URSZULA TOMZA *

ATMOSPHERIC LEVELS OF TRACE ELEMENTS IN KATOWICE

74 aerosol samples were collected on Whatman 41 filters in Katowice, between August 21, 1979 and January 7, 1980. The concentrations of 38 elements (Na, Mg, Al, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Mo, Ag, Cd, In, Sb, I, Cs, Ba, La, Ce, Sm, Eu, Lu, W, Au, and Th) were measured by means of instrumental neutron activation analysis (INAA) method.

Mg, Al, Cl, K, Ca, Fe, Cu, and Zn account for about 85% of the trace element content in the atmospheric aerosol of Katowice, measured by this method. The total aerosol mass, however, measured on Whatman filter samples account for about 25% of the total suspended particulates (TSP) because the elements like carbon, oxygen, nitrogen, sulfur and silicon are not measured by INAA.

1. INTRODUCTION

The town of Katowice is the area of major environmental interest because of the magnitude of its gaseous and particulate emissions. It has been estimated in 1980 that particulate emissions from the main industrial activities in the province of Katowice (6650 km^2) amounted to 7×10^5 t per year, 30% of the total particulate emissions in Poland.

In the past, source-oriented techniques such as emissions inventories and predictive diffusion models were used to trace pollutants from source to receptor. Conventionally, emission rates and associated particle sizes [1]–[3] were determined by in-stack measurements. However, this kind of measurements may not represent the reality, because once particles are emitted from the stack there may occur the changes of their sizes, especially due to the changes in plume temperatures and relative humidity while mixing in the ambient air.

Because of large uncertainties of source-oriented models, the receptor-oriented ones are developed. In these models the concentrations of species at sampling site

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and their fluctuations are used to estimate the contributions to various classes of sources.

Most analyses, which are routinely carried out in Poland, provide information about the total suspended particulates (TSP) and concentrations of some selected elements (Zn, Pb, Fe, Mg, Cu, Mn, Cd) in airborne particulates [4]. However, the concentrations of minor and trace elements are also of interest since they may be responsible for some health effects. Consequently, it is necessary to have at ones disposal the reliable and complete data on the composition of atmospheric aerosol for as many individual elements as possible.

In this work a 2 month survey of atmospheric concentrations of more than 35 trace elements was undertaken. The town of Katowice was considered to be suitable for such a survey, as it is a heavily industrialized complex with a diverse and changing pattern of industry, including coal mining, steel production and non-ferrous metallurgy as well as coal-fired electrical power generation. This complex agglomeration of major sources might provide both high concentrations and strong temporal variations of trace elements. Furthermore, various source processes tend to maximize the number of detectable elements what makes the application of such a multielemental analytical procedure as INAA extremely useful.

2. EXPERIMENTAL

The study of airborne particulate matter in the town of Katowice was initiated in 1979 and some preliminary results have already been published elsewhere [5], [6].

Nearly all samples were collected at the Institute of Physics of the Silesian University, near the center of the town. At the end of the sampling period, however, the collection devices were transferred to an apartment building about 1 km from the original sampling location. The Whatman filter holder ($\Phi = 11$ cm) was situated at the open window (fourth floor at the Institute of Physics, third floor of apartment building) with the air intake facing outside. The average flow rate was about 100 dm^3/min and face velocity about 20 cm/s. The first sampling period (referred to as summer) started on August 10 and lasted to September 21, 1979. The second one (winter) was of shorter duration and lasted only for 2 weeks (December 21, 1979–January 7, 1980). Over the two sampling periods combined 74 Whatman filter samples were collected. All these samples were analysed by INAA method at the reactor facilities of the Institute for Nuclear Sciences, University of Ghent, Belgium.

