RADIONUCLIDE CONTENT IN THE UPPER VISTULA RIVER SEDIMENTS IN A COAL MINING REGION IN POLAND (EAST-CENTRAL EUROPE)

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Abstract. Hard coal mining activity is one of the Technologically Enhanced Natural Radiation sources introduced more than a hundred years ago in the southern part of Poland. This study of radionuclides (K-40, Ra-226, Ra-228) in river sediments showed the principal factors determining the specific activity of radionuclides are distance from mines and sorption by clay particles in river sediments. The K-40 specific activity varies between 185 and 595 Bq/kg. The highest Ra-228 and Ra-226 specific activities, 280–610 and 205–415 Bq/kg, respectively, were detected in sediments of the Vistula in the vicinity of Silesia region.

Key words: hard coal, K-40, Poland, Ra-226, Ra-228, river sediments,

1. Introduction

The Vistula River called *a queen of Polish rivers* is, unfortunately, the most polluted river in Poland. Industrial and municipal sewage from the Upper Vistula Basin, particularly from Silesia industrial region, is the most dangerous threat to the quality of river waters. Bottom sediments of the Vistula River are polluted too. There has not been enough attention paid to the contamination of the Vistula River sediments by radionuclides in previous works.

Technologically Enhanced Natural Radiation (TENR) was introduced in 1975 to describe modification of the natural environment and the possible inereased human radiation exposure associated with it (Gessel and Pritchard, 1975). Intensive coal mining activity introduced more than a hundred years ago in the Silesia region is one of the TENR sources there. The approximate mean specific activity in lithological layers containing hard coals in Poland equals 17 Bq/kg Ra-226, and 11 Bq/kg Ra-228, and 59 Bq/kg K-40 (Table I). A major source of TENR in the Vistula River is from mining activity in Silesia, in which coal with relatively high radioactivity content is extracted from its naturel location. There are 66 underground coal mines extracting approximately 150×10^6 tons of coal per year. The depth of mine works varies from 350 to 1050 m. Some differences in radionuclide concentrations in coal samples in Silesia are observed: Ra-226, Ra-228 and K-40 specific activities ranged from 0 to 121 Bq/kg, from 0 to 105 Bq/kg and from 0 to 758 Bq/kg, respectively (Wysocka and Skowronek, 1991). Specific activities of Ra-226 in deep circulation radium waters in Silesian mines varies from 0.7 to 28 Bq/l (Lebecka, Tomza and

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Table I

Specific activity of natural radionuclides in hard coal and the Earth's crust in Bq/kg (after Wysocka and Skowronek, 1991)

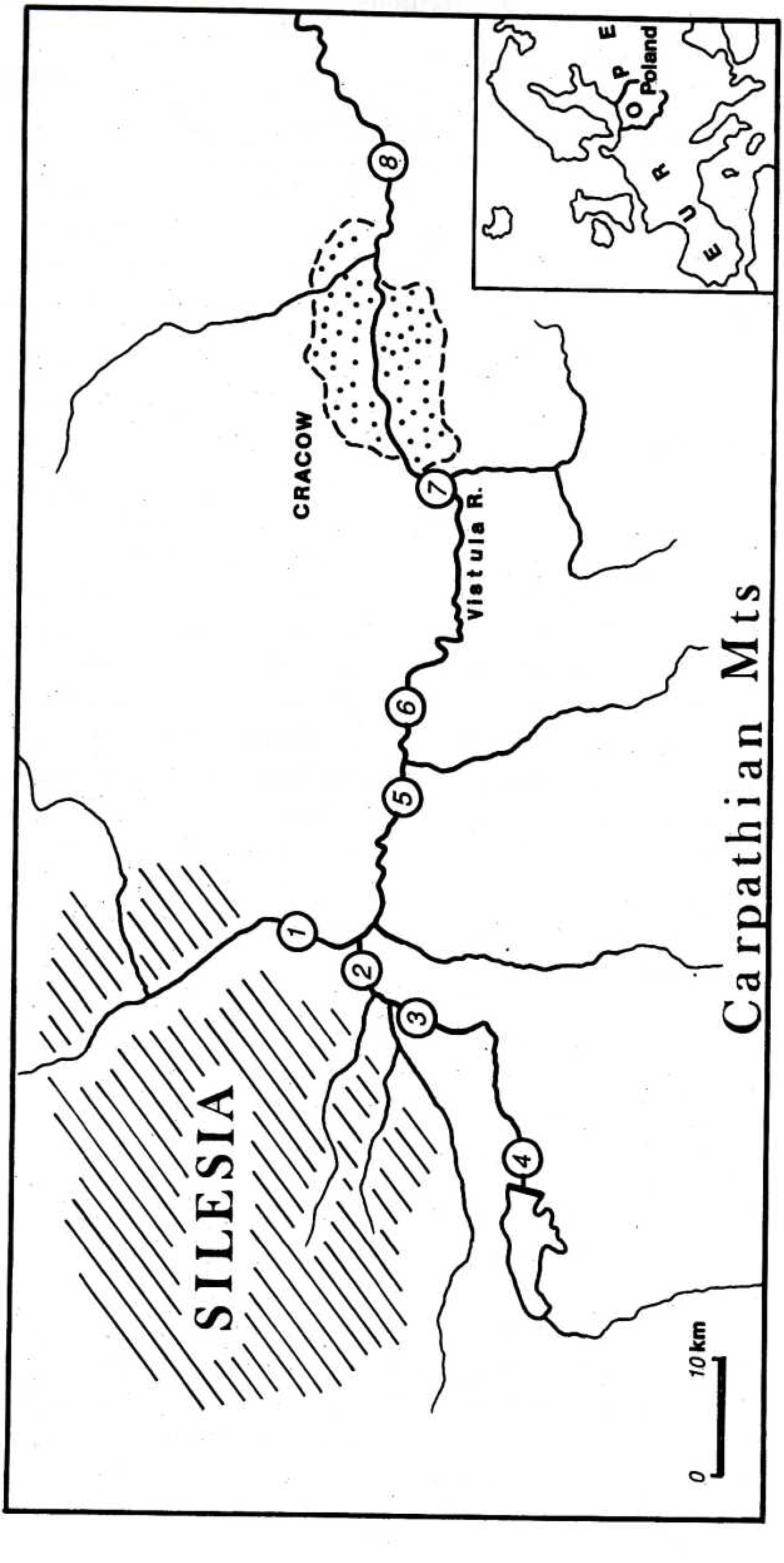
Area	K-40	Ra-228	Ra-226
USA	52	21	18
Rep. of S. Africa	110	20	30
Former USSR	120	22	28
Great Britain	120	17	17
Poland	59	11	17
World average	50	20	20
Earth's crust	370	25	25

