
ATMOSPHERIC RADON CONCENTRATION AROUND A PHOSPHOGYPSUM STACK AT WISLINKA (NORTHERN POLAND)*

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Abstract

The measurements of atmospheric radon concentration were carried out for one year at the turn of 2008/2009, using Kodak LR 115 passive track detectors. The average atmospheric radon activity near the phosphogypsum stack was 104 Bq m^{-3} . This is below the level accepted for indoor air. The results indicate that there are strong positive correlations between radon concentration and temperature ($r=0.9$) or atmospheric pressure ($r=0.9$) and a negative correlation between radon concentration and humidity ($r=-0.7$) or wind velocity ($r=-0.7$). Moreover, for all monitoring points the correlation between radon atmospheric concentrations measured in four seasons of the year were analyzed. The correlation coefficients are as follows: winter-summer 0.7, winter-autumn 0.2, winter-spring 0.2. Influence of radon exhalation from the stack was especially distinct in winter when the background radon activity was low. Spring and autumn farmland cultivation works increase radon exhalation from the soil, so that the contribution of radon emitted from the stack was less obvious.

Key words: radon activity concentration, phosphogypsum stack, correlation coefficients of radon concentration, meteorological parameters.

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STĘŻENIE RADONU W ATMOSFERZE W OKOLICY HAŁDY FOSFOGIPSÓW W WIŚLINCE (PÓLNOĆNA POLSKA)

Abstrakt

Pomiary stężenia radonu w atmosferze przeprowadzono w ciągu jednego roku, na przełomie lat 2008/2009, z użyciem detektorów Kodak LR-115. Średnie stężenie radonu w atmosferze wynosiło 104 Bq m^{-3} , co jest wartością poniżej dopuszczalnego stężenia tego gazu w budynkach mieszkalnych. Wykazano silne dodatnie korelacje między stężeniem radonu i temperaturą ($r=0.9$), i ciśnieniem atmosferycznym ($r=0.9$) oraz ujemne korelacje między stężeniem radonu a wilgotnością powietrza ($r=-0.7$) oraz prędkością wiatru ($r=-0.7$). Ponadto obliczono korelację między stężeniami radonu zarejestrowanymi w wyznaczonych punktach pomiarowych w poszczególnych sezonach. Współczynniki korelacji w sezonach: zima – lato, zima – jesień, zima – wiosna wynoszą odpowiednio: 0,7; 0,2; 0,2. Wpływ ekshalacji radonu z hałdy na stężenie radonu w atmosferze jest szczególnie widoczny w sezonie zimowym, w którym poza obszarami sąsiadującymi z hałdą rejestruje się najniższe w roku stężenie tego gazu. Wiosną i jesienią prace rolnicze wpływają na wzrost ekshalacji radonu z gruntu, przez co udział radonu, który wydostał się z hałdy, staje się mniej wyrazisty.

Słowa kluczowe: stężenie radonu, hałdy fosfogipsowe, współczynniki korelacji między stężeniem radonu a parametrami meteorologicznymi.

INTRODUCTION

The monitoring of atmospheric radon activity around a phosphogypsum stack located on the Baltic coast in northern Poland was carried for four seasons from autumn 2008 to autumn 2009. Phosphogypsum, a byproduct of phosphate processing, is a well known source of radon (RABI, MOHAMAD 2006, DUEÑAS et al. 2007, LYSANDROU et al. 2007). Phosphates, especially sedimentary ones, have a high uranium content. In deposits found in Marocco and Florida, it reaches $1500\text{-}1700 \text{ Bq kg}^{-1}$ (SKOROVAROV et al. 1998). Concentrations of the activity of natural radionuclides in phosphogypsum in different countries are listed in Table 1 (BERETKA, MATHEW 1985, FOURATI, FALUDI 1988, KOBAL et al. 1990, LAICHE, SCOTT 1991, LUTHER et al. 1993, RUTHERFORD et al. 1994, BURNETT et al. 1996, HULL, BURNETT 1996, PAPASTEFANOU et al. 2006, BORREGO et al. 2007, ABRIL et al. 2009, DUEÑAS et al. 2010). High radium and low uranium concentrations in phosphogypsum are connected with radionuclide separation during the wet acid process. Because of the high radium content, using phosphogypsum in civil engineering (RABI, SILVA 2006, REIJNDERS 2007, TAYIBI et al. 2009, MÁDUAR et al. 2011) and agriculture (PAPASTEFANOU et al. 2006, ABRILL et al. 2008, 2009) is controlled. In agriculture, the EPA radium activity limit is set at of 370 Bq kg^{-1} (EPA, *Federal Register* 1999).

