Radon potential of a fly ash pile - a criterion for its use as a building lot

Radonski potencial odlagališča elektrofiltrskega pepela kot merilo za njegovo uporabo kot gradbeno zemljišče

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Abstract: Radioactivity survey on a fly ash pile was carried out. Concentration of radon (^{222}Rn) in fly ash at a depth of 1 m was in the range of 0.3–46.9 kBq m^{-3}, with arithmetic mean of (23.8 ± 0.2) kBq m^{-3}. Radon exhalation rate was about 24 mBq m^2 s^{-1} on the part of the pile covered with grass, and about 37 mBq m^2 s^{-1} on the part with trees and bushes. Gamma dose rate was about 168 nSv h^{-1} and ash permeability around 3.9×10^{-13} m^2 on both parts.

Izvleček: Na odlagališču elektrofiltrskega pepela smo izvedli raziskavo radioaktivnosti. Na globini 1 m smo v elektrofiltrskem pepelu izmerili koncentracije radona (^{222}Rn) 0,3–46,9 kBq m^{-3} s povprečjem (23,8 ± 0,2) kBq m^{-3}. Na s travo poraščenem delu odlagališča smo izmerili hitrost ekshalacije radona okrog 24 mBq m^2 s^{-1}, na delu, kjer odlagališče preraščajo drevesa in grmičevje, pa okrog 37 mBq m^2 s^{-1}. Hitrost doze sevanja gama smo izmerili okrog 168 nSv h^{-1}, prepustnost pepela pa okrog $3,9 \times 10^{-13}$ m^2 na obeh predelih.

Key words: radon, concentration, exhalation rate, gamma dose rate, fly ash

Ključne besede: radon, koncentracija, hitrost ekshalacije, hitrost doze sevanja gama, elektrofiltrski pepel

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Introduction

Radon (^{222}Rn) is a radioactive noble gas (α radioactive transformation, half-life, $t_{1/2}$ = 3.82 d) originating from radioactive transformation of radium (^{226}Ra) in the natural radioactive chain of uranium (^{238}U) (Nazaroff & Nero, 1988). Only a small fraction of radon atoms emanate from the solid and enter the space between mineral grains, from where they migrate through the medium, by both diffusion and advection, and eventually exhale into the atmosphere (Etiope & Martinelli, 2002). Radon is always accompanied by its short-lived products (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) formed by its radioactive transformation and appearing in air as nano aerosols. Together, radon and radon short-lived products contribute more than half to the effective dose a member of the general public receives on the world average from all natural radioactive sources (UNSCEAR, 2000) and are a major cause of lung cancer, second only to cigarette smoking (Darby et al., 2005). Keeping low radon levels in dwellings and at workplaces is therefore a serious social concern and a great scientific challenge.

Radon enters the indoor air mostly from the ground on which a building is standing. Other radon sources, such as outdoor air, building material, burning natural gas and using water, are usually minor if not negligible. It is therefore important where and how the building is constructed. Particularly important is the quality of the basic floor slab and parts of the walls contacting the ground. Obviously, the higher the uranium content in the ground, and, consequently, radon concentration in soil gas, the higher quality is needed to keep radon level in indoor air acceptably low. In this sense, radon concentration in soil gas together with soil permeability, as a measure of radon potential (Wiegand, 2001; Neznal & Šmarda, 1996), should be considered when constructing a new, or remodelling an old building. In Germany, Kemski et al. (2001) have proposed the following ranking of radon risk with respect to radon concentration in soil: low at <10 kBq m^{-3}, medium at 10–100 kBq m^{-3}, increased at 100–500 kBq m^{-3}, and high radon risk at >500 kBq m^{-3}. In Sweden, the ranking is slightly different: low at <10 kBq m^{-3}, normal at 10–50 kBq m^{-3} and high at >50 kBq m^{-3} (EC, 2005). A classification of soil with respect to radon potential in Slovenia has not been accepted.

Radon potential varies markedly from soil to soil. It is reasonable to expect that it will be higher in the technologically enhanced naturally occurring radioactive material (TENORM). Such are wastes and by-products of technological processes in which some members in the uranium radioactive chain are concentrated and hence their contents elevated. An example is burning coal in a thermal power plant. As in any material, also in coal, there is certain, though low, level of ^{226}Ra which, after burning, grows concentrated in the fly ash. A question emerges what is the radon potential of a layer of fly ash and thus which radon risk is expected when using this layer as a building lot.

