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Radiomonitoring of the Opole industrial district

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Abstract: The metallurgic industry is one of the main sources of environmental pollution. It emits dust, which contains manganese oxides, iron oxides, zinc and lead oxides that could be deposited in soil. The aim of this study was to determine the influence of the local metallurgical industry on soil contamination with metals released during processing using Pb-210 as a contamination tracer. Soil samples were collected from 19 sites located around the Opole industrial zone, in which, for example, welding and galvanizing companies operate. In the studies, various types of sampling sites were taken into account, such as the forests, meadows, fields and allotment gardens. The activity measurements of Bi-214, Pb-214, Cs-137 and Pb-210 were performed, using a gamma spectrometer. Applying the properties of the latter isotope, the local level of soil contamination was estimated. The results indicated that the metallurgical industry is not a source of environmental pollution in the immediate vicinity. The excessive lead concentrations ranged from 1 Bq/kg d.m. to 100 Bq/kg d.m. and were not strongly correlated with the distances from the presumed source of contamination.

Keywords: air pollution, metallurgical industry, Pb-210

INTRODUCTION

At the turn of the 1980s and 90s, in the Opole-Metalchem district, a great metallurgical industrial center was located. Its main aim was the production of armaments, creating jobs for several thousand people. Nowadays, in this area, many welding, galvanizing and tinning works take place.

Waste produced in the welding process, such as lead, copper, zinc or nickel can enter the human body through the food chain (Balkhyour et al. 2010, Dartey et al. 2010, Herranz et al. 2013, Murphy 2013). As such, the waste can cause a number of health changes, such as kidney disease, osteoporosis and cataracts (Campbell et al. 2004, Brown et al. 2008, Yang et al. 2010). Therefore, periodic analyses of water, soil and air in this region are necessary.

Lead, especially its isotope Pb-210, is very useful in the analysis of soil contamination. This is due to the fact that its activity in the environment consists of allogeneic (anthropogenic origin) and autogenous (natural origin) contents. The contamination of the environment is indicated by increased activity of allogenic lead (Bem et al. 1998).

In this study the influence of the local metallurgical industry on soil contamination, with metals released during processing, using Pb-210 as a contamination tracer was investigated.

MATERIALS AND METHODS

In the study, soil samples collected from 19 sites, located in the Opole-Metalchem district were analyzed. This work was an extension of research conducted in 2013 (Godyń et al. 2013). In Figure 1 locations of the sampling sites are shown. These sites were mostly farmland (1–4, 6, 7, 15–18), but samples were also collected in woodlands (10),

meadows (9, 11) and the public use areas (Fig. 1). All places were located a short distance from the metallurgical industry buildings. The exception is the 19th point, located in the center of the zone.

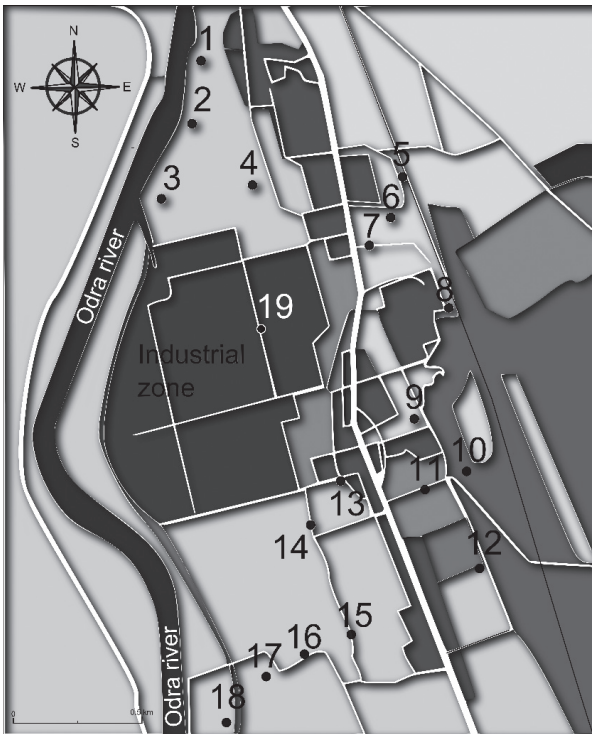

Fig. 1. *Location of sampling points*

The samples were collected from the surface layer to a depth of up to 15 cm and a deeper layer depth of 15–30 cm. At each point, subsamples were collected to get total sample mass of 1 kg. The material was dried at 105°C to constant mass, sieved through a 2 mm diameter mesh and subjected to gamma-spectrometry analysis.

