DETECTION AND IDENTIFICATION OF EMISSION SOURCES ON THE BASIS OF THE TRACE ELEMENT COMPOSITION OF AEROSOLS

By

J. Bérczi, G. Keömley, Zs. Molnár, and T. Várkonyi*

Nuclear Training Reactor of the Technical University of Budapest

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Introduction

Examination of solid pollutants in air and the exact knowledge of their composition are important problems in environmental analysis. Several methods are known to investigate the biologically important or toxic elements quantitatively [1, 2, 3]. A requirement imposed on these methods in general is to give information on as many elements as possible at the highest sensitivity, taking into consideration the large number of samples involved or, in other words, the suitability of the methods for large series tests. It should not be forgotten either that the elements are considered 'important' or 'toxic' on the basis of our present knowledge and, considering the extremely rapid development in the field of biology and biochemistry, the method shall be suited to determine also elements the mechanism of action of which is unknown at present. Thus, in planning for the analysis, all the elements which can be determined with the given method shall be taken into consideration. This is justified also by the fact that man's activity introduces new emission sources increasingly (e.g. nuclear power plant) and thus new problems of environmental analysis are to be solved.

Taking into consideration all what has been said so far we might be right in saying that neutron activation analysis highly meets these requirements so that it has become a useful routine method of aerosol analysis in recent years [4, 5, 6, 7]. The fundamentals, environmental applications, and possibilities of using this method are described in [8].

Nuclear technology can be ideally used in many fields of aerosol analysis [6]. Discussed in this Report are the analytical process and identification of the source of emission.

While the analytical process and the mathematical methods used are dealt with in the literature [9, 10, 11], this approach is the first in Hungarian

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* Institute for Environmental Protection.

applications. Therefore, in addition to the directly applicable informations obtained in resolving an actual problem, also the further possible applications of the method are discussed here.

Experimental

Test material

Samples were obtained from flue No. 2 of the Komló Thermal Power Plant (4 dust samples) and at environmental monitoring stations installed at an average distance of 1.5 km from the central stack of the power plant (9 samples). Sampling was made at 3 test stations at different times using Gravikon samplers and Whatman 41 paper. The amount of air passing through the sampler was 80 to 500 m^3, depending on the filter used.

Sample preparation

The environmental samples on the filter papers were placed in plastic or aluminium foils, depending on the place of irradiation (see later). The dust samples (emission samples) were put into plastic or aluminium capsules, each containing 10 to 50 mg of the sample.

Preparation of standards

As a traditional practice in our laboratory, the comparator method was used as standardizing method to avoid the preparation and measurement of standards of a very large number [8, 13]. Ruthenium was used as comparator, of which 5 to 10 mg were wrapped in aluminium foils and irradiated simultaneously with the samples.

To check the accuracy of the method, an NBS reference sample No. 1633a (coal fly ash) was prepared for irradiation and measurement (packed in aluminium or polyethylene capsules) simultaneously with the samples.

Irradiation

The samples (and standards) were irradiated in the following way:

— Emission samples and environmental samples in the nuclear reactor of the Technical University, Budapest for 15 minutes ($\Phi_{th}=2\text{–}3\cdot10^{15}$ $n\cdot m^{-2}\cdot s^{-1}$).

— Emission samples in the nuclear reactor of the Central Research Institute of Physics, Budapest, for 2 to 8 hours ($\Phi_{th}=2\text{–}3\cdot10^{17}$ $n\cdot m^{-2}\cdot s^{-1}$).

— Environmental samples similarly in the same nuclear reactor for 1 to 2 hours (because of high temperatures and radiolytic processes, the filter papers cannot withstand longer irradiation).

Measurement

The gamma spectrum of the samples was measured by means of an ORTEC Ge/Li semiconductor detector (sensitive volume: 70 cm^3, resolution: 2.0 keV for the ^{60}Co 1332 keV peak) and a measuring chain consisting of a Canberra Model 2010 main amplifier and Model 80 8000-channel 'intelligent' analyzer as well as different peripherals.

