Vol. 15 1989 No. 1–2

J. KWAPULIŃSKI*, B. NOWAK*

OCCURRENCE OF RADON DECAY PRODUCTS IN AIR WITHIN RECREATIONAL AREAS

The research on the occurrence of radon decay products was carried out in the selected industrial and recreational areas during the period of 1976–1985. The characteristics of ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi occurrence was described by the changes of concentrations of these isotopes as a function of time and height. The modified TSIVOGLOU method [8] was adapted for defining the concentrations of radon decay products. In order to eliminate radon stream fluctuation from the soil, the measurements were performed on the dam reservoir Wapienica and on the Seksty lake. The research was made in summer time to exclude additional sources of contaminations from coal combustion.

The investigations of radon decay product concentrations in day-and-night cycle showed only slight variability of concentrations of examined radioisotopes. The frequency of radon decay products occurrence fulfilled normal distribution. Self-purification of air in recreational areas is 50–80 times faster when compared with that in the areas influenced by coal power stations. The washout of these radioisotopes is 10 times lower.

1. INTRODUCTION

In recent years, some natural and artificial radioisotopes attracted more and more attention in determining the general balance of environmental impacts. Therefore the information about the occurrence and migration of these radioisotopes constitutes a very important range of problems considered in the papers which pretend to be called "complex".

The source of ionizing radiation in the atmosphere are natural radioactive elements contained in the respective elements of environment as a result of both radioactive fall-out and economic activity. Coal burnt in power stations contains uranium, radium-226 and thorium, as well as decay products (kBq/t) of these elements. The radioactivity of Polish hard coal ranges within 1.48-34.8 kBq/t [3], whereas the ²²⁶Ra content in coal from other countries varies from 37 Bq/t to 48.1 kBq/t.

^{*} Medical Academy, 41-200 Sosnowiec, ul. Jagiellońska 4, Poland.

Radon is a rare gas formed during decay of ²²⁶Ra. The stream of radon is proportional to the contents of uranium and radium in various rocks and soils. According to EVANS [2], the average value of radon string from the typical soil to atmosphere is equal to 4.43 kBq/day·m², and the characteristic values of radon concentration in air are 3.7–37 mBq/dm³. For agricultural areas in the USA the radon string equals 222–555 mBq/km²·s and for Warsaw, according to PENSKO [7], 703–4070 mBq/km²·s.

2. MEASURING METHODS

The air samples were collected on Whatman 41 filters by means of TFIA-2 Staplex dust-meter with the filtering surface of 10 cm diameter. The samples were collected at the speed of air suction of 0.84 m³/min during 5 min. The numbers of alpha impulses in 5, 15, 30 min periods were measured with a SSA-1 probe immediately after sampling.

The total number of countings for measuring times (t = 0.5, 0.15, 0.30 min) was applied for calculating the concentrations of particular radioisotopes. The allowance for the period between sampling and measuring (30 s) was also made and taken into consideration. Concentrations of radon decay products were calculated according to the Tsivoglou method [8].

3. DISCUSSION OF RESULTS

The research on the occurrence of radon decay products in air was held in 9 stops in the recreational areas (fig. 1, points from 1 to 9). The concentrations obtained were compared with those of radon decay products in industrial areas (fig. 1, points from 10 to 14). The measurements were made in the period of 1976–1985. In the first stage of research, the dust-fall was assumed as a criterion for choice of a given measuring point.

The mean contents of ²¹⁴Pb, ²¹⁸Po and ²¹⁴Bi in the layer of air close to the ground in selected places are presented in the fig. 1. It can be issued that contents of the radioisotopes in recreational areas are lower when compared with those in the industrial ones.

Relative contents of lead, polonium, and bismuth in the air on the areas influenced by the industry were defined as well. For the purpose of these considerations the small town of Szczyrk, situated 70 km southwards of industrial centre, was assumed as a reference system. Concentrations of particular radioisotopes in this place were as follows:

 $^{218}Po - 17.76 \pm 10.3 \text{ mBq/dm}^3,$ $^{214}Pb - 2.59 \pm 1.7 \text{ mBq/dm}^3,$ $^{214}Bi - 3.33 \pm 1.6 \text{ mBq/dm}^3.$

The relative values of radon decay product contents are presented in the tab. 1. It is very characteristic that the values of the quotients discussed are considerably higher in the areas with power stations and thermal-electric power stations in comparison with the values obtained in the recreational areas, where the relative part of radon decay products in the layer of air close to the ground is 10–100 times smaller.