The procedure was similar to that of SCHUTYSER et al. [7] and involved 2 separate irradiations. (for 5 min and 7 h) with subsequent measurement of short- and long-lived product radioactive radionuclides by means of a large Ge(Li) detector [8]. Figure 1 gives one of typical gamma spectrum of an aerosol sample
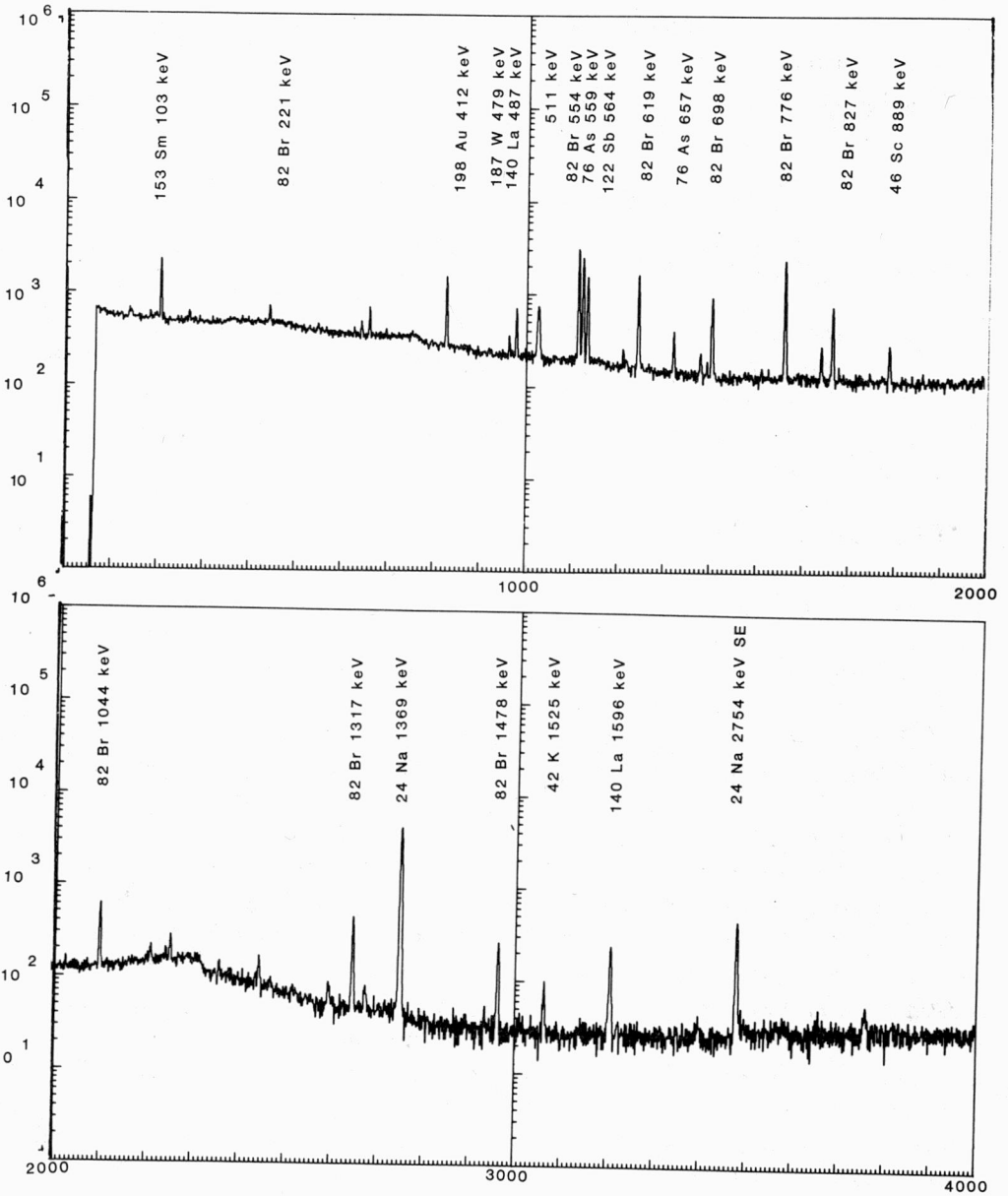
Fig. 1. Gamma-ray spectrum of an aerosol sample $t_{irr} = 7$ h, $t_{cool} = 72$ h, $t_{count} = 2$ h

irradiated for 7 h with the neutron flux of 3×10^{12} n/cm^2 s and recorded 3 days later.