Efforts were made to optimize the irradiation time (t_i), decay time (t_d) and counting time (t_c) to obtain results for as many elements as possible, with the highest attainable accuracy. The procedure is schematically illustrated in Table 1.

Table 1

The procedure of analysis

Elements	Irradiation time	Decay time	Counting time
	t_i	t_d	t_c
Al,Ca, Cl, Mg, Mn, Ti, V	15 min.	1 to 2 min.	20 min.
Br, K, Na		2 to 3 days	20 min.
As, Au, Ba, La, Sm		7 to 10 days	3 hours
Eu, Ce, Cs, Hf, Cr, Fe, Lu, Rb, Sc, Co, Sb, Se, Sm, Tb, Th, Zn, Ta	2 to 8 hours	15 to 20 days	12 hours

The half-lives ($T_{1/2}$) and gamma energies (E_γ) suited for analytical purposes for the radioactive isotopes of the elements to be determined are tabulated in Table 2.

The measurement scheme illustrated above is largely justified by the data tabulated. The comparator method mentioned above was taken as a basis for the quantitative evaluation. The quantity of the element to be determined (m_x) was calculated on the basis of the following relationship:

$$m_x = \frac{I_x}{k_x \cdot I_{sp}^*} \tag{1}$$

where

I_x — analytical gamma peak intensity of the element to be determined

I_{sp}^* — specific intensity of the comparator isotope (* relating to the comparator)

Table 2

Half-lives ($T_{1/2}$) and gamma energies (E_γ) suited for analytical purposes for the radioactive isotopes of the elements to be determined

Element	(n, γ) product	$T_{1/2}$	E_γ (keV)
Al	^{28}Al	2.31 m	1778
As	^{76}As	26.3 h	559.2; 657
Au	^{198}Au	2.70 d	411.8
Ba	^{131}Ba	11.5 d	496.3
Br	^{82}Br	35.87 h	554.3; 777
Ca	^{49}Ca	8.8 m	3083
Ce	^{141}Ce	32.5 d	145.4
Cl	^{38}Cl	37.29 m	1642
Co	^{60}Co	5.24 a	1173.1; 1332.4
Cr	^{51}Cr	27.8 d	320
Cs	^{134}Cs	2.07 a	604.7
Eu	^{152}Eu	12.2 a	121.8; 344.2; 1408
Fe	^{59}Fe	45.1 d	1098.6; 1291.5
Hf	^{181}Hf	44.6 d	482.2
K	^{42}K	12.52 h	1524.7
La	^{140}La	40.27 h	486.8; 1595.4
Lu	^{177}Lu^{m}	155 d	208.4; 228.5
Mg	^{27}Mg	9.45 m	844; 1014
Mn	^{56}Mn	2.58 h	846.9; 1810.7
Na	^{24}Na	15.02 h	1368
Nd	^{147}Nd	11.06 d	531
Rb	^{86}Rb	18.66 d	1076.6
Sb	^{124}Sb	60.9 d	602.6; 1690.7
Sc	^{46}Sc	83.9 d	889.4; 1120.3
Se	^{75}Se	121 d	264.6; 279.6
Sm	^{153}Sm	47.1 h	103.2
Ta	^{182}Ta	115.1 d	1221.6
Tb	^{160}Tb	73 d	879.4; 965.8
Th	^{233}Pa	27.0 d	311.8
Ti	^{51}Ti	5.79 m	320
U	^{239}Np	2.35 d	228; 277
V	^{52}V	3.76 m	1434.4
Yb	^{175}Yb	101 h	396.1
Zn	^{65}Zn	245 d	1115.4

k — so called k-factor, determinable at any time independently of the actual measurement ($k = \frac{I_{sp}}{I^*_{sp}}$; I_{sp} being the specific intensity of the radioactive isotope to be determined).