Erosion capability and erosion speed also affect the variability of ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi concentrations. In most cases specific radioactivity of air decreases by about 50% after atmospheric precipitations [5].

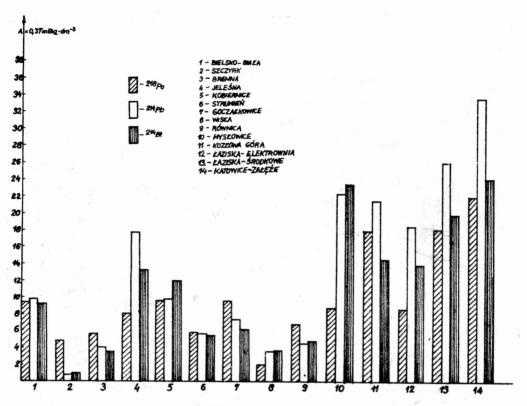


Fig. 1. The mean concentrations of ²¹⁴Pb, ²¹⁸Po, ²¹⁴Bi in the air layer close to the ground in selected places in Poland

Table
Comparison of the relative concentrations of radon decay products in air
occurring within recreational (the town of Szczyrk) and industrial areas

Place	Dustfall t/km ² ·month	²¹⁸ Po	²¹⁴ Pb	²¹⁴ Bi
Recreational areas	1			
Strumień	12.1	1.2	8.2	6.6
Goczałkowice	8.8	1.8	10.0	6.6
Równica	10.3	1.2	5.7	4.4
Wisła-Czarne	8.4	0.5	5.2	6.5
Industrial areas				
Katowice-Załęże	29.8	10.4	91.8	66.1
Łaziska	32.4	3.5	35.7	22.1
Chorzów	34.8	7.8	79.0	52.4

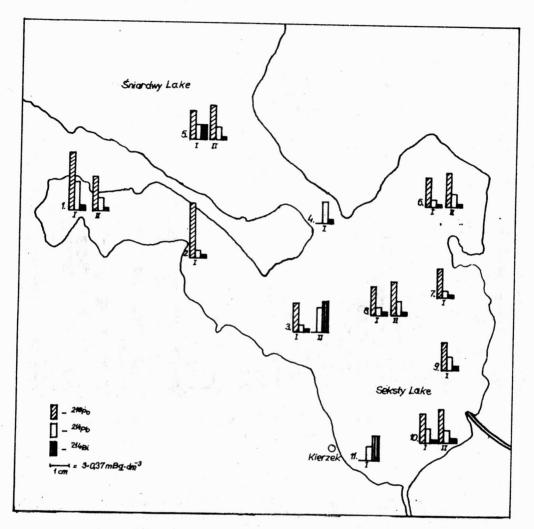


Fig. 2. Concentrations of radon decay products in various spots of the Seksty lake

The degree of air self-purification is determined among others by wash-out ability and wash-out speed. Wash-out ability is 10 times higher and wash-out speed 50-80 times higher in localities in recreational area than those in the industrial one [6].

Radon content in the air differs depending on the time and results from contents of uranic minerals in different parts of litosphere. Temporal changes are the consequence of changes in diffusion rate which depends on meteorological conditions such as atmospheric precipitations, a cover of snow or ice and changes of atmospheric pressure. In order to compare the results concerning the soils, the measurements were also made on the dam reservoir Wapienica and the Seksty lake.

2

			Table
440		214 _{Ph} 214 _{Ri}	
Concentrations of 218Po	, ²¹⁴ Pb	$^{214}_{Bi}$ and quotients of $\frac{^{214}_{Pb}}{^{218}_{Po}}$ and $\frac{^{214}_{Bi}}{^{214}_{Pb}}$	
		Po sirph	

Place	²¹⁸ Po mBq/dm ³	²¹⁴ Pb mBq/dm ³	²¹⁴ Bi mBq/dm ³	²¹⁴ Pb ²¹⁸ Po	²¹⁴ Bi ²¹⁴ Pb
Seksty lake	2.96 ± 0.6	0.74 ± 0.1	0.37 ± 0.1	0.25	0.5
Pisz forest	3.33 ± 0.2	1.48 ± 0.3	0.37 ± 0.2	0.44	0.25
Forest's cottage (Kierzek)	3.33 ± 0.3	1.48 ± 0.3	0.74 ± 0.3	0.44	0.5

The research was carried out in summer to eliminate additional sources of contaminations coming from the coal combustion.

