Station has two pulverised lignite-fired boilers. Samples were collected from appropriate points at the production lines and include pulverised lignite (PL), flyash (FA) and bottom ash (BA). Lignite samples (OL), wooden lignite (WL). Rock samples (RA, RB, RC, RD) were also collected from the stock piles and analysed for the same elements.

As lignite that supplies the power stations varies (even daily), in its physical and chemical state because is coming from various coal-fields, it was decided to take as many samples as it was possible, together with the corresponding flyash and bottom ash. Dates of collection and sample names are shown in Table 1.

Portions of 1.5 kg for each sample were collected first, and after mixing properly, 150-250 gr were selected and were put in plastic containers with air tight covers.

ANALYTICAL METHODS

X-RAY Fluorescence (XRF) was used for the determination of 10 major and 11 trace elements, in all flyash, bottom ash and rock samples. The analyses were carried out with the use of PHILIPS PW 1400 X-RAY GENERATOR. Regarding the lignite samples, because of lack of appropriate standards the analyses carried out are semiquantitative, however the preparation and analysis was performed in the same way as that for flyash, bottom ash and rock samples.

Concerning the flyash and bottom ash, 4 replicates of each sample were prepared, and analysed for trace elements. Concerning the lignite and rock samples, 3 and 2 replicates respectively, were prepared and analysed for trace elements. Bottom ash samples lost in some cases 50-70% of weight and this possibly caused by the presence of sulphur which is volatile and lignite.

Arsenic and Cadmium were determined using classical chemical methods, but not for the whole number of samples available. Atomic Absorption Spectrometry (AAS) was used for the samples listed below: BM, BA, BJ (sample obtained from proper mixing of all the Bottom ash samples, collected in various dates in June 1992). FM, FA and FJ (sample obtained from proper mixing of all the Flyash samples collected in various dates in June 1992). LM, LA and LJ (sample obtained from proper mixing of all the Lignite samples, collected in various dates in June 1992).

The As analyses were carried out with a UNICAM 8625 UV/VIS Spectrometer, while the analyses of Cd with VARIAN GTA 96 Graphite Tube Atomiser, using palladium magnesium nitrate as matrix modifier.

Trace and major elements concentrations determined can be found in

Table 1: List of samples collected and analyzed with XRF Methods

SAMPLE NAME	COLLECTION DATE	REPLICATES ANALYZED		DESCRIPTION
		Trace	Major	
FM	May '92	4	2	Flyash
FA	April'92	4	2	Flyash
F1	1/06/92	4	2	Flyash
F2	2/06/92	4	2	Flyash
F3	3/06/92	4	2	Flyash
F4	4/06/92	4	2	Flyash
F5	5/06/92	4	2	Flyash
F6	6/06/92	4	2	Flyash
F7	7/06/92	4	2	Flyash
BM	May '92	4	2	Bottom ash
BA	April'92	4	2	Bottom ash
B1	1/06/92	4	2	Bottom ash
B2	2/06/92	4	2	Bottom ash
B3	3/06/92	4	2	Bottom ash
B4	4/06/92	4	2	Bottom ash
B5	5/06/92	4	2	Bottom ash
B6	6/06/92	4	2	Bottom ash
B7	7/06/92	4	2	Bottom ash
PLM	May '92	3	-	Pulverized Lignite
PLA	April'92	3	-	Pulverized Lignite
PL1	1/06/92	3	-	Pulverized Lignite
PL2	2/06/92	3	-	Pulverized Lignite
PL3	3/06/92	3	-	Pulverized Lignite
PL4	4/06/92	3	-	Pulverized Lignite
PL5	5/06/92	3	-	Pulverized Lignite
PL6	6/06/92	3	-	Pulverized Lignite
OL	8/06/92	2	-	Lignite (piles)
OW	8/06/92	2	-	Lignite (woody)
RA	8/06/92	2	2	Rock (Sterile band)
RB	8/06/92	2	2	Rock (Sterile band)
RC	8/06/92	2	2	Rock (Sterile band)
RD	8/06/92	2	2	Rock (Sterile band)

Tables 3 4,and 5, for lignite and rock and for flyash and bottom ash samples, respectively. The reported values of major and the most trace elements of Amyntaio samples are more or less similar to values reported for samples from other places in the Ptolemais basin (e.g. FOSCOLOS et al. 1989, FILIPPIDIS % GEORGAKOPOULOS 1992, GEORGAKOPOULOS et al. 1992a,b, 1994, KASSOLI-FOURNARAKI et al. 1993, FILIPPIDIS et al. 1994).

LEACHING TEST

The growth of coal utilisation for electricity generation has raised questions about the problem of ash disposal. Although part of the ash produced is used by the construction industries (road and concrete) a certain amount is disposed on land

(Ptolemais), or into the ocean.In Ptolemais area, the flyash by-product reaches 11,000 tons on a daily basis and approximately 4 millions tons annually.