In gamma-spectrometry, intensity (I) means the ratio per unit time of the background-corrected peak area (N) of the measured gamma peak, taking also into consideration the above mentioned corrections

$$I = \frac{N}{S \cdot D \cdot C} \tag{2}$$

or

$$I_{sp} = \frac{N}{S \cdot D \cdot C \cdot W} = \frac{I}{W},$$

where

$S\left(= 1 - \exp\left(-\frac{\ln 2}{T_{1/2}} t_i\right)\right)$ — saturation factor

$D\left(= \exp\left(-\frac{\ln 2}{T_{1/2}} t_d\right)\right)$ — decay factor for cooling

$C\left(= \frac{1 - \exp\left(-\frac{\ln 2}{T_{1/2}} t_c\right)}{\frac{\ln 2}{T_{1/2}}}\right)$ — correction factor for decay during measurement time and for unit counting time

I_{sp} — intensity per unit mass of the element to be determined and/or of the comparator

W — mass of the element to be determined or of the comparator (Ru in the present case)

The principle and application of the comparator techniques are described in the literature [12, 13].

The peak places the peak areas and their errors were calculated by Canberra-80, Model HP-97 programmable calcultor was used for spectral corrections if necessary, while calculations specified above were made by means of the Model TPA, small computer of the Nuclear Training Reactor. The results of measurements are tabulated in Table 3.

Accuracy, reproducibility

The reliability or, in other words, accuracy and reproducibility of the analytical method elaborated in our laboratory was checked against the NBS reference sample (coal fly ash) already mentioned. Concentrations of about 20 elements are certified while concentrations of 15 additional elements serve as recommended values in the Certificate supplied with this sample.

The relative difference between the arithmetic mean of the results of several parallel measurements and the value given in the Certificate refers to accuracy while the mean error of the mean value to reproducibility. The results of measurements tabulated in Table 4 show acceptable agreement (the deviation obtained for magnesium cannot be explained for the time being). Nevertheless, we are unaware of appreciable errors due to sampling, a fact which may be important especially in case of environmental measurements.

Evaluation of the results of measurement

As seen in Table 3, the 4 emission samples show a fair agreement with respect to the different elements. However, considering the results from the point of view of relation between environmental samples and emission (flue gas), it is by no means so simple to draw the conclusion.

Essentially, we had to answer the following questions:

— Could the responsibility of the tested emission source for environmental contamination at each measuring station and at different times) be proved?

— Could a contribution by another emission in addition to given source be assumed? Which elements can be attributed to this contribution?

To answer these questions, two in principle different approaches were adopted:

1. We tried to find relation with the emission samples on the basis of the correlation between the different elements of the environmental samples [14];
2. We used the method of cluster-analysis to analyze correlation between all test samples (both emission and environmental samples), leaving our knowledge on the origin of the samples out of consideration [15].

Correlation calculations

According to assumption, in case of identical origin, the correlation between the elements of the environmental samples should exist also for the emission samples.

According to the process described in [10], we selected first the elements which could be measured at sufficient accuracy in each sample. Such elements were Na, V, Al, Mn, K, Cs, Sc, Fe, Co, Sb, As, Zn, Th.

Then, on the basis of the measurement results of the environmental samples, the square of the linear correlation coefficient was determined for each pair of the elements listed, using the following relationship:

$$r_{i,j}^2 = \frac{\left(\sum_{k=1}^{n} X_{i,k}X_{j,k} - \sum_{k=1}^{n} X_{i,k} \sum_{k=1}^{n} X_{j,k}\right)^2}{\sum_{k=1}^{n} (X_{i,k}^2 - n\bar{X}_i^2) - \sum_{k=1}^{n} (X_{j,k}^2 - nX_j^2)} \tag{3}$$

where

n — number of chemical elements

$X_{i,k}; X_{j,k}$ — normalized concentration of the k^{th} element measured in the i^{th} or j^{th} sample, respectively (for details of 'normalization' see [14]. The purpose of normalization is to ensure identical treatment of the different elements, independent of the absolute value of their concentration. This is important first of all for the cluster-analysis discussed later).

$$\bar{X}_i = \frac{1}{n} \sum_{k=1}^{n} X_{i,k}$$

$$\bar{X}_j = \frac{1}{n} \sum_{k=1}^{n} X_{j,k} \, .$$

The calculations were made by means of a Model HP-97 programmable calculator. The results are tabulated in Table 5.