The concentration of radon decay products in air was presented as the function of height and time. The results of measurements in various spots of the lake are presented in fig. 2. The average contents of radioisotopes examined were as follows: $^{218}Po - 2.96 \pm 1.7 \text{ mBq/m}^3$, $^{214}Pb - 0.74 \pm 0.5 \text{ mBq/m}^3$, $^{214}Bi - 0.37 \pm 0.2 \text{ mBq/m}^3$, $\pm SD(1.7, 0.5, 0.2 \text{ mBq/m}^3)$. The comparison of results obtained over the water surface of the lake, on the lake bank, and in the little town 12 km away is shown in tab. 2.

The contents of radon decay products in day and night cycle, as well as function of height, change slightly (figs. 3 and 4). The different way of changes was observed in the areas influenced by power industry [6].

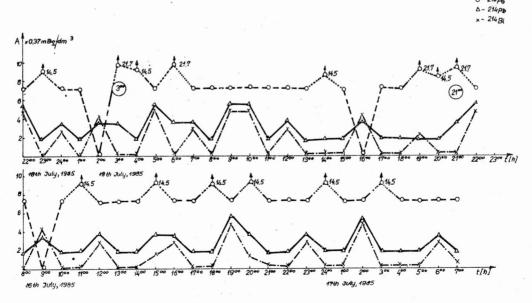


Fig. 3. Typical daily changes of radon decay products

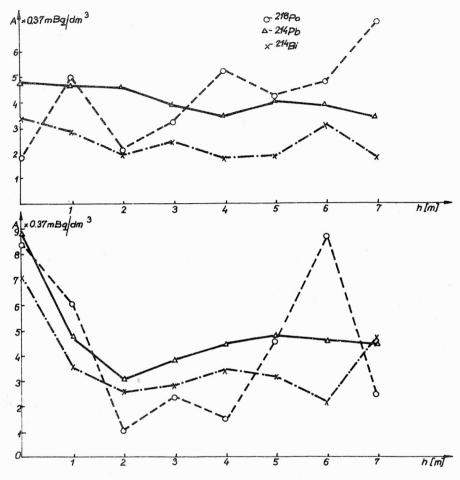
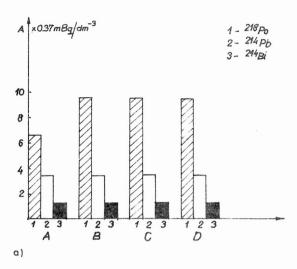


Fig. 4. Concentrations of radon decay products at different distances from the ground

In the Virgin Forest the measurements were made within the piny forest, mixed leafy forest, and birchen-piny one (fig. 5). It may be concluded that radon decay products are in the state of radioactive equilibrium.

When the lack of industry in this region and an essential correlation between concentrations of radioisotopes investigated, i.e., ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi, are considered, these concentrations can be regarded as conditioned by geochemical composition of soils only. Statistically, the values of concentrations of these radioisotopes accomplish the normal distribution, whereas in the industrial region the observed frequency distribution has log-normal character [6]. The stated normal distribution of radion decay products shows the absence of environmental factors.



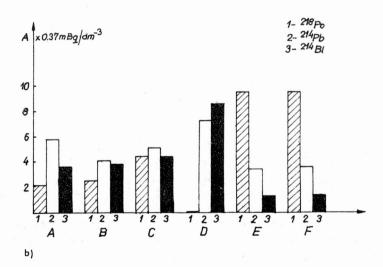


Fig. 5. a) Concentrations of radon decay products in the different wood complexes A - rural area, B - birchen-piny forest, C - mixed leafy forest, D - piny forest

b) Concentrations of radon decay products in the different points of the town Pisz (near the Seksty lake)

A, B, C, D, E, F - sampling points

The other measuring point was the dam reservoir near the town of Wapienica. The surface of this one was 14 ha. Physiography of the reservoir environment protects it effectively against inflow of contaminated air from all directions. The mountain slopes directly adjacent to the

reservoir make it resistant to the wind. The wind and raising of radon from the background are also restricted by small surface of the reservoir due to its relatively considerable depth, i.e., 21 m.

