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ANALYSIS OF ELEMENTAL COMPOSITION OF ATMOSPHERIC AEROSOL IN INDONESIA

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Összefoglalás - Makra László indonéziai terepi mûszeres expedíciója során, 1996-ban 7 db aeroszol mintát gyûjtött Jáva és Bali szigetén, részben tengerparti, részben nagyvárosi, sûrûn lakott környezetben. A mintákat PIXE-módszerrel analizáltuk. Ismertetjük az aeroszol minták elemi összetételét és feldúsulásukat. A minták a klór, a kén, a réz, a cink és a neodímium szignifikáns földúsulását mutatják mind Jáván, mind Balin. Míg a klór tengeri eredetû, a kén részben antropogén, jelentõs része viszont az óceánból származik, s biogén eredetû.

Summary - During a Hungarian expedition in 1996, seven aerosol samples were collected by László Makra in Indonesia, partly in the polluted air of overpopulated Javanese towns, partly in the clean air of Javanese and Balinese seashores,. The samples collected were analysed by the PIXE-method. Elemental composition and enrichment of the elements are shown in this study. As a conclusion, chlorine, sulphur, copper zinc and neodymium are highly concentrated all over the country. While chlorine can be considered of marine origin, sulphur is partly of anthropogenic partly of biogenic origin and this latter comes from the sea.

Key words: PIXE-method, elemental composition, enrichment factor, non-crustal fraction, Java, Bali

INTRODUCTION

Indonesia is situated between the mainlands of Asia and Australia and is surrounded by the Pacific and Indian Oceans. Two-thirds of the total area of Indonesia is water whereas the country itself, with its more than 13700 islands (about 6 thousand of which is inhabited), is spread out along the Equator.

Due to the structure of the islands and their geographical location, the air over Indonesia is warm and humid with near-equal daily and yearly temperatures and water vapour near to saturation at sites near sea level. The climate is almost entirely controlled by the monsoon, especially in the southern part of the country. The Northern Hemisphere winter corresponds to the wet season whereas the dry season occurs during the Southern Hemisphere winter. The region in which air, coming from the Northern Hemisphere, meets air from the Southern Hemisphere plays an important role in the weather of Indonesia. This region is known as the Intertropical Zone of Convergence. Along this region the weather is usually cloudy and rainy, and sometimes a line of cumulonimbus clouds, with tops reaching 15 km or more, can be found.

In general, the wet season prevails in the area north of the Conwergence Zone, and the dry season prevails south of it. Although the season is called "dry", this does not necessarily mean that there is no rain at all. Wet and dry seasons are distinguishable but, in general, there is no month without rainfall. In fact, according to Köppen's classification, a tropical rainforest climate predominates in Indonesia (*Landsberg*, 1969).

The Indonesian islands are situated just at the border of the Asian and Australian mainlands. Hence this is the region of frequent structural movements, consequences of which are earthquakes and volcanic eruptions. The country is usually called as the land of volcanoes. There are altogether 240 volcanoes here, from which 80 are working even nowadays. Because of the intensive volcanic activity, soil is very fertile and is full with minerals.

According to our examinations samples were taken in Java and Bali Islands. While Java is overpopulated and concentrated with heavy industry, on Bali there are no industrial sources for air pollution.

The elemental composition of atmospheric aerosol particles has been studied widely during the last three decades. Aerosols play an important role in the atmosphere, e.g. by modifying radiation, cloud and fog formation and precipitation; even they influence human health. The reason for such measurements is to estimate the environmental impact of the particles on local, regional and global scales. Among other things, samplings in the atmosphere, far from human activities, have been carried out under oceanic and continental conditions to investigate the global cycle of aerosols (e.g. *Mészáros*, 1978; *Penkett et al.*, 1979; *Adams et al.*, 1980, 1983; *Winchester et al.*, 1981a, 1981b; *Morales et al.*, 1990). On the other hand results may show characteristics of anthropogenic pollution in overpopulated cities in Asia being highly industrialised compared with background regions (e.g. *Winchester et al.*, 1981a, 1981b; *Hu et al.*, 1987; *Winchester and Wang*, 1989; *Hashimoto et al.*, 1994; *Ning et al.*, 1996; *Zou and Hooper*, 1997).

Several papers have been published about air pollution characteristics of urban and non-urban regions of Indonesia (e.g. *Zou and Hooper*, 1997; *Cohen*, 1998). According to the analysis of *Zou and Hooper* (1997), crustal dust contributes significantly to both coarse and fine fraction of aerosol in the air of Jakarta. Nevertheless few information is available on the aerosol in the air over Indonesia. Elemental composition of atmospheric aerosols was analysed in the Asian region e.g. over Lake Baikal (*Van Malderen et al.*, 1996); Northern China (*Winchester et al.*, 1981a, 1981b; *Fan et al.*, 1996); and the Loess Plateau, China (*Zhang et al.*, 1993).