In next step, the pairs of elements for which $r^2 \geq 0.90$ (i.e. the correlation is fairly close) were selected, and the parameters of the corresponding regression lines were determined. The average of the concentration values of the 4 emission samples was calculated (and this was indeed justified because of the fair agreement mentioned) and the averages obtained were substituted into the equations of the regression lines accordingly. Then, the percentual deviation between the measured averages and the values so calculated was tabulated. The results of calculations are shown in Table 6.

On the basis of the results obtained, we arrive at the following conclusion:

— The generally high values of correlation coefficients show that the environmental samples are of very similar compositions; that means that the majority of the elements results most probably from the same source. Among the elements tested, Na, Sb, and Zn are exceptions. Note that the close correlation between Sb and Zn indicates, however, that these two elements result from the same source but this source is independent of the source of the other elements.

Table 3

Results of measurements obtained for emission and enviromental samples

Element	Concentrations												
	Emission samples (ppm or %)				Environmental samples (ng/m^3)								
	16	17	18	19	1	2	3	4	5	6	7	8	9
Al	12.8%	14.1%	12.0%	12.9%	1800	1600	680	820	530	690	2100	230	2700
As	90	90	90	90	1.2	1.1	0.4	0.35	0.29	0.5	0.4	0.15	1.9
Au	—	—	—	—	0.075	0.027	0.005	0.02	0.095	—	0.90	0.22	0.15
Ba	390	480	490	380	9.1	—	3.3	11	3.2	—	—	5.7	—
Br	—	—	—	—	0.2	0.5	0.13	0.65	0.35	0.35	0.77	0.41	—
Ce	350	350	350	300	—	—	—	2.0	1.3	—	2.5	0.6	—
Cl	—	—	—	—	0.72	1.3	0.5	2.0	1.2	1.0	1.7	1.2	3.8
Co	27	25	24	23	0.28	0.27	0.1	0.09	0.07	0.09	0.16	0.03	0.44
Cr	97	85	86	84	—	1.8	0.64	1.5	0.8	—	—	0.7	4.2
Cs	23	22	20	21	0.25	0.26	0.1	0.1	0.08	0.09	0.1	0.05	0.44
Eu	3	2.5	2.8	2.8	0.33	0.4	0.02	0.04	0.02	—	0.05	0.02	—
Fe	5.51%	5.22%	5.13%	5.14%	570	600	230	270	190	180	310	80	1100
Ga*	—	x	x	x	x	x	x	—	—	—	—	—	—
Hf	20	23	22	21	0.29	0.26	0.11	0.11	0.08	—	0.16	0.04	0.5
Hg	x	—	x	x	—	—	—	—	—	—	—	—	—
K	2.2%	2.1%	2.1%	2.1%	230	240	95	110	80	90	150	50	160
La	195	195	185	190	2.15	2.10	0.8	0.85	0.65	—	1.2	0.3	3.8

