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Study of a remediated coal ash depository from a radiological perspective

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ABSTRACT

Coal-fired power plants play a significant role in the production of electricity. The Ra-226 concentration of coals mined in the Ajka region can reach up to 3000 Bq/kg. This study focuses on the effects of a Hungarian (Ajka) remediated coal ash depository on the environment and the effectiveness of the cover layer. During the remediation, a method patented in Hungary was used, in which the upper layer of the depository, which had settled like concrete, was ploughed and mixed with woodchips before being planted with vegetation.

The gamma dose rate H*(10) of the depository and its vicinity was measured using Automess 6150AD-b at 32 points, surface Rn-222 exhalation at 19 points and air radon concentration at 34 points; at 32 points, soil gas radon content was measured with AlphaGUARD and soil permeability with RADON-JOK. The nuclide content of nine samples was determined using an HPGe gamma spectrometer and their Rn-222 exhalation rates were measured using the AlphaGUARD.

H*(10) was 290 (130–525) nSv/h at the covered depository; $C_{Ra\text{-}226}$ was 1997 Bq/kg, 960 Bq/kg and 104 Bq/kg for the ash, cover layer and background soil respectively. $C_{Rn\text{-}222}$ in the soil was 25–161 kBq/m^3, and soil gas permeability K was between 6.4E-13 and 1.80E-11 m^2. The radon exhalation of the uncovered and covered depository was 259–1100 mBq/m^{2}s. The exhalation and emanation coefficients of the samples were 0.05–0.32 mBq/kgs and 8–22%. The effects of vegetation on the migration of radon were also examined. The results show that the Ajka coal ash depository involves higher radiological risk than that reported by previously published studies on depositories. The applied cover layer halved the field radon exhalation; in addition, the vegetation reduced the convective airflow and, with this, the migration of Rn.

1. Introduction

In 2008, coal contributed 41% of the world's energy production, and the volume of coal mined in 2015 was 8022.5 Mt (Lior, 2010; World Coal Association, 2015). Combustion waste formed during the combustion of coal, such as ash and slag, has a non-negligible effect on environmental pollution (Papastefanou, 2010). Coal always contains radionuclides of natural origin, and the average specific radioactivity of both U-238 and Th-232 in coal is generally around 20 Bq/kg (range 5–300 Bq/kg); however, this can reach up to 15,000 Bq/kg for uranium (UNSCEAR, 2008).

Uranium and radium are enriched in ash and slag compared to coal (Jala and Goyal, 2006; Mathur et al., 2008; Bhangare et al., 2014; Sahu et al., 2014; Yao et al., 2015). Through the valorization of coal ash, the number of depositories and consequently their environmental impact can be reduced. The most common method of handling of coal ash is deposition, although valorization is becoming more widespread. Fly ash has been used by the cement industry for many years (ASTM C618-08, 2008) and several additional valorization methods exist for coal ash, e.g., the formation of concrete bricks, blocks, dams, lightweight concrete and insulation (Yao, 2013, 2015; Wang et al., 2014; Angjusheva et al., 2013; Somlai et al., 1997a, 1997b, 1997c, 1998, Somlai and Kovacs, 2007). Both in cases of direct valorization and further processing, non-negligible radioactivity must be taken into consideration (Singh et al 2015; Bhangare et al., 2014; Mathur et al., 2008; Feng and Lu, 2016; Norm4building).

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The deposited materials may cause harmful effects, such as increased gamma doses to the proximate habitat, and a release of radon and its progeny, aerosols and leaching (Carlson and Adriano 2009; Mljac and Križman, 1995; Ramola et al., 2013). In order to mitigate these harmful effects, the depositories are covered and remediated. A long-term programme has been introduced for the monitoring of pollution (Máté et al., 2013; Juhász et al., 2001; Mljac and Križman, 1995). The direct gamma dose rate decreases with distance, and is only significant at the surface of the depository and within its direct environment. By contrast, the nuclides that are spread into the environment can travel a long way through the movement of the atmosphere; nuclides therefore have an effect on the environment and human health at greater distances (Papp et al., 2002; Papp and Dezso, 2003; Meszaros et al., 2016). Thus, we examined the efficiency of the cover layer in terms of radon exhalation, since radon can move up through the soil and be exhaled at the surface. The exhalation (E) is the amount of radon released from the examined material related to mass or surface unit of material per time unit, and is measured in Bq/kgs or Bq/m^2s (IAEA Technical Reports Series 474). In the case of coal ash, this radon exhalation depends on Ra-226 content and other factors such as pore size, specific surface, ash content, combustion temperature, superficial morphology, grain size distribution, grain density, homogeneity of Ra-226 distribution within the grain and humidity (Jobbagy et al., 2009; Sas et al., 2012; Stajic and Nikezic, 2014, Vaasma et al., 2014; Kikaj et al., 2016). An increase in outdoor radon concentration can be expected in the ash depository environment (Girault et al., 2016).