The main aim of this study was to evaluate the environmental impact of a phosphogypsum stack on radon activity concentration. Another objective was to describe the seasonal variation of this phenomenon.

Table 1

Concentrations of natural radionuclides in phosphogypsum in different countries

Country/region	²²⁶ Ra	²¹⁰ Pb	²³⁸ U	²³² Th	²²⁸ Ra	⁴⁰ K	References
USA, Central Florida	907 (505-1353)	860 (578-11833)	132 (45-368)				Hull and Burnett, 1996
	820 (340-2000)		76				May and Sweeney, 1984 a,b fide in Papastefanou et al., 2006
	1100 (836-1230)	1370 (1270-1430)	130 (93-190)				Horton et al., 1988 fide in Papastefanou et al., 2006
	1140 (844-1670)						Guidry, 1990 fide in Papastefanou et al., 2006
	1140	1370	130				Rutherford et al., 1994
USA, North Florida	433 (270-598)	435 (347-553)	130 (23-452)				Hull and Burnett, 1996
	500 (477-548)						Rosler et al., 1979 fide in Papastefanou et al., 2006
USA, Louisiana	1100 (700-1700)						Laiche and Scott, 1991
USA, Mississippi	780						Mullins and Mitchell, 1990 fide in Papastefanou et al., 2006
Canada, Alberta	890			5.8			Luther et al., 1993
Australia	451			10			Beretka and Mathew, 1985
	500		10				Rutherford et al., 1994
Sweden	15		390				Rutherford et al., 1994
Hungary	1093				68		Fourati and Faludi, 1988
Yugoslavia	390						Kobal et al., 1988
Greece	633	476	28	3	3	8	Papastefanou et al., 2006
Spain	620		140				Borrego et al., 2007
Spain	730 (670-790)						Abrill et al., 2009
Spain	647			8		33	Dueñas et al., 2010
China	85	82	15				Burnett et al., 1996
Indonesia	473	480	43				Burnett et al., 1996
India	510	490	60				Burnett et al., 1996
Egypt	100						Burnett et al., 1996

MATERIAL AND METHODS

The examined phosphogypsum stack is located in the northern part of Poland, close to the Baltic Sea (Figure 1A), southeast of Gdansk, in the vicinity of a small village called Wiślinka.

The climate of the area transitional between maritime and continental. It is also strongly influenced by the lowland topography of this part of Eu-