In Slovenia, there are several fly ash disposal sites of various sizes. For this study, a small fly ash pile of well defined geometry was chosen. In addition to radon concentration in fly ash also radon exhalation rate from fly ash, permeability of fly ash and gamma dose rate were measured, and the site was classified according to radon risk. These have been the first such measurements in Slovenia, aimed only at showing as an example how radon potential for this kind of sites may be dealt with.

Materials and methods

Site description

To the site selected as an example for this study, the fly ash of a thermal power plant burning lignite had been disposed of for years. At present it is a 5–7 m thick layer of an approximate 150 m × 200 m surface area.

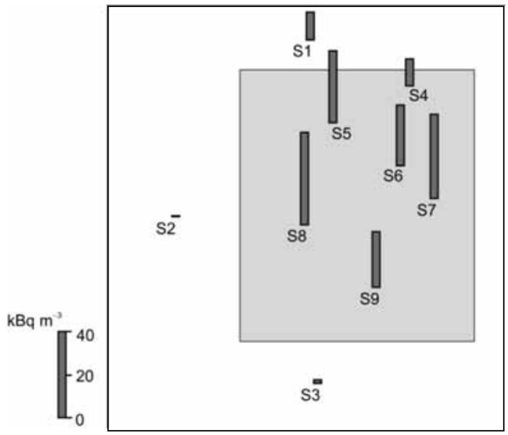

Figure 1. A schematic outline of the fly ash pile: the part covered by grass only is shadowed; measurement points are indicated, with vertical bars representing radon concentration in ash-gas.

A gamma spectrometric analysis of an averaged dry ash sample had shown the following concentrations of radionuclides (Bq kg^{-1}): ^{238}U: 200 ± 26, ^{232}Th: 37 ± 8, ^{226}Ra: 237 ± 8, ^{210}Pb: 102 ± 14, ^{40}K: 410 ± 23. For comparison, the following ranges were found in the terra rossa soil in various points in the karstic area: ^{238}U: 52–70, ^{226}Ra: 53–74, ^{40}K: 320–450, (Vaupotič et al., 2007a). A small part of the pile is a glade covered with grass only, while the rest, with dense bushes and trees (Figure 1). Complete measurements of all the parameters were carried out only in the former part because the movement of the equipment (except of scintillation cells) to the latter was practically impossible.

Radon in fly ash

To measure radon concentration (C_{Rn}/(Bq m^{-3})) in the ash-gas (gas contained in the bulk of fly ash) an AlphaGuard radon monitor and alpha scintillation cells were used. Because the cells showed similar values and are, in addition, more simple to move and use, only at two points in the part covered with grass the AlphaGuard monitor was used and the cells everywhere else.

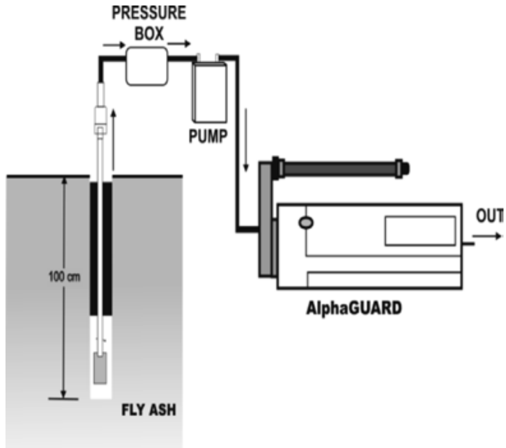

Figure 2. Schematic set-up for measuring radon concentration in ash-gas.

1. AlphaGuard

The measurement set-up to analyse radon concentration in soil gas consisted of an AlphaGuard PQ 2000 PRO (AG) radon monitor, a soil-gas probe and an AlphaPump (AP) (Genitron, Germany) (Figure 2). A borehole of 7 cm diameter was hand-drilled into the fly ash to a depth of 100 cm. The soil-gas probe was inserted to the bottom and the rubber ring around it inflated to isolate the bottom part of the borehole from the outdoor air. Soil gas was then pumped from the bottom through the AG ionization chamber at a flow rate of 0.3 dm^3 min^{-1}. The temporary radon (^{222}Rn) concentration was registered in one-minute intervals over approximately a 20-minute period. After initial growth, the concentration became stabilised. The average of the last few stabilised values was taken as the radon concentration in soil gas. At this low flow rate, contribution of thoron (^{220}Rn, half-life 55 s) was negligible (Žunić et al., 2006).