The accuracy of the method was investigated by analysing standard reference materials for coal (SRM 1632a) and for coal fly ash (SRM 1633a) from the

National Bureau of Standards. The precision of the analysis, calculated from counting statistics, was better than 10% for most elements except for Mg, Ti, Ga, Ag, Cd, In, and Lu where analytical errors were up to 25%.

3. RESULTS AND DISCUSSION

Tables 1 and 2 summarize the concentration data obtained by INAA of Whatman filter samples for summer and winter periods, respectively. They present the ranges, the arithmetic and geometric means and median concentrations of elements. All the concentrations are consequently represented in ng/m^3 of the air applicable to the air temperature and pressure at the sampling time. Of 38 elements detected by INAA 35 elements have been measured in nearly all samples; the remaining elements (Ni, Sr, and Mo) were rarely above detection limits [9].

The tables include also standard deviations (in absolute values) of the arithmetic and geometric means as the measures of the data variability and the representativeness of the averages. Most of the elements show moderate variability with the time and their maximum to minimum ratio ranged within 10–20. For the elements like Cr, Mn, Zn, As, Se, Cd, In, and I, however, larger ranges of concentrations and, consequently, higher standard deviations were noticed.

A strong difference between the arithmetic and geometric averages stated in most elements indicates not exact log-normal distribution of elemental concentrations. A log-probability plot for Al, Si, S, K, V, Cr, Mn, Fe, Zn, As, Se, Br, La, and Pb is shown in fig. 2. As can be seen Al, S, K, V, Fe, As, and La seem to be log-normally distributed, but for Si, Cr, Mn, Zn, Se, Br, and Pb more or less important departures from the ideal log-normal distribution are observed.

When compared our data to mean atmospheric concentration levels in other countries certain agreement is apparent. The atmospheric concentrations of most elements in Katowice are of the same order of magnitude as those reported for the most industrialized areas in the world [10]–[12]. Besides this general similarity there occur also some distinct differences due to different nature of the industries in the area considered. The atmospheric levels of Al, Cr, Mn, Cu, and Se, which seem to be excesively higher, especially for winter, are more likely due to a larger combustion activity taking place in winter and to production of pig-iron, steel and non-ferrous metals [13].

It is worthwhile to note that the elemental concentration levels measured by us are of the same order as the existing elemental data for the Katowice region (tab. 3).

Copper concentrations measured on Whatman filters (tabs. 1 and 2) are unrealistically high in comparison with the corresponding values obtained by the author on Nuclepore filters and cascade impactor samples [6], as well as with the reported concentration of Cu in the current literature [12]–[14]. The increased

Range, arithmetic mean, geometric mean, and median for the concentrations of individual elements
(ng/m^3) measured by INAA on Whatman filters (summer)