Measurements of the activity concentrations of Pb-210, Bi-214, Pb-213 and Cs-137 were performed by means of a gamma spectrometer, with a high-resolution HPGe germanium detector (Canberra) (1.29 keV [FWHM] at 662 keV and 1.70 keV [FWHM] at 1332 keV; relative efficiency: 21.7%). Energy and efficiency calibration of the gamma spectrometer was performed with the standard MBSS 2 type solution (Czech Metrological Institute, Prague, CZ), which covers an energy range from 59.54 keV to 1836.06 keV. The geometry of the calibration source was the Marinelli beaker (447.7 ± 4.5 cm^3), with a density of 0.99 ± 0.01 g/cm^3, containing Am-241, Cd-109, Ce-139, Co-57, Co-60, Cs-137, Sn-113, Sr-85, Y-88 and Hg-203. The geometry of the sample container was Marinelli, 450 cm^3. The measuring process and analysis of spectra were both computer controlled by software GENIE 2000. MDA values for analyzed isotopes, in range of 43.5–79.0 for Pb-210, 0.38–5.47 for Cs-137, 1.26–8.78 for Pb-214 and 1.14–8.41 for Bi-214. Measurement uncertainty ranged from 0.17–3.84 for Cs-137, 8.4–181.0 for Pb-210, 0.17–27.8 for Bi-214 and 0.67–14.6 for Pb-214.

RESULTS AND DISCUSSION

In the studied soil layers, activity concentrations of selected gamma radionuclides were determined. Distribution of Cs-137 activity concentration showed the highest variability, with values ranged from a few hundred becquerels per kilogram of dry mass (Bq/kg d.m.), in forest samples, to several becquerels per kilogram of dry mass, in samples from farmland. Also, the maximum activity concentration of this radionuclide in the surface soil layer was significantly higher than in the deeper one. Similar behavior was observed for Pb-210, with mean activity concentration of 80 Bq/kg d.m.

For Bi-214 and Pb-214, the lowest difference in activity was observed, where the activity concentrations in the both layers were in the range from 6 Bq/kg d.m. to 30 Bq/kg d.m.

In Table 1 the basic statistical parameters, i.e. the maximum (Max), minimum (Min), median (Median), lower quartile (Q1) and the upper quartile (Q3), mean and standard deviation (SD) are shown.

In Figures 2 and 3 relationships between activity concentrations of the radionuclides determined in the upper and lower soil layers are shown. The straight line in the graphs show the same radionuclide activity concentrations in both soil layers. Cs-137 activity concentration in the Opole region comes mainly from air pollution, occurring after the explosion at the Chernobyl nuclear power plant in 1986 (Biernacka et al. 2002, Wołkowicz et al. 2002, Dołhańczuk-Śródka et al. 2007, Korobova et al. 2009). Analysis of the results showed, that the content of Cs-137 in the surface soil layers is much higher than the activity concentration in deeper layers, which confirms the atmospheric nature of its origin (Fig. 2A).

Table 1
Basic statistical values of the determined radionuclides

Layer	Radionuclide	Max	Min	Median	Q1	Q3	Mean	SD
		[Bq/kg d.m.]						
0–15	Cs-137	524	7	82	72	101	114	115
	Pb-210	104	<MDA	36	19	44	36	23
	Bi-214	35	7	11	10	27	17	10
	Pb-214	38	7	12	10	28	18	11
15–30	Cs-137	119	7	69	40	87	64	33
	Pb-210	61	<MDA	20	13	35	24	18
	Bi-214	38	7	14	10	29	18	11
	Pb-214	40	7	14	10	31	20	12

The Pb-210 radioisotope showed a higher activity concentration in the surface layers (Fig. 2B), but this effect was not as evident as in the case of Cs-137. This phenomenon is associated with two sources of the determined lead. The first of them is human activity, leading to atmospheric pollution which is later deposited on the surface. As a result, the surface soil layers are enriched with the Pb-210 isotope. The second source of Pb-210 is radioactive decay of its parent radionuclides, derived from Ra-226, through Bi-214 and Pb-214. These radionuclides are naturally present in the soil and constitute a geological source of lead Pb-210 in the environment.

As shown in Figure 3A and 3B, the activity concentrations of the above-mentioned Bi-214 and Pb-214, in contrast to Cs-137 and Pb-210, are distributed equally in the two layers of soil.

Further analysis was based on the determination of excess lead (Pb_{ex}) content of anthropogenic origin. Calculations are based on the two aforementioned components of Pb-210 in the environment. The allogenic component was calculated from formulas (1) and (2):

$$a_{\text{Ra-226}} = \frac{a_{\text{Pb-214}} + a_{\text{Bi-214}}}{2} \quad (1)$$

$$\text{Pb}_{\text{ex}} = a_{\text{Pb-210}} - a_{\text{Ra-226}} \quad (2)$$

where a – the activity concentration of analyzed radionuclide [Bq/kg d.m.].

Activity concentration of calculated excess lead was significantly higher in the surface soil layer (Fig. 4).