Flyash may yield leachates under certain conditions that contain trace elements (i.e As, B, V, Cd, Cr and Mn), which are not acceptable by water-quality standards. It is apparent that, regarding the waste disposal areas, if some changes take place causing an unacceptable release of elements at some time in the future, this would be of prime environmental importance.

Other important factors are the chemical forms of the leachates, since chemical forms of Mo, W, V, Cr, As, Sb and Se have toxicological significance. Handling a waste disposal problem one must consider the short-term effects and the long-term effects with the former to be controlled by contact time, expected dilution and pH.

a.pH Measurements

The ratio: $(CaO+MgO):(SO_3+00.4Al_2O_3)$, gives an estimation on pH values of flyash: if <1.2 the leaching is expected to be acidic and if >2.2 the leaching is expected to be alkaline. The pH measurements proved the strong alkalinity of the extracts. The pH was measured in a 1:100 ash:water (distilled) ratio and after 1 minute and 10 minutes contact time the pH values obtained varied from 12.20 to 12.37.

b. Experimental methods

0.250 grams of each sample diluted in 25 ml of HCl 0.5M into a centrifusion tubes and were placed in a shaking bath. After a time period of 16 hours the first batch was taken out for centrifusion. The centrifuser used was the MSE High Speed 25 and the conditions of the actual procedure were: Temperature = 12 C, Time = 5 min, Rotation per minute = 5000. The following step was the filtration of the extracts, which was done using Whatman Membrane Filters - Cellulose Nitrate with diameter 25 mm and pore size 0.45.

The extracts from the leaching test were analysed by AAS (flame), using the Varian Spectr AA-10 available at the department of Earth Science of Leeds University. Blank samples were treated the same way through-out the procedure and standards of 0 and 1 ppm were prepared to calibrate the spectrometer.

Leaching was performed for lignite, flyash, bottom ash and shale (standard

Table 2.

Initial concentration in ppm				
Element	*Shale	Lignite	Flyash	Bottom ash
Cu (5)**	53	28	71	40
Ni (3)	122	56	170	140
Cr (5)	123	126	150	198
Leaching test Results in % of the initial concentration leached				
Element	*Shale	Lignite	Flyash	Bottom ash
Cu (5)	20	31	69	21
Ni (3)	11	50	49	32
Cr (5)	11	6	69	<5

** Standard referance material*

*** Detection limit*

reference material, obtained from the X-Ray laboratory), samples. Samples collected on May 1992 (i.e. LM, FM and BM) and the concentrations determined with XRF methods. The percentages of Cu, Ni and Cr leached determined for duplicates of each sample.

DISCUSSION AND CONCLUSIONS

The percentages, of all elements leached, were extremely high in flyash than in all other samples. Cu and Cr extracts were round 69% of the concentration in the flyash while Ni leached by 49%.

Regarding the bottom ash, leachates were in lower percentages than in lignite and therefore, elements concentrate in bottom ash are less readily leached than original lignite, fact that implies that bottom ash would have minimal or no adverse effects on receiving waters.

Comparing the results obtained for flyash and shale we concluded that Cu, Ni and Cr leached in a percentage of 300%, 420% and 690%, respectively, more than what happened in shale, and this acquires environmental significance, since presumably the inputs of these elements due to rock weathering would be in a lesser extent than in the case of weathering of flyash disposal sites.

It should be bear in mind that the experimented leaching test has no great quantitative importance for each particular element, since was applied using weak acid, but it gains an attention when comparing all samples together. In general, it could be stated that, elements concentrated in flyash particles during coal combustion, becoming more leachable than when present in "natural geological materials" like shale and coal. Atleast,this was confirmed for Cu, Ni and Cr.

Health aspects of trace elements in coal and coal residues are not a simple matter and it should be not subject to generalisations. Besides there are many factors controlling whether or not some trace elements (i.e. Pb, Hg, and Cd), could produced health implications.

However, the results obtained from the elemental analysis of flyash from Ptolemais, make possible to indicate some major points of environmental concern. As and Cd levels are high enough to put us in some consideration regarding their potential impact on the environment and the local population, either by contamination of the water supplies or by entering into the food chain in some way. Considering the millions tons of flyash produced annually, and the existence of more than 10 power plants operating in Ptolemais the "case" gains much of interest. Particulate, collected from the stacks, could be very helpful, in determining possible environmental consequences due to emissions caused by combustion of lignite and a further detailed geochemical study could qualify and quantify the problems might occur (or have already) at the flyash disposal sites.