Lu	1.7	1.7	1.7	1.6	0.009	—	0.006	0.009	0.005	0.004	0.01	0.003	0.01
Mg	5.1%	4.7%	4.2%	4,5%	77	230	250	110	150	30	620	180	—
Mn	430	470	420	530	5.2	5.0	2.0	3.0	2	2.3	7.3	1	13
Na	3300	3300	3300	3200	80	86	35	120	61	57	120	45	20
Nd	177	130	—	—	—	—	—	—	—	—	—	—	—
U*	x	—	x	x	x	—	x	x	—	x	—	x	x
Rb	190	160	170	170	1.9	2.1	0.8	—	0.6	—	1.3	—	—
Sb	6	5.5	5.0	5.0	0.10	0.14	0.05	0.11	0.06	0.03	0.33	0.05	0.5
Sc	16.7	16.5	16.0	15.4	0.20	0.19	0.077	0.080	0.057	0.066	0.10	0.027	0.37
Se	43	43	37	37	—	—	—	—	0.17	—	0.9	0.07	—
Sm	26	23	23	23	—	—	—	—	0.11	—	0.02	0.04	—
Ta	14	14	13	13	0.17	0.16	0.07	0.07	0.05	—	0.09	0.03	0.32
Tb	—	6.8	5.8	6.4	0.07	—	—	0.02	—	—	0.05	—	—
Th	53	50	50	48	—	0.65	0.23	0.23	0.17	0.20	0.50	0.08	1.0
Ti	5000	5300	4600	5100	79	—	30	40	20	40	150	20	100
V	140	150	140	130	2,0	1.8	0.70	0.70	0.60	0.96	2.6	0.30	3.7
W*	—	x	x	—	x	—	—	—	—	—	—	—	—
Zn	210	200	210	170	2.5	4.0	1.2	3.6	2.0	1.9	11	1.4	11
Zr	—	—	—	—	x	—	—	—	—	—	x	—	—
Yb	8.6	10	10	9.0	0.11	0.11	0.04	—	—	0.06	—	—	0.17

* No concentrations can be given because of inavailability of appropriate K-factors

Table 4

Results of analysis of IAEA coal fly ash

Element	Reference values %	Results of individual measurements %	Mean Values of measurements %	Difference of reference values and the mean (%)	Mean error of the mean values (%)
Al	14				
Ba	0.15	0.11; 0.11	0.11	27	10
Ca	1.11				
Fe	9.4±0.1	9.3; 9.0; 9.6; 9.4; 9.7; 9.0; 9.6; 9.9	9.4	0	3
K	1.88±0.96	1.72; 1.73; 1.67	1.70	10	2
Mg	0.46±0.01	4.3; 3.9; 4.4; 4.6	4.6	?	17
Na	0.17±0.01	0.14; 0.15; 0.15; 0.15; 0.14; 0.15	0.15	12	7
Si	22.8±0.8				
Ti	0.8	0.77; 0.87; 0.72; 0.79	0.80	0	13
	ppm	ppm	ppm		
As	145±15	137; 132; 131; 131; 144; 148; 138; 149	141	0.3	0.6
Be	12				
Cd	1.0±0.15				
Ce	180	150; 150; 150; 100; 110; 200; 100	140	22	30
Co	46	46; 38; 43; 38; 39; 41	41	10	7
Cr	196±6	190; 150; 185; 167; 154; 180	170	13	12
Cs	11	9.9; 10; 9.7; 11.5	10	9	20
Cu	118±3				
Eu	4	4.5; 3.1; 4.7; 2.9; 3.9; 3.0; 3.8; 3.0	3.6	10	20
Ga	58	52; 57; 57; 61	55	5	9
Hf	7.6	5.1; 6.1; 6.6	6	20	17
Hg	0.16±0.01				
La		78; 80; 77; 78; 82; 84; 71; 74	78		5
Mn	190	193; 190; 196; 192; 186	190	0	2
Mo	29				
Ni	127±4				
Pb	72.4±0.4				
Rb	131±2	134; 125; 121	127	3	6
Sb	7	5.7; 5.7; 5.2; 5.2	5.4	22	6
Sc	40	35; 34; 35; 32; 36	35	13	6
Se	10.3±0.6	10.6; 10.6	1	7	9
Sm		16; 18; 14	15		13
Sr	830±30				
Tb		5.5; 3.2; 3.6	4		50
Th	24.7±0.3	25; 20; 21	23	7	9
Tl	5.7±0.2				
U	300	260; 320; 270; 290; 290	290	3	1
Zn	220±10				