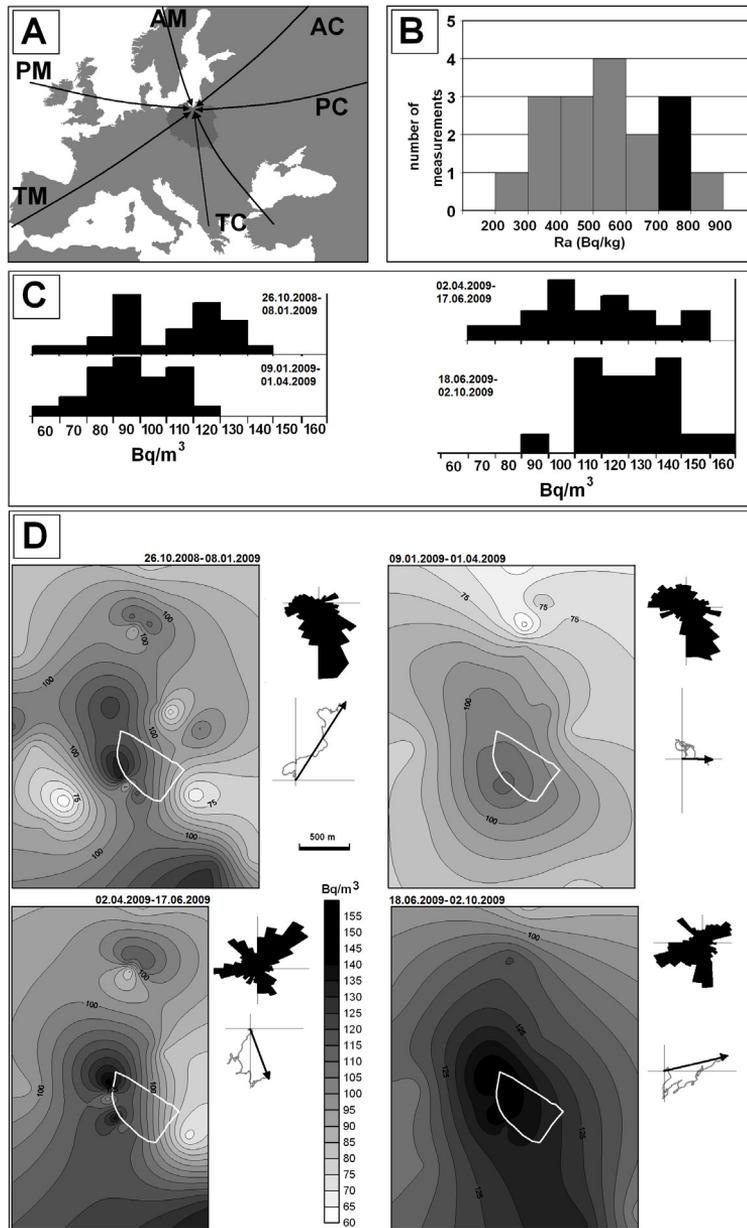


Fig. 1. Results of atmospheric radon monitoring: A) Area of the investigation and main directions of atmospheric air migration, B) Distribution of radium equivalent activity of the gamma dose for the Wiślinka phosphogypsum stack. Black class column corresponds to gamma spectrometric laboratory measurement of Wiślinka phosphogypsum, C) Seasonal variation of average radon activity concentration, D) Seasonal changes in spatial distribution of atmospheric radon activity around the phosphogypsum stack connected with the cumulative air movement vector and wind directions

rope, resulting in air masses moving quickly from the Atlantic Ocean or the North Sea. Arctic marine, arctic continental, polar marine, polar continental, tropical marine and tropical continental air masses enter this area depending on regional meteorological conditions.

The main pressure systems that affect the weather are the Icelandic low (stronger in winter) and the Azores anticyclone (more active in summer), and the changing atmospheric fronts from Asia: the East Asian high in winter and the South Asian low in summer. However, the Baltic Sea is the major contributor to the local climate.

This part of Poland has predominantly westbound winds, caused by the eastward movement of barometric lows from the Atlantic (Figure 1A). In summer months, westerlies (WSW) prevail, whereas in other seasons easterlies (NE) are more significant. The winds are typically weak to moderate, with velocity between 2 to 10 m s⁻¹. Strong and very strong winds occur at the seaside, causing storms; there their velocity may exceed 30 m s⁻¹. Southern and northern winds of local significance are sea and land breezes. In that area, the southern direction of winds is promoted by the local relief, dominated by the broad N-S Vistula River valley.

Spring starts in March, with temperatures varying from -1 to +20°C, and lasts until June. Summer is quite warm, with the temperatures recorded during our study up to 31.3°C. In autumn, days become colder until December, when winter begins and the temperature oscillates between a few degrees above 0°C, dropping to -16.7°C during our study. Winters last from December to March and include shorter or longer periods of snow. The ground is frozen from December to March. The average number of cloudy days a year, with the sky more than 80% overcast, is 120-160; there are 30-50 sunny days, with cloudiness below 20%. The annual precipitation ranges between 400 mm and 750 mm. The maximum precipitation occurs in autumn.

This area is a lowland and partly a depression, situated in the delta of the Vistula River. The fertile soils developed on alluvial muds encourage intensive agricultural activity. Locally, the subsoil bedrock consists of glacial sediments of the Pleistocene era as well as lacustrine and shallow marine sediments connected with various stages of Baltic Sea history.

The phosphogypsum stack covers an area of 36 ha and is 46 m high. Phosphogypsum dumping started in 1972. Since 1998, phosphogypsum stack has been reclaimed in the parts where the disposal was finished. Mixtures of water, sewage and grass seeds were used for rehabilitation of its surface.