2. Alpha scintillation cells

For this experiment, the Spanish 0.3 dm^3 alpha scintillation cells were used (Quindos-Poncela et al., 2003). After the measurement with the AlphaGuard had been finished, a cell was connected to the soil-gas probe and pump, and ash-gas was flushed through the cell at a flow rate of 1 dm^3 min^{-1} for 3 minutes, necessary to exchange the air in the cell by the ash-gas sample. In a time longer than three hours, when the secular equilibrium between radon and its short-lived products had been reached, gross alpha activity of ^{222}Rn, ^{218}Po in ^{214}Po was measured in an PRM 145 α-counter (AMES, Ljubljana) (Figure 3). Cell efficiencies are from 0.000218 s^{-1} Bq^{-1} m^3 to 0.000428 s^{-1} Bq^{-1} m^3 and their background from 0.1 min^{-1} to 0.5 min^{-1}, thus assuring a lower limit of detection from 50 Bq m^{-3} to 110 Bq m^{-3} at counting times of 15 min.

Figure 3. Alpha counter with a scintillation cell.

Radon exhalation from fly ash

The radon exhalation rate E_{Rn}/(Bq m^{-2} s^{-1}) from soil was measured using the Exhalation Box (EB, dimensions 0.7 m × 0.7 m × 0.2 m) and the same AG monitor and AP pump as in the previous section (Figure 4). The air was circulated in the closed circuit for about 90 min and the concentration of radon accumulated in EB was recorded every 10 min. The exhalation rate was calculated using the formula:

$$E_{Rn} = B \times \frac{V}{F} \quad (1)$$

in which: B – slope of the straight line fixed to the increasing radon concentration points in the EB, V/m^3 – volume of the EB, F/m^2 – surface area covered by EB (Žunić et al., 2006).

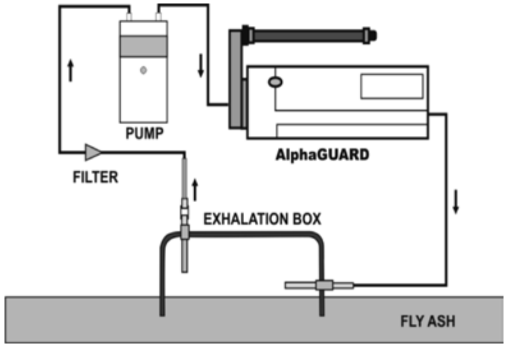

Figure 4. Schematic set-up for measuring radon exhalation rate from fly ash.

Fly ash permeability

The system to measure fly ash permeability k_{ash}/m^2 at 1 m depth consisted of a Multisensor Unit D/D device (Genitron, Germany) and the same AG monitor, AP pump and soil-gas probe as for measuring radon concentration (Figure 5). Ash-gas was sucked from soil by soil-gas probe and pumped through the AG and Multisensor. The pressure difference (ΔP) between ash and open air, and flow rate of ash-gas (Q) were measured by the Multisensor D/D. The fly ash permeability was calculated using a modified equation of Fick's law of diffusion (Janik, 2005):

$$k_{ash} = \mu \frac{Q}{W \times \Delta P} \quad (2)$$

in which: k_{ash}/m^2 – permeability of fly ash, μ – dynamic viscosity of air (Pa s), W/m – shape parameter of the soil-gas probe, Q/(m^3 min^{-1}) – gas flow rate and ΔP/Pa – pressure difference measured (Žunić et al., 2006).

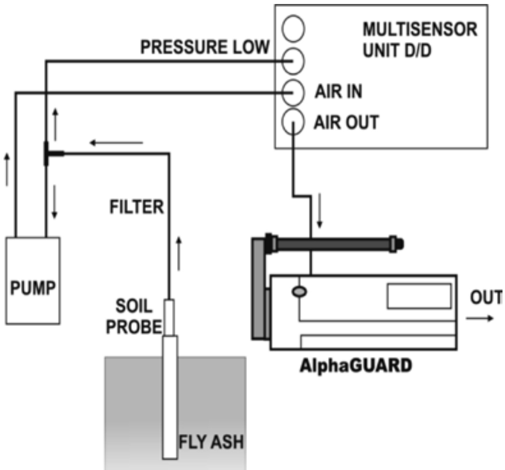

Figure 5. Schematic set-up for measuring fly ash permeability.