One of the frequently applied techniques of elemental analytics of atmospheric aerosols is the PIXE method. Possibilities of this procedure are discussed, among other papers, in comprehensive studies of *Koltay* (1990), furthermore *Maenhaut and Malmqvist* (1992). Authors of the present manuscript applied this technique, among other papers, in *Borbély-Kiss et al.*, (1991, 1999), *Molnár et al.*, (1993b, 1995) for qualification of regional aerosols. Technical details are given in these papers.

The aim of this paper is to characterise the elemental composition of aerosol samples collected in the air over Indonesia.

SAMPLING AND ANALYSIS

Atmospheric aerosol particles were collected on Nuclepore polycarbonate filters of 15 mm diameter with pore size of 0.4 μm. After samplings, the filters were placed into small plexiglas boxes sealed carefully. The filters were kept in these boxes until their analysis in Hungary.

We collected four samples in Java and three in Bali Island (*Fig. 1*). The latter mentioned three samples were taken at Adika Sari, Bali, at the same site. Three of the samples were taken from urban areas [Jakarta, Bandung and Yogyakarta (Java Island)] and the other four samplings were performed at seashore [Carita, Labuan (sampling site No. 1.), one measurement and Adika Sari, Bali (sampling site No. 5.), three measurements]. Carita and Adika Sari (with four samples connected actually to two sites) can be considered as background areas. This means that they are not influenced directly by local pollution sources. The geographical positions and characteristics of the sampling sites are given in *Fig. 1* and *Table 1*.

Table 1 Date of samplings; geographical positions and description of sampling sites, Indonesia

Sample No.	**Date**	**H (m)**	**φ (°S)**	**λ (°E)**	**Site description**
1	04 May	2	6° 50'	105° 57'	Carita, Labuan (seashore)
2	07 May	7	6° 11'	106° 50'	Jakarta (urban site)
3	09 May	709	7° 05'	107° 56'	Bandung (urban site)
4	11 May	116	7° 38'	110° 34'	Yogyakarta (urban site)
5a	17 May	12	8° 41'	115° 26'	Adika Sari, Bali (seashore)
5b	19 May	12	8° 41'	115° 26'	Adika Sari, Bali (seashore)
5c	21 May	12	8° 41'	115° 26'	Adika Sari, Bali (seashore)

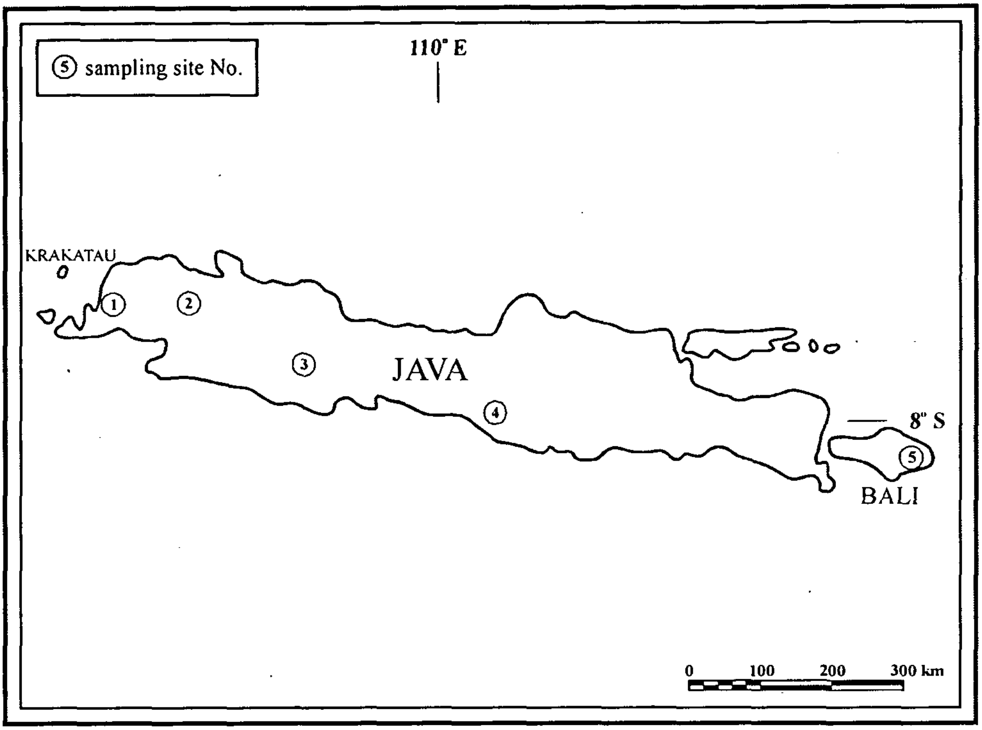

Fig. 1 Position of sampling sites in Indonesia with spatial distribution of elements enriched over ten, excluding sulphur, chlorine, copper and zinc