On the other hand, the concentrations of other trace elements like U, Cu or Zn indicate that further studies (i.e. both epidemiological and geochemical), need to be carried out, in an effort to define any implications from coal development on health in the greater area of Ptolemais. Meanwhile the data obtained from elemental analysis and the monitoring itself gains a lot of interest for further research with many applications.

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Table 3: Trace elements (in ppm) of air-dried lignite and rock samples from Amyntaion power plant stations

	SAMPLE NAME								mean		
ELEMENT	PLA	PLM	PL1	PL2	PL3	PL4	PL5	PL6	PL1-6	OL	WL
Pb (5)*	9	8	8	6	7	7	7	5	7	<5	<5
Zn (3)	31	26	28	25	25	27	25	24	26	11	14
Cu (5)	35	28	22	22	25	25	24	21	23	21	19
Ba (7)	177	169	146	149	156	158	146	147	150	91	58
Cr (5)	137	126	170	147	130	145	124	136	142	64	75
Co (3)	18	18	18	20	15	18	19	17	18	17	17
Ni (3)	67	56	75	64	60	68	54	56	63	11	18
V (5)	96	89	90	91	80	82	94	92	88	70	269
U (3)	4	4	5	6	6	5	6	5	6	5	<3
Th (3)	8	8	6	6	8	8	6	7	7	4	3
Rb (2)	23	21	19	19	19	22	18	19	19	14	<2
Sr (2)	130	127	127	111	126	126	116	114	120	106	52
Y (3)	10	9	8	8	7	9	8	8	8	5	4
Nb (2)	5	4	4	4	4	4	4	4	4	4	<2
Zr (5)	36	35	33	34	33	36	33	33	34	32	<5
Sc (5)	14	12	15	13	13	15	14	12	14	8	12
As (1)	3	2							3		
Cd(0.1)	0,4	0,3							0,5		

	Rock Samples					Major Elements Analysis			
ELEMENT	RA	RB	RC	RD		RA	RB	RC	RD
Pb (5)*	<5	35	<5	<5	SiO2	2,66	58,51	57,29	15,94
Zn (3)	14	166	16	31	TiO2	0,05	1,35	1,30	0,28
Cu (5)	<5	40	<5	13	Al2O3	1,02	23,26	22,76	5,05
Ba (7)	137	543	40	114	Fe2O3	1,08	5,35	5,21	3,89
Cr (5)	<5	298	18	98	MnO	0,05	0,07	0,07	0,11
Co (3)	3	40	3	8	MgO	4,46	2,51	2,49	6,24
Ni (3)	4	169	20	109	CaO	89,49	3,28	3,20	66,13
V (5)	<5	184	8	20	Na2O	1,80	2,11	1,96	0,69
U (3)	<3	<3	<3	<3	K2O	0,22	2,72	2,65	0,50
Th (3)	4	18	5	7	P2O5	0,18	0,20	0,21	0,09
Rb (2)	4	123	7	21					
Sr (2)	263	212	98	262	Total	100,95	99,40	99,70	98,99
Y (3)	<3	40	4	5					
Nb (2)	<2	21	<2	2					
Zr (5)	<5	244	6	16					
Sc (5)	17	20	21	20					

* Detection limit

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Table 4: Trace element Results in Flyash and Bottom ash from Amyntaio power plant stations.

	SAMPLE NAME							mean		
ELEMENT	**FA**	**FM**	**F1**	**F2**	**F3**	**F4**	**F5**	**F6**	**F7**	**F1-7**
Pb (5)*	23	21	21	21	19	18	18	16	19	19
Zn (3)	77	73	91	85	61	67	65	66	60	71
Cu (5)	77	71	66	70	56	62	69	68	54	64
Ba (7)	463	469	429	460	441	438	461	445	405	440
Cr (5)	158	150	240	189	144	141	186	198	103	171
Co (3)	20	20	22	24	19	19	23	21	16	20
Ni (3)	175	170	262	214	161	173	198	206	123	191
V (5)	114	107	107	118	104	104	121	122	88	109
U (3)	14	14	14	17	19	18	17	17	16	17
Th (3)	15	14	14	15	15	14	15	14	15	14
Rb (2)	61	63	63	70	59	58	67	71	57	63
Sr (2)	331	342	330	332	336	346	330	326	326	332
Y (3)	27	26	28	27	24	25	28	28	22	26
Nb (2)	13	13	13	15	13	12	15	15	12	13
Zr (5)	97	100	110	111	106	100	118	119	92	108
Sc (5)	17	18	18	17	17	18	17	18	13	17
As (1)	16	15	-	-	-	-	-	-	-	10
Cd(0.1)	0,7	1,0	-	-	-	-	-	-	-	2
										mean
	BA	BM	B1	B2	B3	B4	B5	B6	B7	B1-7
Pb (5)*	10	7	7	<5	6	<5	5	<5	<5	5
Zn (3)	51	43	43	30	30	24	29	24	24	29
Cu (5)	51	40	19	12	8	<5	14	<5	<5	8
Ba (7)	344	319	188	186	168	95	129	98	107	138
Cr (5)	222	198	300	216	193	227	255	237	166	228
Co (3)	28	27	30	28	25	27	26	25	25	26
Ni (3)	125	118	144	90	77	64	83	67	53	83
V (5)	169	148	159	172	163	251	255	218	192	201
U (3)	9	7	7	9	8	11	12	7	8	9
Th (3)	13	11	9	8	8	5	6	5	6	7
Rb (2)	46	44	36	31	24	19	25	19	20	25
Sr (2)	183	191	128	139	132	81	100	83	89	107
Y (3)	22	19	18	14	12	9	14	11	11	13
Nb (2)	10	9	8	7	5	4	5	4	4	5
Zr (5)	70	66	60	48	41	30	40	30	30	40
Sc (5)	17	15	14	16	17	14	13	11	14	14
As (1)	3	5	-	-	-	-	-	-	-	5
Cd(0.1)	0,4	0,4	-	-	-	-	-	-	-	2

