

LIQUID SCINTILLATION COUNTING OF ^{210}Pb IN GRASS AND SOIL

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ABSTRACT. We measured concentrations of ^{210}Pb and ^{226}Ra in grass and soil from areas of increased natural radioactivity resulting from coal mine drainage. We developed a method of determining ^{210}Pb using a Quantulus 1220™ liquid scintillation (LS) spectrometer; this method allows direct measurement of ^{210}Pb in a large number of samples in a short period of time. Ra isotopes can be determined simultaneously. The method consists of decomposition of biological matter by combustion, followed by dissolution in a mixture of inorganic acids. PbSO_4 is then separated from $(\text{BaRa})\text{SO}_4$, the precipitates are mixed with a gelling liquid scintillator, and the activity is measured by LS counting. The detection limit of ^{210}Pb is 0.02 Bq per sample with 2 h counting time. This enables detection of 0.3 Bq of ^{210}Pb in 1 kg of fresh grass. Concentrations of ^{210}Pb varied from 0 Bq kg^{-1} to 5.8 Bq kg^{-1} , whereas ^{226}Ra varied from 0.2 Bq kg^{-1} to 1.0 Bq kg^{-1} .

INTRODUCTION

Coal mining in the Upper Silesian coal basin causes serious environmental pollution. One pollutant is saline water released from underground mines. Such water often contains radioactive isotopes, mainly ^{226}Ra and ^{228}Ra . We recognized two types of Ra-bearing waters (Lebecka *et al.* 1986):

- Type A, with barium ions, but with no sulfate ions. In this case, Ra always co-precipitates with Ba forming $\text{BaSO}_4 + \text{RaSO}_4$ (precipitation takes place when these waters mix with sulfate-containing waters, which are very common in nature).
- Type B, with no Ba ions, but with sulfate ions. In this case, precipitation of Ra does not occur, but only dilution in rivers and sorption (because there are no Ba ions for co-precipitation; water containing Ba ions is not very common in nature).

Ra-bearing waters released from coal mines cause contamination of river beds and neighboring areas by radioactive sediments or elevated radioactivity of river water. This contamination is not only caused by Ra, but also by its daughter products. One of the most radiotoxic daughters is ^{210}Pb , a decay product of ^{226}Ra . The half-life of ^{210}Pb is rather long ($t_{1/2} \approx 22.3$ yr). The annual limit of intake (ALI) for ^{210}Pb (2×10^4 Bq) is even lower than that for ^{226}Ra (7×10^4 Bq) (ICRP 1980). Thus, the measurement of ^{210}Pb in water, soil and plants is important for the evaluation of doses obtained by inhabitants of polluted areas.

Pollution caused by Ra-bearing water and deposits from coal mines is limited to areas near small rivers and streams where Ra-bearing water is released or was released in the past. Settling ponds and waste dumps are also contaminated. The contamination is irregularly distributed, with a total area of several square kilometers.

Contamination of plants and grass is caused mainly by radioactive deposits spread in the ground adjacent to streams and river beds during periodic dredging. In most cases, radioactive isotopes in plants cultivated in such areas may elevate doses obtained by inhabitants, due to consumption of contaminated meat originating from cattle or sheep grazing in affected pastures and meadows. We chose four different contaminated areas to investigate ^{210}Pb and ^{226}Ra concentrations in soils and grasses and describe our results.

^{210}Pb emits low-energy beta ($E_{\beta\text{max}} = 20$ keV (81%), 61 keV (19%)) and gamma rays ($E_\gamma = 46.5$ keV). In both cases, measurements are difficult because of poor counting efficiency, self-absorption in the source and absorption in the detector shield. To avoid these problems, we do not usually measure ^{210}Pb directly, but through its daughter product, ^{210}Po , which is an alpha emitter. Here we

used liquid scintillation counting (LSC) to measure ^{210}Pb directly (Lebecka & Chalupnik 1990). This method consists of chemical separation of ^{210}Pb with PbSO_4 carrier and measurements by a low-background LS spectrometer.

METHODS

After drying at 105°C , soil samples were ground and sieved. Then, 50 g of the homogeneous material were dissolved in a mixture of inorganic acids. We used the same technique to measure ^{210}Pb in this solution as we used for water samples. ^{210}Pb , ^{226}Ra , ^{228}Ra and ^{224}Ra were determined on the same sample (Chalupnik & Lebecka 1993) by chemical separation of Pb and Ra isotopes with carriers, PbSO_4 and BaSO_4 , followed by LSC. Figures 1 and 2 show the procedure.

Samples (mainly grass) were dried at room temperature, cut and ground. Then, 15 g of the homogeneous material were oxidized at 500°C (4 h), mixed with HNO_3 and heated at the same temperature. The residue was treated by HCl and H_2O_2 , then rinsed with hot water. A filtrate was used for measuring ^{210}Pb according to the technique described above.