Table 5

Square of correlation coefficients for environmental samples

	Square of correlation coefficient (r^2)												
	Sodium	Vanadium	Aluminium	Mangenese	Potassium	Cesium	Scandium	Iron	Cobalt	Antimony	Arsenic	Zinc	Thorium
Sodium	—												
Vanadium	0.71	—											
Aluminium	0.67	0.98	—										
Manganese	0.83	0.96	0.91	—									
Potassium	0.71	0.81	0.63	0.87	—								
Cesium	0.56	0.70	0.68	0.97	0.97	—							
Scandium	0.64	0.77	0.76	0.82	0.99	0.99	—						
Iron	0.67	0.77	0.76	0.82	0.99	0.99	1,0	—					
Cobalt	0.58	0.92	0.82	0.81	0.97	0.97	0.98	0.97	—				
Antimony	0.85	0.84	0.78	0.92	0.68	0.51	0.61	0.62	0.58	—			
Arsenic	0.50	0.69	0.68	0.71	0.94	0.99	0.97	0.96	0.97	0.46	—		
Zinc	0.74	0.77	0.72	0.92	0.45	0.30	0.38	0.40	0.39	0.92	0.26	—	
Thorium	0.50	0.74	0.59	0.90	0.95	0.90	0.93	0.94	0.98	0.79	0.89	0.64	—

Table 6

Comparison of concentration calculated on the basis of the regression line parameters with the measured concentrations

Pair of element X—Y	Correlation coefficient (r^2)	Regression line equation	Measured Concentration (Y)	Calculated Concentration (Y)	Deviation in %
Fe—Co	0.97	$Y=0.01+4.14\cdot10^{-4}x$	25	22	12
Cr—Fe	0.99	$Y=10.5+2470\cdot x$	5.25%	5.43%	3
K—Sc	0.99	$Y=-0.01+8.36\cdot10^{-4}x$	16.2	18.4	14
Cs—As	0.99	$Y=-0.03+4.48\cdot x$	90	99	10
Sc—Co	0.97	$Y=0.01+1.23\cdot x$	25	20	20
Co—Th	9.98	$Y=0.03+2.28\cdot x$	50	57	12
Mn—Cs	0.97	$Y=-0.02+0.04\cdot x$	22	19	14
K—Sc	0.97	$Y=9.84\cdot10^{-4}x$	22	22	0
K—Co	0.97	$Y=1.025\cdot10^{-3}x$	25	23	8
Cs—Co	0.97	$Y=1.03x$	25	23	8
Co—As	0.97	$Y=-0.02+4.24\cdot x$	90	110	22
Mn—Sb	0.92	$Y=0.02+0.04x$	5.4	18.5	240
Sb—Zn	0.92	$Y=0.69+23.7\cdot x$	200	128	36
Mn—Zn	0.92	$Y=0.9+0.97\cdot x$	200	449	115
Cs—Th	0.90	$Y=0.04+2.24\cdot x$	50	49	2
Sc—Th	0.93	$Y=0.06+2.71\cdot x$	50	44	12
Na—Mn	0.83	$Y=-1.48+0.07\cdot x$	463	230	50
Na—Sb	0.85	$Y=-0.09+2.72\cdot10^{-3}x$	5.4	9	67
Na—Sc	0.64	$Y=-0.02+1{,}62\cdot10^{-3}x$	16.2	5.3	67

— The relatively good agreement between the measured and calculated values (Table 6) also identifies the common emission source (in the present case, the Komló Thermal Power Plant) exactly. Deviation was found for elements already mentioned indicating that these elements were emitted by a different source.

Cluster analysis

The method of cluster analysis can be used to group the different elements i.e. to calculate the correlation between the different elements if the fact that both emission and environmental samples were involved in the analysis were disregarded in analyzing the data of Table 3. Instructive examples have been found in archeology, geology, botany [14, 15, 16] and more recently also in environmental studies [11] for grouping samples in such a way (without any 'a priori' information).

Essentially, the method defines a measure (similarly index) describing the difference between two samples unambiguously.