Pluta, 1986). The precipitates containing, $BaSO_4$, $SrSO_4$ and $RaSO_4$ with very high radioactivity occur in channels and streams with water pumped from Silesian mines. These substances can be transported to the Vistula. The Ra-226 specific activities in these solids vary from 110 to even 133 200 Bq/kg and Ra-228 specific activity varies from 95 to 42 000 Bq/kg (Lebecka, Tomza and Pluta, 1986). It has been estimated that mine waters contribute about 420 MBq of Ra-226 and 107 MBq of Ra-228 to the Vistula River every day (Wiktorowski, 1989).

Assessing the impact of mining activity on radionuclide contents in the Vistula River sediments is the **aim** of this paper. Measurements of Ra-226, Ra-228 and K-40 radionuclide concentrations in the Vistula River and its tributary sediments have been done.

2. Area of Investigation

The upper part of the Vistula flows generally from the west to the east and passes a few different physiographic regions: the Carpathian Mts. Chain and the Silesian and Cracow Uplands (Figure 1). Carpathian Mts. are built of sandstones, slates, crystallic rocks, and Uplands are built of limestones, dolomites, and sandstones. The slope of the river bed is low and fine alluvial material can be transported. Mean annual discharge of the Vistula in Nowy Bierun and in the vicinity of Cracow (Tyniec) equals 22.7 and 96.6 cubic meters per sec, respectively. There is an artificial reservoir on the river course before the Vistula passes the Silesian region, and then a few Carpathian rivers with relatively good quality of water inflow to the Vistula. Also the Przemsza River from Silesia industrial and mining region and other streams with very polluted water and considerable amount of coal mud flow in to the Vistula.



EUROPE
Poland
CRACOW
Vistula R.
Carpathian Mts
SILESIA
1
2
3
4
5
6
7
8
0
10 km

Figure 1.

3. Methods

Sediment samples were collected in December 1993. Samples were taken from the river bottom at 7 sites located in the Vistula River channel and 1 site in the Przemsza River channel (Figure 1). Sample sites were situated 2–3 m off-shore, at not deeper than 1.5 m of water. Each sample was placed in a clean plastic bag and over-dried at 105 °C for 48 hours, followed by disaggregation and then a small amount of each sample was analyzed by laser Fritsch Particle Sizer to quantify the clay fraction ($\leq$0.020 mm). The measurements were performed in the laboratory of the Department of Geomorphology and Hydrology of Mountains and Uplands, Polish Academy of Sciences, Cracow.

Radium isotopes: Ra-228 ($\rightarrow$Ac-228), and Ra-226 as well as K-40 specific activity measurements were done using a gamma-ray spectrometer (manufactured by Silena Ltd.) with Ge(Li) detector. The typical measurement time was about 80 000 sec. The energies of photo-peaks of interest were: 186 keV (Ra-226), 911 keV (Ac-228, the isotope in equilibrum with Ra-228), and 1461 keV (K-40). The U-235 contributes to the 186 keV Ra-226 line in our standard on the level of 7.3%. Although the contribution of U-235 to the 186 keV line in samples spectra remains unknown, the upper limit of it can be obtained in two ways: the direct estimation from other its lines 144, 163, 205 keV and the estimation based on the Pa-234 1001 keV line (this radionuclide is in equilibrium with U-238, and there are no reasons for other than natural uranium isotopic ratio). Both methods give the upper limit for the systematic error of the determination of radium specific activity lower than 25%. The detector was shielded with low-background lead. Dried and homogenized samples were put into so-called Marinelli beakers (Cywicka-Jakiel *et al.*, 1988). Measurements were done by comparison with standards of the same geometry, which had known concentrations of all analysed radionuclides. The standards were prepared from uranium and thorium ores at the Institute of Nuclear Physics, Cracow. Radionuclide concentration A in a sample was calculated from simple formula