Measurements were made in a Quantulus 1220TM LS spectrometer with an anticoincidence shield, where very low background can be achieved, and very low activities of ^{210}Pb and ^{226}Ra can be measured. The lower limit of detection (LLD) for ^{226}Ra is as low as 0.001 Bq per sample, whereas the LLD for ^{210}Pb is 0.02 Bq per sample (2 h counting time). This enables detection of 0.02 Bq kg^{-1} of ^{226}Ra and of 0.3 Bq kg^{-1} of ^{210}Pb in fresh grass. Figure 3 shows spectra of ^{210}Pb obtained for a grass sample and standard solution of ^{210}Pb . The activity of ^{210}Pb is measured directly by

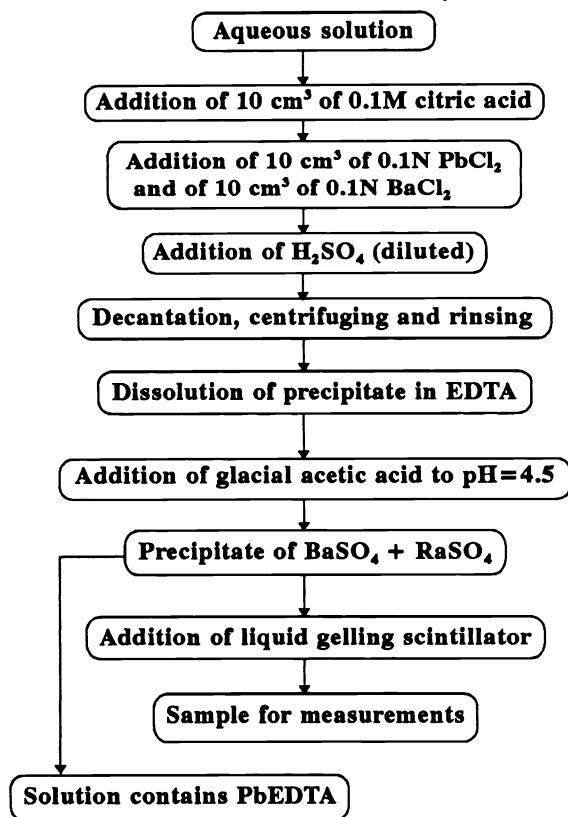


Fig. 1. Preliminary preparation of sample

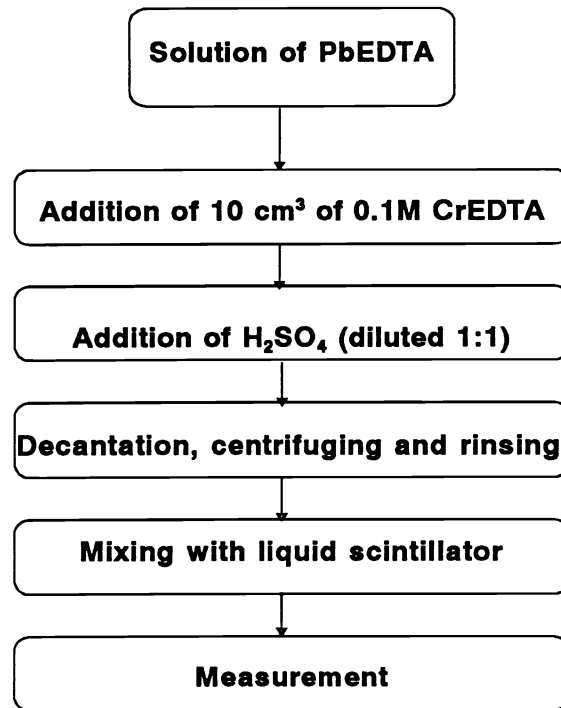


Fig. 2. Preparation of ^{210}Pb

counting low-energy β particles; thus, counting can be done immediately after sample preparation, versus waiting several days to build up the ^{210}Bi or ^{210}Po activity.

RESULTS

We investigated four sites near locations where Ra-bearing water from coal mines is released into the natural environment. Sites 1 and 2 contained type-A (with Ba ions) Ra-bearing brines. Sites