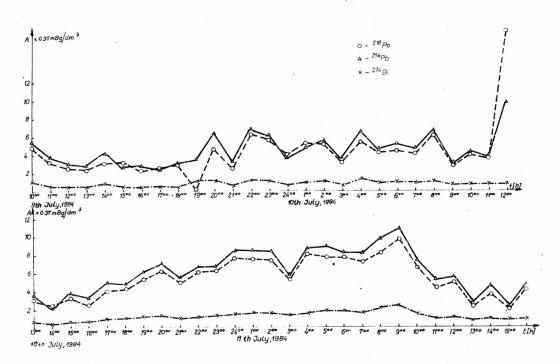


Fig. 6. Daily changes of ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi on the surface of the Wapienica lake

The results discussed above showed slight variability of ²¹⁸Po, ²¹⁴Pb, and ²¹⁴Bi concentrations in air samples collected in 1 m height. This remark concerns also the measurements made in day-and-night cycle. Similarly, as it was observed in the case of the lake described above, only small oscillations of radon decay product concentrations were observed in respective times of day-and-night cycles. Ranges of variability of the examined radioisotope contents are presented in the tab. 3. The mean contents of radon decay products were as follows:

```
^{218}Po -1.73 \pm 0.6 \text{ mBq/dm}^3, ^{214}Pb -1.90 \pm 0.7 \text{ mBq/dm}^3, ^{214}Bi -0.37 \pm 0.1 \text{ mBq/dm}^3.
```

The small variability of the concentrations of radon decay products in air as well as the essential correlation of the functions $^{214}\text{Pb} = f(^{218}\text{Po})$ and $^{214}\text{Bi} = f(^{214}\text{Pb})$ for two independent series of measurements (n = 70 and n = 58) justify using the results of average concentrations as the reference level for making up air estimations in different parts of Katowice district.

Table 3
Concentrations of ²¹⁸Pe, ²¹⁴Pb, ²¹⁴Bi over the surface
of the Wapienica lake water

Range	²¹⁸ Po mBq/dm ³	²¹⁴ Pb mBq/dm ³	²¹⁴ Bi mBq/dm ³
Night maximum	3.03	3.58	0.72
Day maximum	6.55	4.29	0.82
Night minimum	1.15	1.33	0.25
Day minimum	0.07	0.56	0.15
Mean value for $n = 127$	1.73 ± 0.6	1.90 ± 0.7	0.37 ± 0.1

It should be stressed that, similarly to those of Seksty lake, the values of free term b are extremely low, close to zero, which means that radon decay products are in equilibrium. This result gives evidence of the lack of external sources of charging these areas with 222 Rn. Subsequent series of measurements (n = 99) confirmed the above conclusions.

Table 4

Statistical characteristics of radon decay products' occurrence in air on the Wapienica lake, July, 1984

Dependence	Correlation coefficient	Determination coefficient	Test F $\alpha = 0.05$	Regression equation
214 Pb = $f(^{218}$ Po) $n^* = 58$	+ 0.977	0.047	1001.6	y = 1.19x - 0.001
$n = 58$ $2^{14}\text{Bi} = f(^{218}\text{Pb})$ $n = 58$	+ 0.987	0.974	2059.9	y = 0.205x
214 Pb = $f(^{218}$ Po)	+ 0.906	0.822	313.3	y = 0.845 x + 0.013
$n = 70$ $^{214}\text{Bi} = f(^{214}\text{Pb})$ $n = 70$	+ 0.952	0.906	657.6	y = 0.192x

 n^* – number of measurements.

In order to illustrate the differences in the concentrations of radon decay products in air, the examplary results of the research carried out in the Katowice district area are given. The values of mean concentrations of particular radioisotopes and their ranges measured at constant meteorological conditions in the Chorzów area are as follows:

Spring: n = 99,

average: $^{218}P_0 = 19.98 \text{ mBq/dm}^3$,

range: 17.02-24.05, average: $^{214}\text{Pb} = 28.27 \text{ mBq/dm}^3$,

range: 22.94-30.71, average: 214 Bi = 16.65 mBq/dm^3 , range: 14.06-19.61.

Summer: n = 75.

average: $^{218}Po = 17.76 \text{ mBq/dm}^3$,

range: 6.22-21.09,

average: $^{214}Po = 17.39 \text{ mBq/dm}^3$,

range; 6.22-19.98,

average: $^{214}Bi = 9.62 \text{ mBq/dm}^3$,

range: 7.77-19.98.

Autumn: n = 29.

average: $^{218}Po = 39.59 \text{ mBq/dm}^3$,

range: 34.78-5.17,

average: $^{214}\text{Pb} = 77.33 \text{ mBq/dm}^3$,

range: 67.71-88.43, average: $^{214}\text{Bi} = 61.42 \text{ mBg/dm}^3$,

range: 52.54-72.15.