Gamma dose rate

Gamma dose rate $\dot{H}_\gamma$/(nSv h^{-1}) was measured in outdoor air at the height of 1 m above the ground using a GammaTracer TM Wide Type E probe (Genitron, Germany). The values of gamma dose rate were registered in 5-min intervals. The average value of 12–15 records was taken as a final result.

RESULTS AND DISCUSSION

Figure 6 shows the initial increase in readings when measuring radon concentration in the ash-gas with the AlphaGuard monitor. A time of about 20 min was needed to reach the correct, saturated value, taken as the results for C_{Rn}. Accumulation of radon under the exhalation box during exhalation rate measurements is presented in Figure 7. The initial slope of the curve was used to calculate E_{Rn} (in this case (10 ± 1.2) Bq m^{-3} min^{-1}).

Values of radon concentration in ash-gas are given in Table 1, together with radon exhalation rate, fly ash permeability and gamma dose rate. Based on good agreement between radon concentrations obtained with both devices in boreholes 1 and 6 (less than 15 % difference) we decided to use the AlphaGuard monitor only at two points easily accessible for the equipment, and scintillation cells everywhere else. Except at points 2 and 3, concentrations were in the range from about 10 kBq m^{-3} to about 45 kBq m^{-3}, with an average of (23.8 ± 0.2) kBq m^{-3}. This is similar as in gravel deposits and lake sediments (38.1 kBq m^{-3} and 20.3 kBq m^{-3}, respectively) but lower than at carbonates (50.4 kBq m^{-3}) in Slovenia (VAUPOTIČ et al., 2007b; VAUPOTIČ et al., 2008), although ^{226}Ra concentration in fly ash is about four times higher, and therefore higher radon concentration would be expected, than in an ordinary soil (VAUPOTIČ et al., 2007a). The lowest two values (points 2 and 3) belong to boreholes at the very edge of the pile where the ash layer was thinner. Differences in values among points (except 2 and 3) are normal, as it is well known that even at a homogeneous distribution of ^{226}Ra, radon levels may differ markedly from borehole to borehole because of micro fractures in the ground (DURRANI

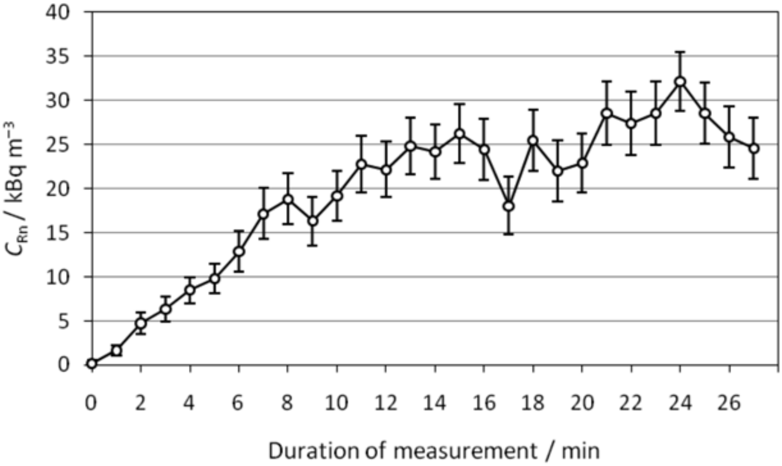

Figure 6. Initial increase in readings (S6) when measuring radon concentration in the ash-gas with the AlphaGuard monitor.