Element	Range ng/m^3	Arithmetic mean ± standard deviation	Geometric mean (standard deviation)	Median	Numbers of samples analyzed
Na	400–2800	1120 ± 550	1000 (1.6)	1050	37
Mg	300–7800	1880 ± 1900	1320 (2.3)	1280	28
At	1500–13600	6680 ± 3000	6000 (1.6)	6500	37
Cl	1030–9200	3200 ± 2060	2720 (1.8)	2800	37
K	960–7760	2760 ± 1521	2410 (1.7)	2300	35
Ca	< 500–23000	5630 ± 4050	4630 (1.9)	4400	35
Sc	0.5–3.1	1.3 ± 0.6	1.2 (1.6)	1.2	37
Ti	150–1100	480 ± 250	430 (1.7)	410	34
V	5.5–35.8	20.0 ± 8.2	18 (1.6)	18.5	37
Cr	1.8–190	46 ± 36	35 (2.3)	36	37
Mn	80–7800	710 ± 1430	320 (2.9)	250	37
Fe	1760–17500	6700 ± 3870	5700 (1.7)	5600	37
Co	1.7–14.7	5.5 ± 3.5	4.5 (1.9)	4.4	37
Ni	< 13–104	54 ± 30	46 (1.8)	< 35	7
Cu	340–3900	1740 ± 960	1500 (1.8)	1380	37
Zn	250–11006	1760 ± 2350	1140 (2.4)	920	37
Ga	1.6–46.0	8.9 ± 9.2	6.7 (2.0)	5.7	32
As	2.7–51	15.7–9.9	13.1 (1.8)	13.5	37
Se	2.8–120	14.5 ± 18.9	10.3 (2.0)	10.3	37
Br	33–570	120 ± 100	97 (1.9)	90	37
Rb	6.5–49	18.7 ± 9.5	16.6 (1.6)	14.8	33
Sr	< 35–520	190 ± 220	120 (2.7)	70	4
Mo	2.7–32	13.0 ± 9.5	9.2 (2.4)	7	8
Ag	< 0.9–22	5.2 ± 5.6	3.7 (2.3)	2.9	15
Cd	3.4–250	36 ± 50	22 (2.6)	15.6	31
In	0.03–1.3	0.140 ± 0.217	0.098 (2.0)	0.084	34
Sb	5–50	19.7 ± 11.9	16.5 (1.8)	17.6	37
I	9–1740	200 ± 330	73 (4.3)	80	37
Cs	0.6–7.0	2.6 ± 1.5	2.3 (1.8)	2.3	37
Ba	54–1080	270 ± 230	205 (2.1)	180	27
La	0.8–6.4	3.3 ± 1.4	3.0 (1.6)	3.3	37
Ce	1.6–46	11 ± 9	8.4 (2.1)	8.4	36
Sm	0.12–1.13	0.606 ± 0.250	0.550 (1.6)	0.55	32
Eu	0.05–0.37	0.190 ± 0.103	0.160 (1.8)	0.12	18
Lu	0.012–0.106	0.050 ± 0.030	0.040 (1.8)	0.022	22
W	0.9–72.7	16.4 ± 20.7	8.4 (3.2)	7.2	37
Au	0.002–0.130	0.038 ± 0.040	0.020 (3.0)	0.020	36
Tn	0.3–3.9	1.30 ± 0.75	1.14 (1.7)	0.90	29

Table 2

Range, arithmetic mean, geometric mean, and median for the concentrations of individual elements (ng/m^3) measured by INAA on Whatman filters (winter)