Because of the ongoing reclamation works, the direct access to the tailing area was impossible, so the radiological characteristics of its material are based on the gamma dose data obtained by courtesy of the Fosfory Ciech company (BIERNACKA et al. 2005). Gamma dose variation in the tailing area varies in the range 144-400 nSv h⁻¹, which is an equivalent of radium activity in the range 337-937 Bq kg⁻¹ (Figure 1B). Radium activity of the

Wiślinka phosphogypsum, according to the gamma spectrometric laboratory measurements obtained by courtesy of Greenpeace (personal communication), is of the order 870 Bq kg^{-1} . Published data on radium activity from other phosphogypsum stacks show that in Moroccan phosphorites it varies in the range 580 ± 30 to $670 \pm 35 \text{ Bq kg}^{-1}$ (BOLÍVAR et al. 2009). This corresponds to the modal class distribution of the diagram presented in Figure 1B.

The monitoring of atmospheric radon activity was carried out from autumn 2008 to summer 2009 in 30 sites (some of the detectors during exposition were lost) localized around the phosphogypsum stack at Wiślinka. The measurements were carried out using Kodak LR-115 passive track detectors. The detectors were placed in chambers designed for protection against insulation and humidity. The chambers were fixed on trees at a height of 1.5 m above the ground. The detectors were exposed in the periods: 26.10.2008-8.01.2009, 9.01.2009-1.04.2009, 2.04.2009-17.06.2009 and 18.06.2009-2.10.2009, roughly corresponding to the seasons: autumn, winter, spring and summer. After exposure, the detectors were transported to a laboratory, where they were etched for 1.5 h in 2.5 N NaOH at 60°C . Tracks generated by the alpha-particle impact were counted using an optical microscope at the magnification of 100x. Density of the tracks was converted into radon concentration using calibration coefficients as described by SRIVASTAVA et al. (1995).

Data on the temperature, humidity, atmospheric pressure, wind velocity and direction were obtained by courtesy of the Foundation Armaag, which provides information on meteorological conditions in the Gdansk agglomeration. Their measurements were carried out in an automatic measuring station in the vicinity of the phosphogypsum stack. Average values for one-hour periods were obtained. Temporal variation in temperature, pressure, humidity and wind velocity was analyzed for series covering the radon monitoring period. In respect of winds, not only distribution of wind directions and velocity but also the combined parameter called cumulative air movement was analyzed.

RESULTS AND DISCUSSION

The average radon concentration for the whole measurement period was 104 Bq m^{-3} , which is significantly higher than 4.4 Bq m^{-3} and 6.5 Bq m^{-3} obtained for whole Poland by BIERNACKA et al. (1991) and Jagielak et al. (1998), respectively. Comparable results have been reported from Bad Gastein in Austria ($30\text{-}100 \text{ Bq m}^{-3}$; POHL-RÜLING, HOFMANN 2002) and from the coal mining town Nowa Ruda in Poland (98 Bq m^{-3} ; TCHORZ-TRZECIAKIEWICZ, SOLECKI 2011).

The seasonal variation in radon concentration in the atmosphere is presented in Figures 1C and 2. The highest values were measured in summer (the average 100 Bq m^{-3}), and the lowest – in winter (the average 88 Bq m^{-3}), whereas in spring and autumn the radon concentrations were intermediate.

The seasonal variation in meteorological parameters such as temperature, atmospheric pressure, relative humidity, wind velocity, wind direction and cumulative air movement is shown in Figure 2 and 1D. The average temperature varied from 1°C in winter to 17°C in summer; the average atmospheric pressure was from 1009 hPa in winter to 1015 hPa in spring; the average relative humidity was from 71% in spring to 84% in autumn; the average wind velocity was 2.1 m s^{-1} .

Correlation of the seasonal variation in the atmospheric radon concentration with the meteorological parameters is presented in Figure 3A. Pearson's correlation coefficients are shown above the trend lines. The correlation coefficients indicate that radon concentration was strongly positively correlated with temperature ($r=0.9$) and pressure ($r=0.9$). Correlation with humidity and wind speed was negative (inverse) and the same in both cases ($r=-0.7$).

The positive correlation between temperature and radon concentration is consistent with the results obtained by ISRAELSSON et al. (1972), MORISS and FRALE (1989), STRANDEN et al. (1991), SUNDAL et al. (2008), PRASAD et al. (2008). Those researchers have noticed that higher temperature increases radon exhalation from the ground and thus increases radon concentration in the atmosphere. The negative correlation between relative humidity and radon concentration is the result of negative correlation between temperature and relative humidity. It is commonly known that temperature increase results in the decrease of relative humidity.