The sampling was performed at 1.5 m height. The filter holder was connected to a vacuum pump, which was operated at a flow rate of approximately 20 l min^{-1}. The pump was equipped with an automatic timer system and a gas meter. The gas meter was calibrated before the programme. On the basis of this calibration the accuracy of volume measurements was ± 2 %. The sampling time was about 1.5-2 hours during which at least 1 m^3 but not more than 2 m^3 of air was sampled. The electric current to function the pump was supplied by an electric generator. Exhaust gases from the generator were conducted downwind with a tube. Thus seven aerosol samples were obtained.

The elemental composition was determined by the particle induced X-ray emission (PIXE) method. The details of the analysis are described elsewhere (*Koltay*, 1988). The samples were irradiated by 2 MeV proton beam supplied by the *Van de Graaff* nuclear accelerator of the Nuclear Research Institute of the Hungarian Academy of Sciences, Debrecen, Hungary. Eighteen elements (with atomic number 13 and over) were detected by this procedure: Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Ba, Nd and Pb. The detection limits are between 2-100 ng m^{-3} depending on the elements. The blank value for each element was carefully checked. It was found that, except for bromine, blanks could practically be neglected compared to measured concentrations. For this reason concentrations and enrichment factors for bromine were omitted from further examinations. The error of quantitative analysis was lower than 10 % as discussed in more details by *Koltay* (1988).

RESULTS AND DISCUSSION

The absolute concentrations as well as the enrichment factors of 17 elements found in the samples are shown in *Table 2*.

Table 2 Concentration (x: ng m^{-3}) and enrichment factor (EF) of different elements, relative to average crust data, in the atmospheric aerosol over the regions of Indonesia

Ele-ments	*Sample 1*		*Sample 2*		*Sample 3*		*Sample 4*		*Sample 5*		*Sample 6*		*Sample 6*	
	x	*EF*	*x*	*EF*	*x*	*EF*	*x*	*EF*	*x*	*EF*	*x*	*EF*	*x*	*EF*
Al	302	0.54	421	0.46	869	0.53	255	0.58	222	0.80	< 120		< 120	
Si	514	0.27	1415	0.45	2775	0.50	1043	0.69	498	0.53	164	0.43	294	0.52
S	813	458.7	603	204.1	357	68.7	317	223.5	488	550.5	351	990	419	787.9
Cl	5138	5798	105	71.1	277	106.5	756	1066	2932	6615	1396	7874.5	1847	6946.1
K	218	1.2	181	0.61	310	0.6	300	2.1	208	2.4	134	3.8	118	2.2
Ca	654	2.6	974	2.4	1882	2.6	677	3.4	456	3.7	159	3.2	407	5.5
Ti	30	=1	50	=1	88	=1	24	=1	15	=1	6	=1	9	=1
V	11	12	7	4.6	10	3.7	3	4.1	9	19.6	6	32.6	5	18.1
Cr	32	46.9	30	26.4	23	11.5	17	31.2	37	108.5	35	256.7	30	146.7
Mn	17	2.6	21	1.9	36	1.9	15	2.9	7	2.2	< 5		6	3.1
Fe	439	1.3	627	1.1	1093	1.1	279	1.02	157	0.92	65	0.95	97	0.95
Ni	5	9.8	5	5.9	8	5.3	< 3		4	15.6	2	19.6	3	19.6
Cu	28	74.7	20	32	10	9.1	6	20	38	202.6	13	173.3	7	62.2
Zn	11	23.1	48	60.3	30	21.4	14	36.7	4	16.8	< 6		3	21
Ba	46	15.9	14	2.9	16	1.9	7	3	26	17.9	16	27.6	23	26.5
Nd	64	335.3	19	59.7	26	46.4	15	98.2	41	429.5	22	576.2	31	541.3
Pb	< 12		31	209.8	217	834.6	22	310.2	< 12		< 12		< 12	

Considering the absolute concentrations, the concentration of chlorine is found to be at least one order of magnitude higher than that of the other elements in case of the seashore sites. Following chlorine, silicon, sulphur and iron show the highest concentrations. The concentration of silicon is one order of magnitude higher over urban areas (Jakarta, Bandung and Yogyakarta) contrary to seashore sites.