| Element	Range ng/m³	Arithmetic mean ± standard deviation	Geometric mean (standard deviation)	Median	Numbers of samples analyzed
Na	1600–20000	4030 ± 3170	3460 (1.6)	3000	36
Mg	970–8400	4000 ± 2300	3340 (1.9)	1800	22
Al	2400–83100	16000 ± 13800	12500 (2.1)	12100	36
Cl	7000–52700	15200 ± 9050	13600 (1.5)	12500	36
K	1770–14390	5100 ± 2500	4700 (1.6)	4900	34
Ca	1700–17000	8000 ± 4400	6700 (1.9)	6100	31
Sc	0.6–13.0	2.9 ± 2.1	2.4 (1.9)	2.8	36
Ti	200–3700	970 ± 670	820 (1.8)	580	25
V	13 – 130	40 ± 20	37 (1.6)	36	36
Cr	20–470	105 ± 96	80 (1.9)	74	36
Mn	70–3400	580 ± 650	350 (2.8)	360	36
Fe	3500–30900	8900 ± 5440	7700 (1.7)	7700	36
Co	2.7–42.4	9.1 ± 7.6	7.4 (1.8)	6.1	36
Ni	80–190	110 ± 50	100 (1.5)	94	4
Cu	210–4400	1080 ± 1020	790 (2.1)	600	31
Zn	490–17200	2030 ± 2800	1400 (2.1)	1350	36
Ga	8.3–67	22 ± 12	19 (1.6)	18.4	33
As	18–170	44 ± 30	38 (1.7)	35	36
Se	8.8–440	40 ± 90	22 (2.4)	18	36
Br	90–1280	320 ± 270	260 (1.8)	230	36
Rb	10–97	30 ± 17	30 (1.6)	31	34
Sr	120–340	220 ± 80	208 (1.5)	210	6
Mo	9–70	30 ± 36	19 (3.2)	11	3
Ag	1.5–65.0	14 ± 20	7.6 (3.1)	6.0	8
Cd	6.0–290	60 ± 70	35 (2.9)	25	20
In	0.097–0.680	0.240 ± 0.150	0.220 (1.70)	0.190	34
Sb	11–260	58 ± 56	42 (2.2)	40	36
I	18–90	49 ± 20	45 (1.5)	43	36
Cs	1.9–15.0	5.1 ± 2.5	4.6 (1.6)	5.0	36
Ba	< 90–1150	470 ± 290	410 (1.7)	370	14
La	1.8–45	7.6 ± 7.1	6.2 (1.8)	6.0	36
Ce	4.4–65	14.3 ± 10.5	12.2 (1.7)	11	35
Sm	0.6–15.0	2.0 ± 2.0	1.5 (1.9)	0.9	23
Eu	0.21–1.30	0.520 ± 0.370	0.420 (1.9)	0.30	8
Lu	0.037–0.280	0.110 ± 0.057	0.099 (1.7)	0.098	21
W	1.40–380	18.5 ± 64.9	5.6 (3.0)	4.1	36
Au	0.001–0.230	0.078 ± 0.066	0.050 (3.2)	0.043	32
Th	0.5–12.7	2.5 ± 2.0	2.0 (1.9)	2.1	36
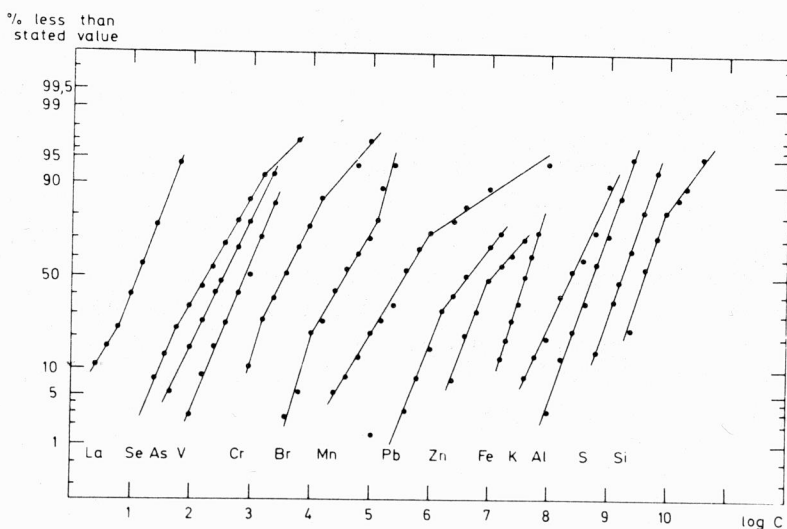
Fig. 2. Log-probability plots for concentrations of selected elements of the Katowice aerosol

Table 3

Atmospheric concentrations of some pollutants in the Katowice region. A comparison of the present and literature data (ng/m^3)

Pollutant	Katowice [a] this work	Katowice [b] [4]	Siersza [14]
TSP	250000 [c]	295000	—
Mn	645	654	—
Cr	75	24	—
Fe	7800	20000	9800
Co	9	—	5.2
Cu	1300	525	—
Zn	1700	2190	2470
As	30	—	69
Se	30	—	7.5
Cd	50	35	—
Pb	480 [d]	1110	670
Th	2	—	3.5

[a] — averaged over summer and winter, Whatman filters.
[b] — averaged over 5 years, 1978/1982.
[c] — averaged over 6 Whatman filters for winter.
[d] — measured by PIXE method [6].

copper level on Whatman filters was assumed to be debris from malfunctioning pump motor within the sampler as such an effect has been suggested before [15]. However, experiments with 2 successive Whatman filters did not show any significant contamination of the second filter with Cu and the increased amount of Cu on Whatman filters remains unclear.

