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Radiological survey of the covered and uncovered drilling mud depository

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ABSTRACT

In petroleum engineering, the produced drilling mud sometimes contains elevated amounts of natural radioactivity. In this study, a remediated Hungarian drilling mud depository was investigated from a radiological perspective. The depository was monitored before and after a clay layer was applied as covering.

In this study, the ambient dose equivalent rate H*(10) of the depository has been measured by a Scintillator Probe (6150AD-b Dose Rate Meter). Outdoor radon concentration, radon concentration in soil gas, and in situ field radon exhalation measurements were carried out using a pulse-type ionization chamber (AlphaGUARD radon monitor). Soil gas permeability (k) measurements were carried out using the permeameter (RADON-JOK) in situ device. Geogenic radon potentials were calculated. The radionuclide content of the drilling mud and cover layer sample has been determined with an HPGe gamma-spectrometer. The gamma dose rate was estimated from the measured radionuclide concentrations and the results were compared with the measured ambient dose equivalent rate.

Based on the measured results before and after covering, the ambient dose equivalent rates were 76 (67–85) nSv/h before and 86 (83–89) nSv/h after covering, radon exhalation was 9 (6–12) mBq/m^{2}s before and 14 (5–28) mBq/m^{2}s after covering, the outdoor radon concentrations were 11 (9–16) before and 13 (10–22) Bq/m^{3}after covering and the soil gas radon concentrations were 6 (3–8) before and 24 (14–40) kBq/m^3 after covering. Soil gas permeability measurements were 1E-11 (7E-12-1E-11) and 1E-12 (5E-13-1E-12) m^2 and the calculated geogenic radon potential values were 6 (3–8) and 12 (6–21) before and after the covering. The main radionuclide concentrations of the drilling mud were $C_{U\text{-}238}$ 12 (10–15) Bq/kg, $C_{Ra\text{-}226}$ 31 (18–40) Bq/kg, $C_{Th\text{-}232}$ 35 (33–39) Bq/kg and $C_{K\text{-}40}$ 502 (356–673) Bq/kg. The same radionuclide concentrations in the clay were $C_{U\text{-}238}$ 31 (29–34) Bq/kg, $C_{Ra\text{-}226}$ 45 (40–51) Bq/kg, $C_{Th\text{-}232}$ 58 (55–60) Bq/kg and $C_{K\text{-}40}$ 651 (620–671) Bq/kg.

According to our results, the drilling mud depository exhibits no radiological risk from any radiological aspects (radon, radon exhalation, gamma dose, etc.); therefore, long term monitoring activity is not necessary from the radiological point of view.

1. Introduction

Oil is the main energy source of our civilization. In 2013, it contributed 31% of the total energy consumption. Oil and gas not only play a role in power and transportation, but are also the raw materials for the petrol chemistry industry (Alessandro, 2013; Lior, 2010; IEA, 2015). In 2014, 4200 million tons of crude oil was mined in the world and 3524 billion m^3 of gas was produced. During the production of oil, various types of waste are produced, such as sludge, mud and water. These waste products of the oil industry (mud, sludge, sand, oil shale, ash, etc.) can contain elevated concentrations of radionuclides from the uranium and thorium series, and because of the elevated radioisotope concentrations they often fall into the NORM (Naturally Occurring Radioactive Material) category (EU BSS, 2013; IAEA-TECDOC- 474, Unscear, 2008 B). An oil well can produce up to 1000 tons of NORM solid waste annually (AL Nabhani et al., 2015). The radionuclide concentrations in the waste of the oil industry can have variations of several orders of magnitude (0–1000 Bq/kg) depending on which part of the technology produced the mud or scale (Bakr, 2010, Gazineu et al., 2005; Shawky et al., 2001; Al Saleh et al., 2008; Xhixha et al., 2015; Hilal et al., 2014). This can cause a significant increase in the radiation dose experienced by the workers in the oil industry, leading to detrimental health effects (Darby et al., 2005). This is the reason why the waste products from the oil industry have to be characterized radiologically, and if the characterization reveals that it is necessary, constant monitoring is also required.

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In this study, drilling mud deposits were surveyed from a radiological point of view. The deposits originated from the digging of Hungarian oil and gas wells. The drilling mud provides cooling and adequate lubrication to the drilling head during operation, regulates the pressure in the drilling hole, seals off porous geologic formations, and carries geologic drill cuttings from the bottom of the well up to the surface (Penn et al., 2014; Oreshkin et al., 2015; Ukeles and Grinbaum, 2004). Typically, water-based drilling mud (WBM) is composed of colloidal clays (bentonite), potassium-chloride, sodium-hydroxide, lignite, barium-sulphate, mica, ground nutshells, polymers, and a number of other additives, depending on the needs of the particular well (Whitaker et al., 2016). There are multiple opportunities for the valorization of drilling mud that has been used several times and that cannot be reused again. Possible uses for drilling mud include underground injection, application to agricultural fields, and usage in the building industry as a raw material. There are multiple choices for the handling of material that cannot be utilized, either because of its properties or for economic reasons, such deposition in pits or mud boxes, use in landscaping, dispersal over the ground, discharge to the sea or ocean, slurry injection, disposal in salt caverns, on-site burial (reserve pits) or storage in hazardous waste landfills. In the case of NORM disposal, the routes of exposure include external radiation, inhalation of dust (including radon and thoron progenies) and intake through the food chain (consumption of leafy vegetables, drinking water, fish and meat) (Kontol et al., 2015). These waste products, if placed in nature after utilization, can contribute to the radiation dose of the general population through the aforementioned routes; thus, in addition to the standard chemical characterization of the materials, it is wise to consider the radiological aspects as well. The harmful effects can be mitigated by the insulation and covering of the depositories (Várhegyi et al., 2013; Jonas et al., 2017).