The Hungarian coal ashes, in particular, exceed the NORM limits (1,000, 1000 and 10,000 Bq/kg for Ra-226, Th-232, K-40 respectively) (Norm4building; IAEA TECDOC 1712); this is confirmed by previous studies of the uranium content of coal from the Ajka region, which found that this level can reach up to 800–900 Bq/kg, and even 2000 Bq/kg in exceptional circumstances (Szalay et al., 1959). Following combustion, this value increases further to 600–3000 Bq/kg Ra-226 in the coal ash (Somlai et al., 1996; Papp et al., 2002). The grey tailing is dried and bound to concrete-like converted coal ash. Since 1986, approximately 11 million tonnes of ash have been produced by local industry (Horváth, 2002). The application of an inactive cover layer can reduce by as much as several orders of magnitude the radon exhalation of the depository (Exh_{field}) (Gorjánácz et al., 2004; Juhász et al., 2001; Várhegyi et al., 2013; Szerbin et al., 2005a; 2005b).

A conventional cover layer of soil, loess or clay was not used in Ajka. Instead, a new method patented in Hungary was employed for covering the depository. Woodchips were placed on the dried and bound surfaces of the depository (minimum 1000 m^3/acre) and the concrete-like ash was subsequently ploughed to a depth of about 40–50 cm and mixed with woodchips. Following this, a forest was planted (Horváth, 2002). The Ra-226 content of the cover layer depends on the depth of ploughing. The remediation was finished in 1992; it was successful and the landscape is similar to the undisturbed area. Radiological surveys were carried out sporadically at this depository following the remediation; however, it is only now, more than 20 years later, that enough time has elapsed to allow a full examination of the efficiency of the remediation.

In this study, the radiological risk, gamma dose, soil gas, outdoor radon concentration, geogenic radon potential (GRP) and radon availability (RA) of the remediated depository are investigated.

2. Materials and methods

2.1. Sampling area

The measuring points shown in Fig. 1 were chosen to represent the different surfaces that can be found on the depository. The two stars represent the GPS coordinates. The depository was divided into three main sections, based on the characteristics of its ground and vegetation, and an area located beside the ash dump was taken as the background. The details of these three main sections and the background were as follows:

1) Belt line: Meeting line between the wall of the depository and its vicinity; no cover layer, the ash here is cut up. Despite this, the trees were already in leaf (closed foliage). Point 0.
2) Slope: Uncovered ash; the soil here consists partly of cut-up ash. Similarly to the belt line, it is covered with vegetation. Another part of this area is a concrete-like layer, which is completely uncovered over a 3–4 m high strip. Point 11.
3) Cover layer with vegetation: More than 90% of the depository falls into this category. This area was further categorized into three characteristic groups according to the vegetation appearing (these groups were situated sporadically and irregularly): open forest with sparse undergrowth, situated at the edge of the covered depository (points 1, 2 and 3); closed forest with sparse undergrowth (points 6 and 9); closed forest with thick undergrowth (points 4, 5, 7, 8, 10, 12 and 13).
4) Vicinity grassland: Points 14, 15 and 16. These points were taken as the background, 50 m away from the depository, and 20 m away from each other

2.2. Sampling for laboratory measurements

In all, nine samples were collected for laboratory measurements, three from the cover layer, three ash (points 1,4 and 6) and three background (points 14–16). The following procedure was used. From the cover layer and the background, (soil) samples were taken from the upper 5 cm layer after the vegetation was removed. The ash samples were collected from the upper 10 cm layer after removing the 20–50 cm cover layer.

2.3. Field measurements

2.3.1. Gamma dose rate H*(10)

In order to determine the gamma dose, the 6150AD-b Dose Rate Meter (Automess), a device suitable for the measurement of H*(10), was chosen (detection limit 1.3nSv/h). H*(10) was measured in units of μSv/h (UNSCEAR, 2013).

The measurements were carried out at 16 points, at a height of 100 cm from the soil surface and directly on the soil surface; in total, therefore, 32 sampling points were used for measuring values. At the soil surface, the measurement was carried out at a single point, while at a height of 100 cm, an area of approx. 10 m^2 was measured, and an average value calculated for this area. The height used for measurement was chosen as 100 cm since the terrestrial component dose conversion factors are given for the absorbed dose at this height (Quindos et al., 2004; Clouvas et al., 2000).

2.3.2. Outdoor radon activity concentration (C_{Rn-222} in air)

Measurements of C_{Rn-222} in air were carried out using the AlphaGUARD PQ2000 (Saphymo) radon device (referred to as AlphaGUARD) with the 10-min diffusion mode, for a measurement time of one hour for each sampling point, although only the final four data were used for calculation. The lower limit of detection was less than 2 Bq/m^3. The measurements were performed at heights of 0, 100, and 150 cm between 8 a.m. and 5 p.m., using the same device. For the examination of C_{Rn-222} in air vs. height, measurements were carried out from 0 to 250 cm at 50 cm intervals, on

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