The enrichment of element X relative to average earth crust composition is given by the enrichment factor $EF_{Ti}(X)$ defined as

$$EF_{Ti}(X) = (X/Ti)_{air} / (X/Ti)_{crust} , \qquad (1)$$

where Ti is used as a reference crust derived element (*Mason*, 1966), $(X/Ti)_{air}$ is a concentration ratio of an element X to that of Ti in atmospheric particulates and $(X/Ti)_{crust}$ is a concentration ratio of an element X to that of Ti in crust. By using this approach, enrichment factors less than 1 for Al and Si were found at each station, which means that even Ti is partly enriched. The use of titanium as reference element is due to the applied analytical method. At PIXE-analysis the detection limit and the accuracy of the method depend on the atomic number of the given element. The error of determining Al is bigger than that of Ti. Furthermore, Ti has less other (anthropogenic) sources than Al and Fe (see e.g. *Borbély-Kiss et al.*, 1991).

According to the enrichment factors aluminium, silicon, potassium, calcium, manganese and iron are normally taken to be crustal elements, namely their EF at continental sites are below ten, while that of sulphur, chlorine and chromium is well over ten. The enrichment of these elements requires some explanation. Sulphur seems to be enriched everywhere over Indonesia. Generally, most part of S in the air can be found in fine modes of aerosols (size of them is less than 2.5 μm) which are of secondary origin. These aerosols transform to ammonium sulphate drops by chemical processes and can reach remote regions from their sources. In polluted urban areas it is due to oil combustion and industrial thermal processes, while at seashore sulphur comes from other source. Atmospheric sulphates are either emitted directly as primary particles or are products of gas-to-particle conversion which takes place in the atmosphere. Subsequently they can undergo numerous reactions and give rise to a wide variety of sulphates. Sources of S in the atmosphere are well known; oxidation of natural H_2S and $(CH_3)_2S$ results finally in SO_2. On a global scale, the anthropogenic input of SO_2 is also very important. Biological particles can provide an attractive nucleating surface for SO_2 absorption and conversion to sulphates (*Mamame and Noll*, 1985). The absolute concentration of sulphur at sampling site No. 1. is the highest of all sites. This might partly be originated from the Krakatoa volcano settled in the Sunda strait, about 40 km away from the coasts of Western Java. During the period of sampling at site No. 1, the Krakatoa was very active with smoke and ashes following heavy explosions. The EF of chlorine at each site (with the exception of site No. 2.) is well over 100. However, chlorine can be considered of marine origin all over Indonesia, since even typical non-seaside cities are not further from the ocean than 200 km. At sites near to seashore (about 200 km away from the sea) some 80 % of chlorine might be of marine origin (*Cohen et al.*, 1996; *Cohen*, 1998). *Cohen* (1998) received similar result for Jakarta. We did not analyse sodium which, being of marine origin, is also enriched in Jakarta (*Zou and Hooper*, 1997). According *to Cohen et al.* (1996), about 70 % of sodium at seashore sites is of marine origin. The enrichment of chromium in Indonesia shows similarity with that in North-western China. Cr might be partly of industrial origin both in urban sites (Jakarta, Bandung and Yogyakarta) and at seashore. Contrary to this assumption, urban sites show both lowest absolute concentrations and lowest EFs of chromium. Higher concentrations of chromium at seashore sites and its one order of magnitude higher EFs at sampling site No. 5, relative to those at urban sites, can be traced back partly to its soil origin. However, no local samples of soil are at disposal to confirm this hypothesis (*Table 2*).

The rest of the elements (nickel, copper, zinc, barium, neodymium and lead) are mainly of non-crustal ones. According to their enrichment factors, our most important results are that copper, barium and neodymium are enriched at seashores one order of magnitude higher than at urban sites; furthermore, lead is detected and at the same time highly enriched only at urban sites. The cities, where samplings were made, are the most populated (Jakarta: 10 million people; Bandung: 0.5 million people; Yogyakarta: 1 million people) and are full of various types and quality of cars. As an example Jakarta has the second largest traffic in the world (after Bangkok). The high concentration of lead in the cities origins directly from the gasoline of vehicles. Lead compounds can be on the one hand sulphates which are the result of automobile exhausts, on the other hand halides which are emitted by cars and can easily be converted into sulphates by reactions with SO_2 or H_2SO_4 (*Van Malderen et al.*, 1996). In accordance with our results *Zou and Hooper* (1997) found also high concentration of lead and even zinc in Jakarta. Nickel, copper and zinc seem to be enriched all over Indonesia. They are mainly products of non-ferrous metallurgy. Seashores are enriched with them, probably owing to their short distance from industrial centres (*Fig. 1* and *Fig. 2*).