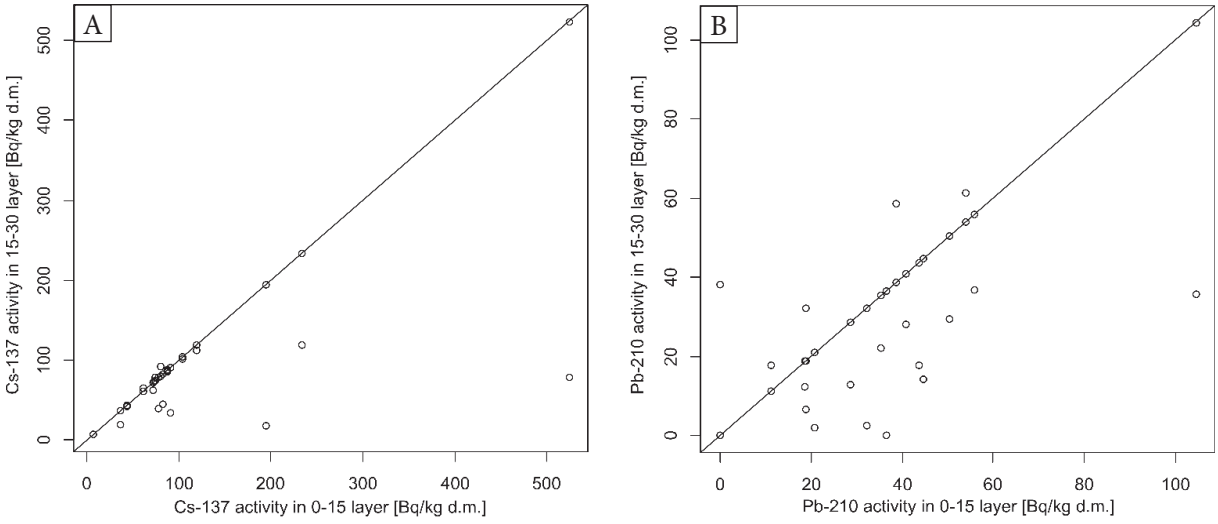

Fig. 2. *Relationship between the activity concentrations of Cs-137 (A) and Pb-210 (B) in upper and lower soil layers*

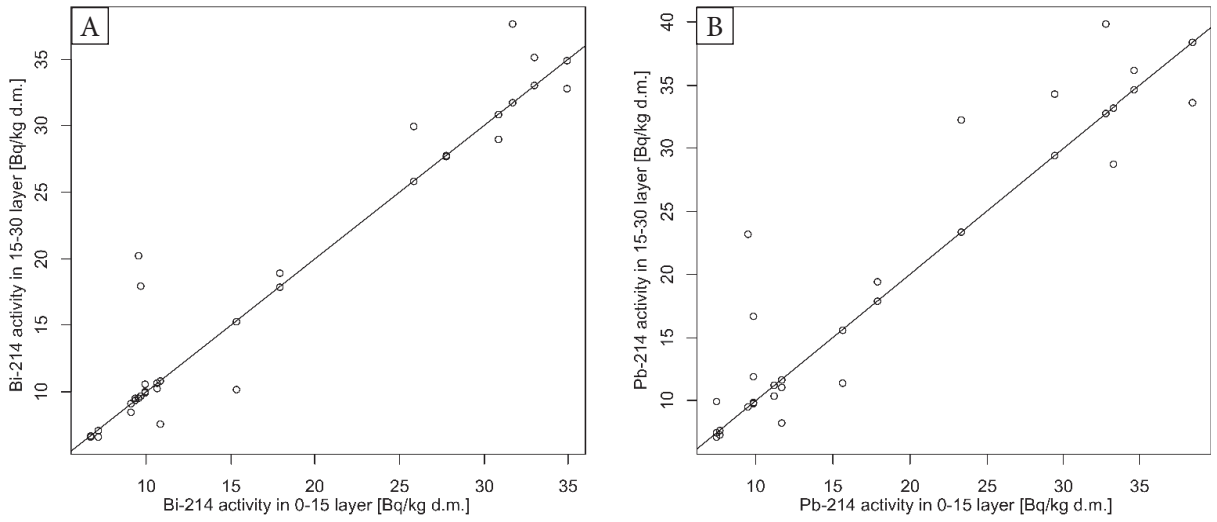

Fig. 3. *Relationship between the activity concentrations of Bi-214 (A) and Pb-214 (B) in upper and lower soil layers*