$$A = A_w \frac{N - n_o}{m(n - n_o)} \qquad (1)$$

where

A_w	=	activity of a given radionuclide in standard [Bq];
N, n_o, n	=	the numbers of the counts in the unit of time for given peak for sample, background or standard, respectively;
m	=	sample mass [kg].

Spectra analyses were performed using P.I.M.P. computer code (Mietelski, 1989).

Table II

Radionuclide specific activity and clay content in river sediments

No. of sampling site	^{40}K	^{228}Ra	^{226}Ra	Clay
	Bq/kg			(%)
1	230±25	45±5	45±15	11.0
2	360±35	610±25	415±70	31.0
3	310±35	280±20	205±50	50.5
4	480±45	40±5	70±20	59.0
5	185±20	130±5	80±20	8.5
6	270±15	75±5	55±15	19.0
7	440±40	40±5	65±20	48.5
8	595±40	60±5	55±15	84.0

Table III

Correlation coefficient (0.0005 significance level)

	K-40	Ra-228	Ra-226	Clay
K-40	1.000			
Ra-228	–0.125	1.000		
Ra-226	–0.047	0.992	1.000	
Clay	0.942	–0.120	–0.475	1.000

4. Results

Results of measurements are shown in Table II. Generally, there are not high radionuclide concentrations in the Vistula nor in the Przemsza rivers sediments. Clay prevails in sediments (48.5–84.0%) in the upper and lower parts of investigated section of the Vistula River bed.

The K-40 specific activity is not high and varies between 185 and 595 Bq/kg. It can be observed, that lower values occur in the middle part of the investigated river bed section and in the Przemsza River bed. The higher concentrations take place in the Vistula River bed in the vicinities of Silesia and Cracow. There is a strong correlation ratio between K-40 concentration and clay content in river sediments (Table III). It amounts 0.942 with significance level 0.0005.

The highest Ra-228 and Ra-226 specific activities (280–610 and 205–415 Bq/kg, respectively) were detected in sediments of the Vistula in the vicinity of Silesia region. Much lower values (45–130 and 45–80 Bq/kg, respectively) are observed in the rest of the area.

5. Conclusions

The K-40 activity concentration appeared to be more related to clay contribution in sediments than location. Clay particles may carry a predominantly negative surface charge making them attractive to cations, which sorb onto the surface of the clay particles. Radionuclides may also be held by the clay particles (Kathreen, 1984).

In general, the Ra-228 and Ra-226 specific activities in the Vistula River sediments are higher than mean contents of the Earth's crust. The water and suspended sediment input from coal mines appear to contribute to the elevated amounts of these radionuclides in sediments of the Vistula River adjoining to the coal mine region.

This study of radionuclides in river sediments showed the principal factors determining radionuclide specific activity are distance from mines and sorption by clay particles in river sediments.

Further investigations should be concentrated on relationship of sediment contamination to particle size and radionuclides concentration in aquatic organisms.

Also migration aspects of these radionuclides under natural environmental conditions via aquatic transport should be analyzed.

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References

Cywicka-Jakiel, T., Grychowski, P., Hajdas, I., Jasińska, M., Kołakowski, L., Łoskiewicz, J., Mazgaj, Z., Mikulski, J. and Ochab, E.: 1988, 'Radioactivity Measurements in Kraków Surroundings in the Aftermath of Chernobyl Reactor Accident', Raport IFJ No 1407/AP, Kraków.

Gessel, T. F. and Pritchard, H. M.: 1975, 'The Technologically Enhanced Natural Radiation Environment', *Health Physics* **28**, 361.

Kathreen, R. L.: 1984, *Radioactivity in the Environment. Sources, Distribution and Surveillance*, London-Paris-New York, p. 397.

Lebecka, J., Tomza, I. and Pluta, I.: 1986, 'Naturalne radionuklidy w wodach zrzutowych kopalń węgla kamiennego'. *Post. Fiz. Med.* **21**(3), 255–261 (Polish).

Mietelski, J. W.: 1989, 'P.I.M.P. – program analiz widm promieniowania gamma z detektora germanowego na komputerze klasy PC', Raport IFJ No. 1435/s, Kraków (Polish).

Wiktorowski, W.: 1989, 'A rad wciąż płynie', *Przegląd Techniczny* **24**, 7–8 (Polish).

Wysocka, M. and Skowronek, J.: 1991, 'Promieniotwórczość naturalna węgli w obszarze Rybnicko-Jastrzębskim'. *Publ. Inst. Geophys. Pol. Acad. Sc.* **M-15**(235), 341–346 (Polish).