Winter: n = 37,

average: $^{218}Po = 39.59 \text{ mBq/dm}^3$,

range: 34.05–46.25,

average: 214 Pb = 54.02 mBq/dm³, range: $^{49.21-61.42}$,

average: 214 Bi = 47.73 mBq/dm³,

range: 42.18-54.02.

The detailed discussion of the above results will be given in our next paper [6].

4. CONCLUSIONS

- 1. The content of radon decay products within the recreational areas is smaller than that within the industrial region. Distribution of the frequency of occurrence of ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi fulfills the normal distribution.
- 2. In Poland in order to estimate the exposure to coal power station, the Mazurian Lake Region can be accepted as the reference system.

REFERENCES

- [1] CLEMENTS W. E., WILKENING M. H., Atmospheric pressure effects on ²²²Rn transport across the earth-air, Journal of Geophysical Research., 79 (1974), pp. 5025-5029.
- [2] EVANS R.D., Engineers quide to the elementary behaviour of radon daughters, Health Physics. 17 (1968), pp. 229-252.
- [3] JAWOROWSKI Z., Radiotoxicity influence of coal ash on people (in Polish), Postępy Fizyki Medycznej, 102 (1975), pp. 150-155.

- [4] KOPCEWICZ T., The variability of radioactive substances and radon decay products in the lower layer of troposphere (in Polish), Przegląd Geofizyczny, z-1, XIII(XXI) (1968), pp. 43-73.
- [5] NOWAK B., KWAPULINSKI J., Some parameters modelling the occurrence of products decay of radon.
- [6] NOWAK B., The dynamics of the variability of some radioisotopes in air under influence of power industry pollutions, Doctor's Thesis, Technical University, Gliwice 1986.
- [7] PEŃSKO J., WARDASZKO T., WADOCHA M., Natural atmospheric radioactivity and its dependence on some geophysical factors, Atompraxis, 14 (1968), pp. 255-258.
- [8] TSIVOGLOU E.C., AYER W.E., HOLADAY D.A., Occurrence of nonequilibrium atmospheric mixtures of radon and its daughters, Nucleonics, 11 (1953), pp. 40-48.

WYSTĘPOWANIE PRODUKTÓW ROZPADU RADONU W POWIETRZU NA OBSZARACH REKREACYJNYCH

Przedstawiono występowanie produktów rozpadu radonu w obrębie wybranych obszarów przemysłowych i rekreacyjnych w okresie 1976–1985. Występowanie ²¹⁸Po, ²¹⁴Pb, ²¹⁴Pb scharakteryzowano prezentując zmiany natężeń tych izotopów jako funkcję czasu i wysokości. Do określenia stężeń produktów rozpadu radonu użyto zmodyfikowanej metody Tsivoglou [8]. Aby wyeliminować fluktuację radonu z gleby, pomiary zostały wykonane na zbiorniku zaporowym Wapienica i jeziorze Seksty. Badania przeprowadzono w lecie, by uniknąć dodatkowych zanieczyszczeń tworzących się podczas spalania węgla.

Stężenia produktów rozpadu radonu w cyklu dzień-noc różnią się nieznacznie. Częstość ich występowania jest zgodna z rozkładem Gaussa. Szybkość samooczyszczania się powietrza na obszarach rekreacyjnych jest 50-80 razy większa, a wypłukiwanie badanych radioizotopów 10 razy mniejsze niż na obszarach z elektrowniami.

ВЫСТУПАНИЕ ПРОДУКТОВ РАСПАДА РАДОНА В ВОЗДУХЕ В ЗОНАХ ОТДЫХА

Представлено выступание продуктов распада радона в области избранных промышленных территорий и зон отдыха в период с 1976 по 1985 гг. Выступание ²¹⁸Ро, ²¹⁴Рb и ²¹⁴Ві охарактеризовали, представляя изменения концентраций этих изотопов как функцию времени и величины. Для определения концентраций продуктов распада радона употребили модифицированный метод Тсивоглию [8]. Чтобы устранить флуктуацию радона из почвы, измерения выполнили в бассейне Вапенница и на озере Сексты. Исследования проводили летом, чтобы избечь добавочных загрязнений, возникающих во время сгорания угля.

Концентрации продуктов распада радона в цикле день—ночь отличаются незначительно. Частота их выступания согласна гауссовскому распределению. Скорость самоочистки воздуха в зонах отдыха 50-80 раз больше, а вымывание исследуемых радиоизотопов 10 раз меньше, чем на территориях с электростанциями.