Frozen soil and snow cover reduce radon exhalation from the ground (YAMAZAWA et al. 2005). In the investigated area, the snow cover and frost kept the winter average value of radon concentration the lowest of all seasons. The present results (Figure 1D) indicate that radon anomaly around the stack was more distinct. The most probable explanation is that the snow cover was thinner on the stack and thicker on the ground around the stack. Moreover the wind velocity and cumulative air movement vector were the lowest in winter (Figure 1D). The highest concentrations of radon were measured near the stack; further away the values were lower. Apart from the strong influence of the stack, the effect of the wind direction was noticeable. The concentric contours indicating radon concentrations around the stack are stretched in the NW-SE direction. It was only in winter that the relation between radon activity in the atmosphere and the distance from the stack and wind direction was so clear.

The positive correlation between atmospheric pressure and radon concentration indicates that increase in atmospheric pressure results in higher radon concentration. This is consistent with the results obtained by COSMA

et al. (1999) and CIGOLINI et al. (2009), but contradicts the findings of CLEMENTS and WILKENING (1974) and DÓRR and MÜNNICH (1990). The latter concluded that a 1% drop in atmospheric pressure results in an increase of radon exhalation by 6%.

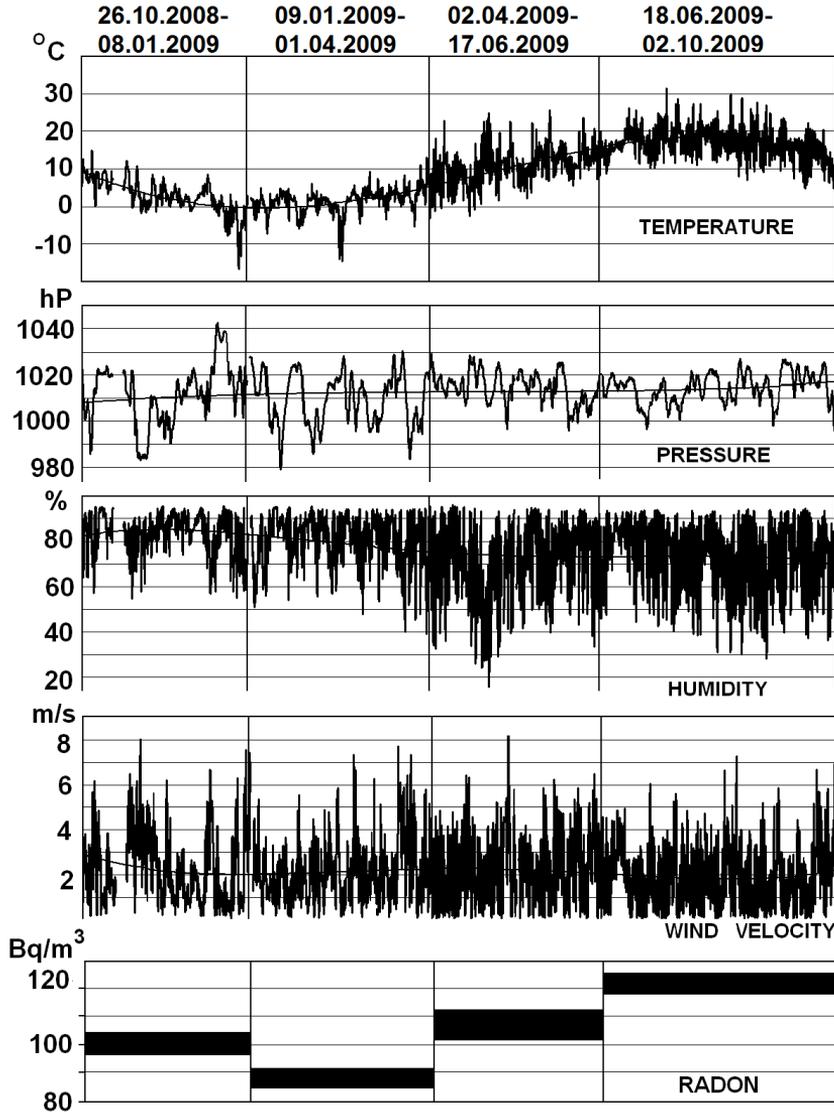


Fig. 2. Correlation of atmospheric radon activity with meteorological parameters

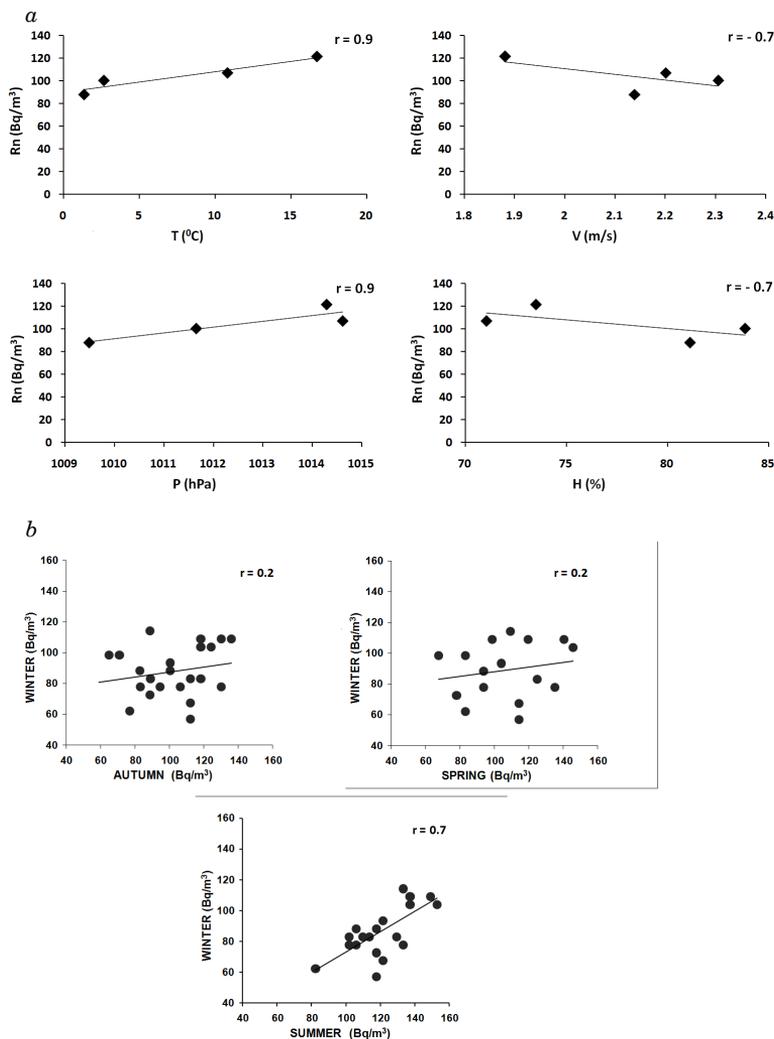


Fig. 3. Correlations of atmospheric radon activity concentration: *a* – with meteorological parameters, *b* – for results obtained at measurements points in various seasons