3.1. TOTAL AEROSOL MASS MEASURED AND ITS MAJOR COMPONENTS

Total suspended particulates (TSP) were determined by weighing the filters before and after sampling and by re-equilibration for at least 24 h at $50^0/_0$ relative humidity [16]. The accuracy of TSP obtained in this manner is of order of $20^0/_0$. As this procedure was applied to a limited number of Whatman filters an attempt was made to account for the total aerosol mass being measured by INAA method. Table 4 presents the average total aerosol mass (the sum of elemental concentrations measured). For comparison the range and arithmetic mean of TSP with the associated standard deviation are listed. It appears that the total aerosol mass measured by INAA of Whatman filters accounted for about $25^0/_0$ of TSP. The remaining about $75^0/_0$ is composed largely of carbon, oxygen and nitrogen, the main missing elements not measured by INAA.

Table 4

Total aerosol mass measured by INAA (ng/m^3)

| | Total aerosol mass measured by INAA | | |
	Range	Arithmetic mean ± standard deviation	Numbers of samples
Summer	10–70	30 ± 16	37
Winter	20–180	60 ± 30	36
TSP (winter)	140–390	250 ± 100	6

Table 5 lists the average elemental composition of total aerosol mass measured on Whatman filter samples. It is striking, though perhaps not unexpected, that the mean elemental composition of aerosol mass is quite constant over the two sampling periods. Aside from this statement one distinct difference can be noticed, namely that the concentration of Cl in winter and, consequently, its relative contribution to the total mass of aerosol is about 2–5 times higher than in summer. This may be partly due to the losses of Cl from Whatman filters during handling, which will by discussed later.

As can be seen from tab. 5, eight elements (Mg, Al, Cl, K, Ca, Fe, Cu, and Zn) account for about $85^0/_0$ of the total aerosol mass measured. These elements can be considered (apart from Si and S not measured in the study) as major constituents of the trace element content of the atmospheric aerosol in the studied area.

3.2. Cl, Se AND I LOSSES FROM WHATMAN FILTERS DURING HANDLING

All Whatman filters collected during the sampling compaign were divided into two equal parts and only one half of each filter was used for INAA analyses. The

second part of several Whatman filters was reanalysed about three years after the collection of the samples to test the reproducibility of the INAA technique. The results of the 2nd and 1st analyses expressed as their ratios are listed in tab. 6. These ratios and the corresponding standard deviations were averaged over the total number of samples taken into consideration. For all but three elements the ratios are equal to unity within the error showing the precision of INAA for the elements measured. However, the concentrations of Cl, Se, and I obtained from the 2nd analysis are significantly lower (at a confidence level of 99.9% for Cl and 95% for Se and I) than those measured in the 1st analysis. This indicates that substantial losses of Cl, Se and I occur during the storage of Whatman filters. Considerable losses of Cl and to a smaller extent of Br stated in aerosol samples have already been reported before [17].

Table 5

Main components of the atmospheric aerosol in Katowice: average element-tal composition of total aerosol mass measured by INAA on Whatman filters

Element	Average percentage ± standard deviation	
	Summer	Winter
Na	3.5±0.8	6.5±2.4
Mg	5.2±3.3	5.6±2.8
Al	21.3±4.7	23.7±9.0
Cl	9.7±2.5	25.5±8.4
K	8.8±1.7	8.2±1.5
Ca	16.6±0.4	12.2±4.5
Ti	1.6±0.4	1.5±0.3
V	0.06±0.13	0.07±0.01
Cr	0.16±0.13	0.17±0.12
Mn	1.9±2.6	0.97±1.11
Fe	10.5±5.0	14.4±3.5
Cu	6.1±3.2	2.2±2.6
Zn	5.1±4.7	2.9±2.3

4. SUMMARY AND CONCLUSIONS

The atmospheric levels of 38 trace elements determined in the airborne particulates collected in Katowice ranged from 83 $\mu g/m^3$ for Al to 0.097 ng/m^3 for In. Of 38 trace elements investigated in the study eight elements, e.g., Mg, Al, Cl,