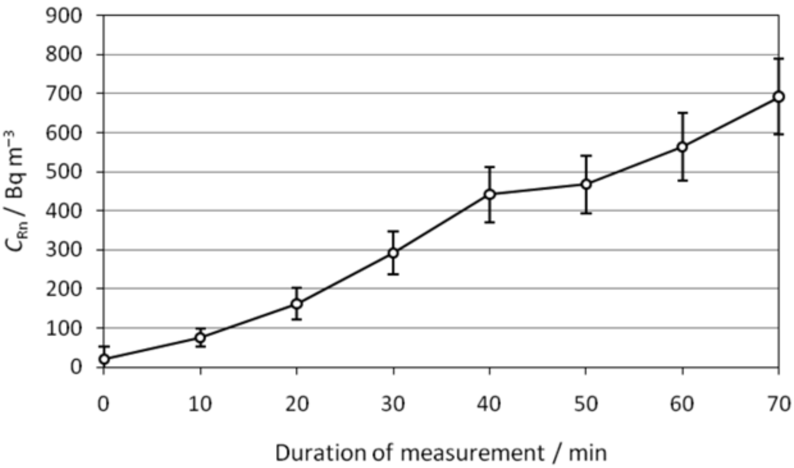

Figure 7. Accumulation of radon under the exhalation box during exhalation rate measurements (S1).

& Badr, 1995; Baixeras et al., 1996; Kemski et al., 2001; Winkler et al., 2001; Iakovleva & Ryzhakova, 2003; Vaupotič et al., 2007a). Point 4 is at the same level as other points but only 1 m or so away from the steep edge of the pile. We may speculate that atmospheric air may penetrate horizontally into the borehole, diluting the ash-gas and thus reducing radon concentration. Such a situation has not been observed at point 7 where radon concentration was second the highest. Although the radon exhalation rate is similar as somewhere else in Slovenia (VAUPOTIČ et al., 2010; VAUPOTIČ et al., 2007b) it is higher at point 1 (36.9 mBq m^{-2} s^{-1}) than at point 8 on glade (24.2 mBq m^{-2} s^{-1}), most probably because the roots of trees and bushes here break up the bulk structure of the pile and thus enhance radon diffusion (MOLDRUP et al., 2000). Also the permeability of fly ash does not differ from that measured at other places (VAUPOTIČ et al., 2010; VAUPOTIČ et al., 2007b). It is practically the same at point 1 and 6, thus pointing out that higher exhalation rate at point 1 is due to enhanced diffusion (MOLDRUP et al., 2000) and not higher advection in the ash.

Gama dose rate of 168 nSv h^{-1} is about 50 % higher than over a nearby ground and about 15 % (27 nSv h^{-1}) lower than over the Pohorje granite (195 nSv h^{-1}) (BRAJNIK et al., 1992).

With respect to radon concentration in ash-gas (never exceeding 50 Bq m^{-3}), this fly ash pile may be considered as a building lot with a normal radon risk according to the Swedish (EC, 2005) and a medium radon risk according to the German (KEMSKI et al., 2001) classification. This classification is conservative, because the measurements were carried out in summer after a long period of dry weather when radon concentration is higher than it would be in wet condition (HOSODA et al., 2007; PAPACHRISTODOULOU et al., 2007).

CONCLUSIONS

Concentration of radon in ash-gas was in the range of 0.3–46.9 kBq m^{-3}, with arithmetic mean of (23.8 ± 0.2) kBq m^{-3}, and radon exhalation rate about 24 mBq m^2 s^{-1} on the part of the pile cov-

Table 1. Radon concentration (C_{Rn}), obtained with alpha scintillation cells and AlphaGuard monitor, radon exhalation rate (E_{Rn}), permeability of fly ash (k_{ash}) and gamma dose rate ($\dot{H}_{\gamma}$) on the fly ash pile.

Meas. point	C_{Rn}/(kBq m^{-3}) Scintillation cell	C_{Rn}/(kBq m^{-3}) AlphaGuard	E_{Rn}/(mBq m^{-2} s^{-1})	k_{ash}/m^2	$\dot{H}_{\gamma}$/(nSv h^{-1})
S1	13.6 ± 0.3	15.5 ± 2.9	36.9 ± 4.9	3.7×10^{-13}	166
S2	0.3 ± 0.05				
S3	1.2 ± 0.08				
S4	13.0 ± 0.4				
S5	37.0 ± 0.6				
S6	30.8 ± 0.6	26.5 ± 3.3		4.0×10^{-13}	169
S7	42.8 ± 0.6				
S8	46.9 ± 0.7		24.2 ± 2.5		
S9	28.9 ± 0.5				

ered with grass, and about 37 mBq m^2 s^{-1} on part with trees and bushes. According to the German classification the pile may be considered as a building lot with medium radon risk, and according to the Swedish, normal radon risk.

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