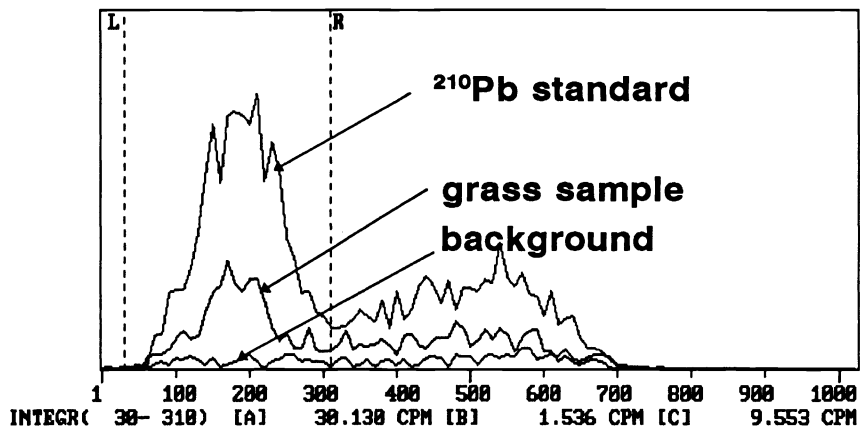


Fig. 3. Different spectra of ^{210}Pb

3 and 4 contained type-B (with sulfate ions) Ra-bearing brines. Site 1 was located on a bank of a small river, where, in the past, radioactive deposits, $\text{BaSO}_4 + \text{RaSO}_4$, precipitated from Ra-bearing water released from a mine. These deposits covered the river bed and were spread on the banks by periodic dredging. Site 2 was near a settling pond containing type-A Ra-bearing brines. Sites 3 and 4 were located near large settling ponds containing type-B Ra-bearing waters. The investigation sites represent ~10 ha of pasture land. Table 1 presents results of these measurements at all four sites.

TABLE 1. Results of Measurement of ^{226}Ra and ^{210}Pb in Soil and Grass

Sample no.	Soil (Bq kg^{-1})		Fresh grass (Bq kg^{-1})		Transfer coefficients*	
	^{226}Ra	^{210}Pb	^{226}Ra	^{210}Pb	^{226}Ra	^{210}Pb
<i>Site 1</i>						
1	107 ± 13	67 ± 7	0.80 ± 0.07	1.5 ± 0.4	.007	.023
2	1898 ± 190	1618 ± 162	0.63 ± 0.06	0.9 ± 0.4	.0003	.0006
3	71 ± 9	56 ± 6	0.55 ± 0.05	0.4 ± 0.3	.008	.008
4	93 ± 10	56 ± 6	0.38 ± 0.05	0.6 ± 0.3	.004	.011
5	41 ± 8	37 ± 4	0.45 ± 0.05	<0.3	.011	<.006
6	22 ± 6	25 ± 3	0.26 ± 0.04	0.6 ± 0.3	.012	.025
<i>Site 2</i>						
1	25 ± 6	14 ± 2	0.53 ± 0.06	3.8 ± 0.6	.021	.279
2	18 ± 5	18 ± 2	0.49 ± 0.05	2.0 ± 0.4	.027	.111
<i>Site 3</i>						
1	42 ± 8	43 ± 5	0.54 ± 0.06	1.0 ± 0.4	.013	.024
2	32 ± 7	10 ± 1	0.66 ± 0.07	2.0 ± 0.5	.021	.194
3	33 ± 7	15 ± 2	1.03 ± 0.10	4.5 ± 0.6	.031	.297
4	6 ± 4	18 ± 2	0.28 ± 0.06	1.7 ± 0.4	.047	.098
5	62 ± 9	64 ± 6	0.76 ± 0.08	5.8 ± 0.7	.012	.090
<i>Site 4</i>						
1	21 ± 6	25 ± 3	0.20 ± 0.04	0.9 ± 0.4	.010	.036
2	13 ± 5	24 ± 3	0.26 ± 0.04	3.0 ± 0.6	.020	.125
3	7 ± 4	5 ± 1	0.27 ± 0.04	1.1 ± 0.4	.039	.224

*The transfer coefficients for ^{226}Ra and ^{210}Pb are defined as follows: k_g = the concentration of isotope in fresh grass divided by the concentration of isotope in soil.

We observed large differences in transfer coefficients, which probably are due to the chemical forms of Ra and Pb present in the soil. In areas contaminated by almost-insoluble Ra sulfate, the transfer coefficient was much lower than in areas where Ra is soluble in water. Transfer coefficients of ^{210}Pb are usually higher than those of ^{226}Ra . A possible explanation is that transfer of Ra probably occurs mainly through the root system, whereas Pb transfer also occurs through air by emanation of radon from soil and fallout or rainout of Rn daughters from air onto grass.

CONCLUSIONS

We developed a method of sample preparation and measurement of ^{210}Pb concentration in grass and soil. The method consists of mineralization of biological matter by dissolution in a mixture of inorganic acids followed by co-precipitation of sulfates with non-radioactive carrier. Samples were

measured in a low-background Quantulus 1220™ LS spectrometer, which enables determination of ^{210}Pb and ^{226}Ra simultaneously in a large number of samples.

The detection limit of ^{210}Pb is 0.02 Bq per sample (2 h counting time), which enables detection of 0.3 Bq kg⁻¹ of ^{210}Pb concentration in fresh grass. Preliminary results of ^{210}Pb measurements in grass and soil samples collected from areas contaminated by Ra-bearing waters show large differences of transfer coefficients; these may be caused by different chemical forms of Ra in soil and perhaps by differences in the emanation of ^{222}Rn from soil.

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