From among the measures applied [9, 15], the correlation coefficient as defined in (3) was selected. A fundamental difference was, however, that while in equation (3) the values of two elements measured in each sample had been involved in summation, here we summed all the elements two samples had in common. The choice fell on the correlation coefficient calculation because this method permitted the emission measured in ppm and the environmental samples measured in $ng \cdot m^{-3}$ to be compared directly.

After the correlation coefficients as defined above had been determined for each pair of samples, the pairs having highest $r_{i,j}$ were selected and then, after these latter had been lumped, the similarity index was calculated again now for a smaller number of samples. This procedure was repeated until all samples were involved. The results obtained were illustrated in a so called dendrogram supplied along with the calculations described by a programme (for the original programme see [14]) adapted to the Model R-32 computer of the University.

The dendrogram is essentially a system of coordinates where the sampling points are indicated along the X-axis while the similarity indices along the Y-axis. We simply interconnected the samples and groups of samples lying closest to each other. In this way, we obtained the diagram indicating the 'genetic' relation of all samples, which, as plotted by the computer, is shown in Fig. 1.

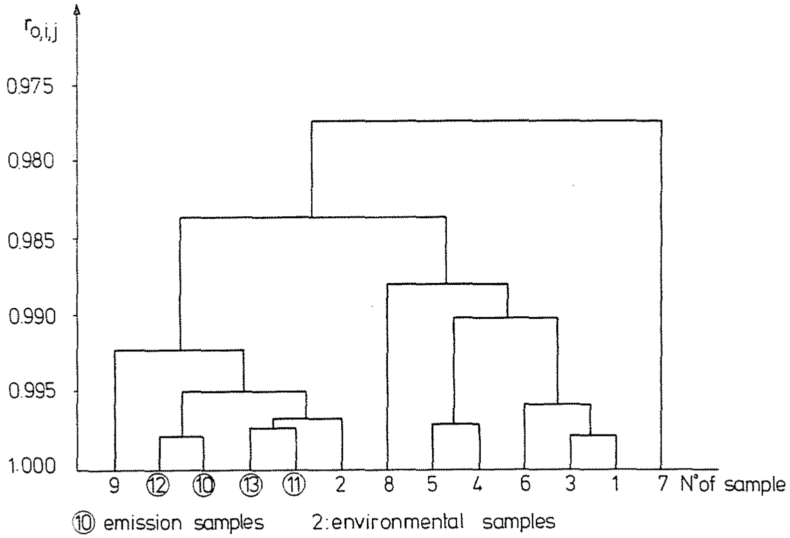

Fig. 1. Dendrogram illustrating the relation of the samples tested

In compliance with what has been said when evaluating the results of correlation calculation, the Figures show closest similarity between emission samples (samples No. 10, 11, 12, 13) but all the samples are very similar to each other so that an isolation of the samples within more groups is unjustified. It is sample No. 7 alone which slightly differs, but even this difference is not likely to be specific. All what has been said above also refers to a common source of emission of the elements.

Note that in this application the capability of cluster-analysis of grouping the samples in independent groups doesn't show up since samples of very similar composition were tested. The advantages of the method can be fully utilized if more intricate problems (more sources of emission) are involved.

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Summary

In the course of our investigations, a method to determine accurately the main and trace elements of aerosols in the atmosphere has been developed. On the basis of the results of chemical analysis, two methods, different in principle, to identify the emission sources are discussed. These methods were used to prove the doubtless relation between environmental samples and samples obtained from the flue of the power plant in different points and at different times on the basis of an analysis of emission and environmental samples resulting from the Komló Power Plant and its environment.

The analytical and computation method developed is considered to be suited for handling more complex environmental problems where more emission sources have to be identified, and their contribution to the contamination of the environment has to be determined exactly.

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Dr. János Bérczy
Dr. Gábor Keömley
Dr. Zsuzsa Molnár
Dr. Tibor Várkonyi

H-1521 Budapest
Institute for Environmental Protection