*detection limit

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Table 5: Analytical results for major elements in Flyash and Bottom ash samples in % wt.

	FA	FM	F1	F2	F3	F4	F5	F6	F7
SiO2	34,81	35,74	35,38	35,07	33,59	31,02	37,58	37,98	28,34
TiO2	0,72	0,70	0,70	0,72	0,67	0,64	0,78	0,78	0,60
Al2O3	13,85	13,57	13,62	14,21	13,46	12,55	15,28	15,47	11,89
Fe2O3	6,79	6,50	6,50	6,89	6,35	6,54	6,87	6,87	5,72
MnO	0,07	0,07	0,07	0,06	0,06	0,06	0,06	0,06	0,06
MgO	4,07	3,95	5,04	4,46	3,79	3,96	4,23	4,44	3,38
CaO	32,31	31,51	31,01	30,95	35,09	37,20	29,52	28,71	40,85
Na2O	1,54	1,92	2,33	1,37	0,86	1,35	1,93	1,16	1,18
K2O	1,16	1,20	1,19	1,26	1,14	1,10	1,31	1,30	1,09
P2O5	0,26	0,25	0,22	0,24	0,28	0,27	0,26	0,24	0,23
L.o I.	4,05	3,37	2,44	3,88	5,55	2,71	5,83	5,29	6,10
Total	95,57	95,38	96,00	95,21	95,29	94,66	97,00	97,00	93,34

	BA	BM	B1	B2	B3	B4	B5	B6	B7
SiO2	44,56	44,44	44,98	40,90	40,15	37,82	37,96	40,73	37,55
TiO2	0,88	0,83	0,86	0,80	0,65	0,81	1,31	0,78	0,95
Al2O3	16,41	15,38	15,87	15,32	12,50	13,99	14,18	14,03	14,57
Fe2O3	8,55	8,39	8,56	8,93	8,72	10,65	8,57	9,41	9,81
MnO	0,06	0,08	0,07	0,07	0,07	0,08	0,06	0,07	0,08
MgO	3,72	3,69	4,99	3,97	3,30	3,40	3,42	3,91	3,39
CaO	19,61	21,71	18,17	22,15	25,65	26,07	19,39	19,01	25,81
Na2O	0,76	0,58	0,73	0,77	0,89	0,63	0,68	0,93	0,72
K2O	1,35	1,30	1,30	1,34	1,14	1,22	1,15	1,17	1,27
P2O5	0,25	0,23	0,19	0,25	0,25	0,25	0,22	0,21	0,24
L.o.I	38,34	37,51	52,15	57,64	57,66	74,88	66,91	72,68	71,90
Total	96,16	96,88	95,70	94,49	92,61	94,60	86,96	90,36	94,20

Table 6: Final characterization of Flyash

Major Elements (%) wt, in flyash			
	MIN	MAX	MEAN
SiO2	28,3	38,0	34,5
TiO2	0,6	0,8	0,7
Al2O3	11,9	15,5	13,7
Fe2O3	5,7	6,9	6,6
MnO	0,1	0,1	0,1
MgO	3,4	5,0	4,1
CaO	28,7	40,8	32,9
Na2O	0,9	2,5	1,6
K2O	1,1	1,3	1,2
P2O5	0,2	0,3	0,2

Trace Elements (ppm) in Flyash	
Pb	19
Zn	71
Cu	64
Ba	440
Cr	172
Co	21
Ni	191
V	109
U	17
Th	14
Rb	63
Sr	332
Y	26
Nb	13
Zr	108
Sc	17
As	10
Cd	1,5

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