For all the monitoring points, the correlation between radon atmospheric concentration measured in various seasons was analyzed. The correlation coefficients were as follows: 0.7 for winter-summer, 0.2 for winter-autumn and 0.2 for winter-spring (Figure 3b). The positive correlation coefficient between winter and summer ($r=0.7$) means that the spatial distribution pattern of the measured values was similar in both seasons. The correlation coefficients between winter-autumn and winter-spring ($r=0.2$) indicate that there is no correlation between these seasons.

The lack of correlation of winter measurements with autumn and spring ones cannot be explained only by the changes of wind direction, because in the summer the wind direction was different than in the winter, but the correlation between these two seasons is still significant. There must be an additional factor other than wind direction which modifies the spatial distribution of the atmospheric radon concentration. The whole area around the stack is rural in character and most of the farming practice takes place in autumn and spring. In these seasons, the plant cover is less dense than in summer. Soil cultivation causes loosening of the soil structure, which can stimulate radon exhalation from the ground. The surveys done by CHAUHAN and CHAKARVARTI (2002), SHWEIKANI et al. (1995) support the hypothesis that loose soil structure is a factors that facilitates radon transport. Radon exhalation from cultivated fields strongly influenced its distribution pattern around the stack.

CONCLUSIONS

The measured radon concentrations do not exceed the values accepted for indoor air. The influence of radon exhalation from the stack was especially distinct in winter, against the background of generally low values of winter radon activity. Spring and autumn soil cultivation increased radon exhalation from the soil, so that the contribution of radon emitted from the stack was less obvious.

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