Table 6

Results of reanalysis of Whatman filter samples

Element	Average concentration ratio ± standard deviation	Numbers of samples
Na	1.15 ± 0.12	12
Mg	0.91 ± 0.24	4
Al	0.88 ± 0.13	13
Cl	0.38 ± 0.18	13
K	1.21 ± 0.20	11
Ca	0.89 ± 0.46	9
Sc	1.02 ± 0.16	10
Ti	1.16 ± 0.21	10
V	0.91 ± 0.12	13
Cr	1.04 ± 0.19	11
Mn	0.97 ± 0.09	14
Fe	0.98 ± 0.18	12
Co	0.98 ± 0.17	12
Cu	1.02 ± 0.25	12
Zn	0.95 ± 0.16	12
Ga	1.09 ± 0.28	7
As	1.08 ± 0.13	12
Se	0.60 ± 0.32	12
Br	1.12 ± 0.37	14
Ag	1.17 ± 0.29	7
Cd	0.97 ± 0.20	8
In	1.06 ± 0.25	12
Sb	1.09 ± 0.11	12
I	0.84 ± 0.13	12
Cs	0.93 ± 0.20	12
Ba	1.14 ± 0.31	8
La	1.12 ± 0.19	12
Ce	0.91 ± 0.12	5
Sm	1.26 ± 0.35	9
Eu	0.95 ± 0.16	6
W	1.02 ± 0.16	10
Th	1.14 ± 0.20	7

K, Ca, Fe, Cu, and Zn account for about 85% of the total aerosol mass measured. These elements (apart from Si and S not measured by INAA) may be considered as major constituents of the trace element content of the atmospheric aerosol in Katowice.

The total aerosol mass measured by INAA in Whatman filters accounts for about 25% of the total suspended particulates (TSP) collected on the filters. The remaining about 75% is composed mainly of carbon, oxygen and nitrogen, the main missing elements not measured by this method.

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POZIOM PIERWIASTKÓW ŚLADOWYCH W ATMOSFERZE W KATOWICACH

Zbadano 74 próbki aerozolu pobrane w Katowicach za pomocą sączków Whatmana nr 41 w okresie od 21 sierpnia 1979 r. do 7 stycznia 1980 r. Zmierzono stężenia 38 pierwiastków (Na, Mg, Al,

Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Mo, Ag, Cd, In, Sb, I, Cs, Ba, La, Ce, Sm, Eu, Lu, W, Au i Th) stosując instrumentalną analizę aktywacji neutronowej (INAA). Zgodnie z tymi pomiarami Mg, Al, Cl, K, Ca, Fe, Cu i Zn stanowią 85% pierwiastków śladowych w aerozolu atmosferycznym w Katowicach. Masa całkowita aerozolu zatrzymanego na sączku Whatmana stanowi jednak tylko około 25% wszystkich rozproszonych cząstek, gdyż takie pierwiastki jak węgiel, tlen, azot, siarka i krzem nie są mierzone metodą aktywacji neutronowej.

УРОВЕНЬ МОКРОЭЛЕМЕНТОВ В АТМОСФЕРЕ В Г. КАТОВИЦЕ

Были исследованы 74 пробы аэрозоля отобранных в г. Катовице с помощью фильтра Утомана № 41 в период от 21 августа 1979 г. до 7 января 1980 г. Были измерены концентрации 38 химических элементов (Na, Mg, Al, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Mo, Ag, Cd, In, Sb, I, Ba, La, Ce, Sm, Eu, Lu, W, Au и Th), применяя инструментальный анализ активации нейтронами (INAA). Согласно этим измерениям в Като-вицах Mg, Al, Cl, K, Ca, Fe, Cu и Zn составляют 85% микроэлементов в атмосферном аэрозоле. Полная масса аэрозоля, задержанного на фильтре Уотмана составляет, однако, только около 25% всех рассеянных частиц, так как такие элементы, как углерод, кислород, сера и кремний не измеряются методом активации нейтронами.