The fraction of elements coming from non-crustal sources [$(c_x)^*$] can be calculated by the following formula (*Mason*, 1966):

$$(c_x)^* = \frac{(c_x)_{air} - (c_{Ti})_{air} \cdot (\frac{c_x}{c_{Ti}})_{crust}}{(c_x)_{air}} \text{, where} \qquad (2)$$

$(c_x)_{air}$: is the concentration of an element x in the air
$(c_{Ti})_{air}$: is the concentration of titanium in the air
$(c_x)_{crust}$: is the concentration of an element x in the crust
$(c_{Ti})_{crust}$: is the concentration of titanium in the crust.

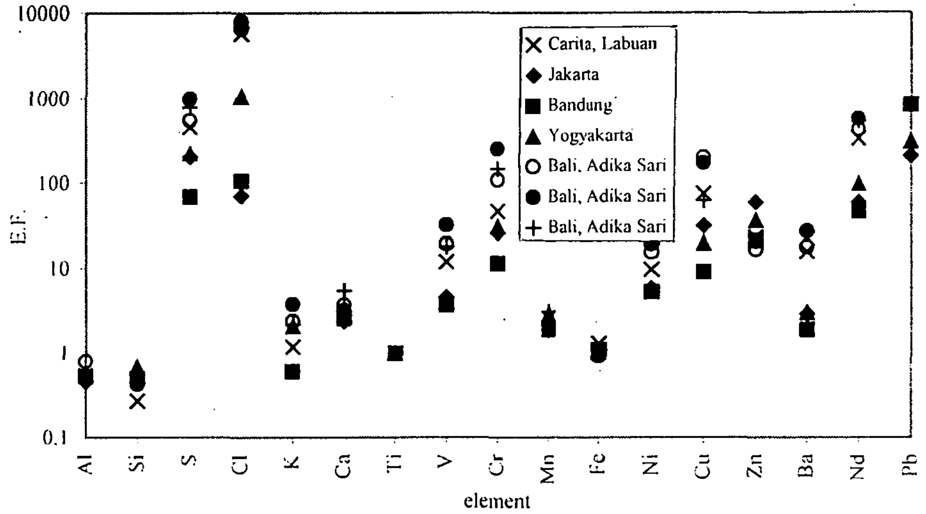

Fig. 2 Enrichment factors of 17 elements at five Indonesian sites

The non-crustal fractions of the detected elements are shown in *Table 3*. According to the results a substantial part of potassium, calcium, manganese and iron comes to the air from the Earth' crust, while aluminium and silicon is entirely of crustal origin. V, Cr, Ni, Cu, Zn, Ba, Nd and Pb are mostly emitted by anthropogenic sources. Chlorine comes basically from the sea, while a substantial part of sulphur originates from anthropogenic sources.

CONCLUSION

Absolute concentrations of chlorine are higher than those of sulphur only at seashore sites. S and Cl are highly enriched at each sampling sites. Most part of chlorine is supposed to come from sea spray, while a substantial part of sulphur presumably comes also from natural source, from biogenic emission of the sea. Chromium is highly enriched at seashore sites, much higher than at urban sites. This shows similar result to high chromium concentrations in the Takla Makan Desert (*Molnár et al.*, 1993a; *Makra et al.*, 1999), which confirms our hypothesis of its soil origin. Cu, Zn, Ba and Nd are highly enriched at each site, while Pb is only enriched at urban sites. They all can mostly be attributed to anthropogenic sources.

Table 3 Non-crustal fractions of aerosol particles, relative to average crust data, %

Ele-ments	*Sample No.*						
	1	*2*	*3*	*4*	*5a*	*5b*	*5c*
Al	0	0	0	0	0	0	0
Si	0	0	0	0	0	0	0
S	100	100	99	100	100	100	100
Cl	100	99	99	100	100	100	100
K	17	0	0	52	58	74	55
Ca	62	58	62	71	73	69	82
Ti	0	0	0	0	0	0	0
V	92	78	73	76	95	97	94
Cr	98	96	91	97	99	100	99
Mn	62	47	47	66	55		68
Fe	23	9	9	2	0	0	0
Ni	90	83	81		94	95	95
Cu	99	97	89	95	100	99	98
Zn	96	98	95	97	94		95
Ba	94	66	47	67	94	96	96
Nd	100	98	98	99	100	100	100
Pb		100	100	100			

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