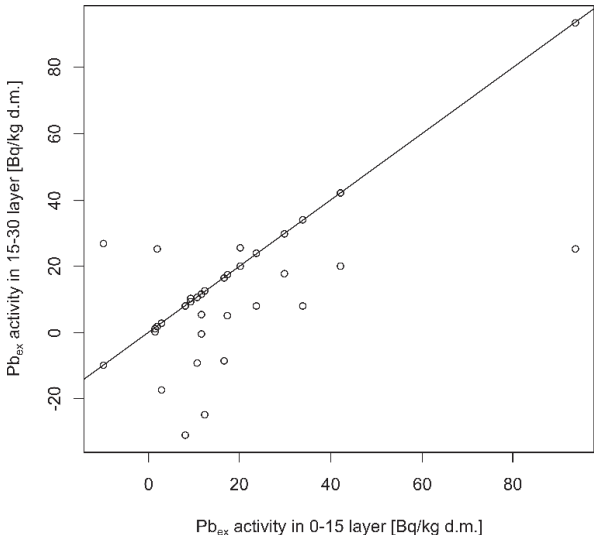

Fig. 4. *Relationship between the activity concentrations of Pbex in upper and lower soil layers*

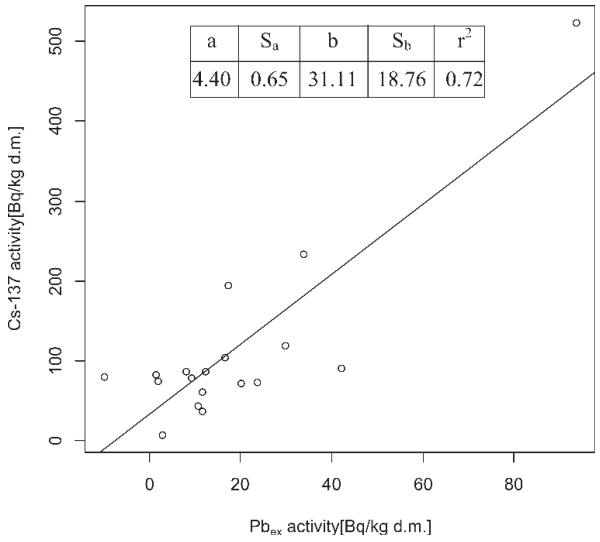

Fig. 5. *Relationship between the activity concentrations of Cs-137 and Pb_{ex} in sampling sites*

Only in the case of three sampling sites, was the activity concentration value higher in the deeper soil layer than that in the 0–15 cm layer. These disorders were probably caused by agrotechnical treatments, conducted before collecting the samples. The obtained results confirm the higher activity concentration of Pb-210 and the atmospheric origin of the Pb_{ex} component.

The activity concentration of excess lead in the surface soil layer was well correlated with the activity concentration of Cs-137. A high Pearson correlation coefficient (0.86) indicates the crucial influence of local weather conditions on deposition of pollutants from the air (Fig. 5).

Analysis of Pb-210 soil contamination in the metallurgical industry zone surrounding Opole was based on the relationship between the activity concentration of excess lead reduced by the activity concentration determined in the middle of the zone and the distance from the center of the zone. The obtained relationship is shown in Figure 6.

The obtained correlation coefficient indicates a lack of association between the distance from the zone of metallurgical industry and the activity concentration of excess of lead in the soil. It can be supposed that the metallurgical industry in Opole is not the source of local soil contamination with lead compounds.

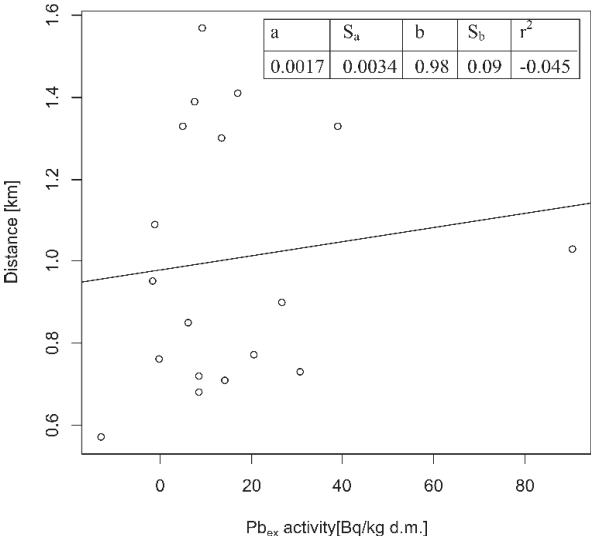

Fig. 6. *Relationship between the activity concentrations of Pb_{ex} and distance from center of industrial zone*

CONCLUSIONS

The methodology used in soil analysis enabled us to determine the two Pb-210 components in the environment. Therefore, Pb-210 can be a good tracer of deposition of atmospheric pollutants. High correlation of excess lead with Cs-137 indicates a link between soil contamination and local weather events. In locations with elevated Pb_{ex} activity concentration in the surface soil layer, an increase in the deposition of other pollutants of atmospheric origin can be expected.

Analysis of the results led us to conclude, that the local metallurgical industry is not a significant source of contamination containing compounds Pb-210.

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