The main aim of this survey was to determine whether the application of any long-term radiological monitoring system is necessary for the covered or uncovered depositories.

1.1. Area description

Drilling mud depositories in Zalatárnok, Hungary have been surveyed from a radiological point of view both before and after they were covered with clay. The ambient dose equivalent rates on the field have been measured and the gamma dose rates calculated from the concentrations of the terrestrial radionuclides. The migration through air has been checked by the measurement of radon in the soil gas, in situ radon exhalation (exhalation) and the radon concentration in air. There are no active faults or tectonic movement that could cause radon anomalies (Iovine et al., 2017). By measuring the U-238 and Ra-226 concentrations and their ratios in the water of monitoring wells, the radioactive contamination of ground water and deeper water layers has been investigated. In the case of oil and gas fields, the ratio of U-238/Ra-226 in the reservoir rock and the new NORM material is different. The U-238 series, up to Th-230, is not or is hardly able to be mobilized from the reservoir rock, while Ra-226, as is true of other elements in the II group of the periodic table, is water soluble (IAEA-SAFETY REPORTS-34). This is the reason why NORM materials from the oil and gas industry can have this disequilibrium, and in cases in which increased amounts of radionuclides get into the environment (if there is no other disturbing effect) this disequilibrium (which can be greater than an order of magnitude) can be observed (Hilal et al., 2014).

2. Materials and methods

2.1. Sample collection

In the current study, we have carried out a radiological survey of the waste depository of the MOL Group at Zalatárnok. There are three depositories in the area (Fig. 1.) Two of them (Depository C, 90 × 150 m, and Depository B, 90 × 150 m) were already covered before the start of this study (and consequently only the covered state could be surveyed), but the third (Depository A 70 × 30 m) was investigated both in a covered and in an uncovered state.

During recultivation, a 1.5 m clay covering layer has been used on all depositories. Drilling mud samples have been taken from the uncovered depository at four points, 15 m apart, and at two different depths (0.2 m, 0.4 m). For the covered depositories, only the cover layer (clay) has been sampled at a 0.2 m depth at three points, 20 m apart on each depository (the clay used for the cover has been considered homogeneous, since the same mine has been used for the whole operation). Soil and drilling mud samples were 10 kg each. The soil and drilling mud samples have been carried to the laboratory in plastic vessels. Water samples (5 L) were also taken at two borehole wells (at depths of 12 m and 30 m) with the help of a windlass from the three groundwater monitoring wells around the waste depository (the distance of the wells from the depository is approximately 50 m), which were acidified and carried to the laboratory for further measurements.

2.2. Field measurements

The field measurements were carried out at the soil sampling points.

2.2.1. Ambient dose equivalent rate H*(10), and the calculated gamma dose rate $D_{\gamma r}$

The ambient dose equivalent rate measurements were carried out at a height of 1 m with a NaI(Tl) Scintillator Probe (6150AD-b Dose Rate Meter, Automess) (detection limit 1.3nSv/h). The device is suitable for the measurement of H*(10), in units of nSv/h (UNSCEAR, 2013). In every case, a 10 m^2 area was measured at 10 points and the average calculated. In case of Depository A, H*(10) was measured in four places (4 × 10 = 40 points) both before and after covering, while for the other two depositories only the covered state was measured at three places (3 × 10 = 30 points) in every depository. The gamma dose rate at 1 m from the soil has also been calculated from the concentrations measured in the upper 0.2 m of the soil using Equation (1), according to Quindos et al. (2004):

$$D_{\gamma r} = 0.4551 * C_{Ra-226} + 0.5835 * C_{Th-232} + 0.0429 * C_{K-40} \quad (1)$$

where $D_{\gamma r}$ is the calculated gamma dose rate at 1 m in nGy, and $C_{Ra\text{-}226}$, $C_{Th\text{-}232}$ and $C_{K\text{-}40}$ are the radionuclide concentrations in the soil, expressed in Bq/kg. During calculations, the terrestrial and cosmic components have been calculated separately followed by the summation. For the terrestrial dose rate component, the radionuclide concentrations determined in Section 3.5 have been used, while for the cosmic component the value of 31 nSv/h for cosmic radiation measurable at sea level and a value of 0.2 km height above sea level have been used in the equation described by Bouwille and Lowder (Unscear, 2008; Bouville and Lowder, 1988). The contribution of other sources of gamma radiation in the air has been considered negligible.

2.2.2. Outdoor radon concentration ($C_{Rn\text{-}222}$ in air)

Outdoor radon concentration measurements were carried out using a pulse-type ionization chamber containing a radon device (AlphaGUARD PQ2000 Saphymo, referred to as AlphaGUARD) with a 10-min diffusion mode. Measurement were taken for 1 h at each sampling point at a height of 1 m (only the final four values were used for calculation). The lower limit of detection was less than 2 Bq/m^3.

2.2.3. Radon concentration in soil gas ($C_{Rn\text{-}222}$ in soil)

Radon concentration in the soil gas measurements were carried out using an AlphaGUARD with the 10-min flow mode, using a soil gas probe and a pump (AlphaPUMP, Genitron Instruments) (0.5 L/min). To eliminate the disturbing effect of thoron, the following method was applied (Jonas et al., 2016):