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Characterization of the natural radioactivity of the first deep geothermal doublet in Flanders, Belgium

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ABSTRACT

Deep geothermal energy is a local energy resource that is based on the heat generated by the Earth. As the heat is continuously regenerated, geothermal exploitation can be considered as a renewable and, depending on the techniques used, a sustainable energy production system. In September 2015, the Flemish Institute for Technological Research (VITO) started drilling an exploration well targeting a hot water reservoir at a depth of about 3 km on the Balmatt site near Mol. Geothermal hot water contains naturally occurring gases, chemicals and radionuclides at variable concentrations. The actual concentrations and potentially related hazards strongly depend on local geological and hydrogeological conditions. This paper summarizes the radiological characterization of several rock samples obtained from different depths during the drilling, the formation water, the salt and the sediment fraction. The results of our analyses show low values for the activity concentration for uranium and thorium in the formation water and in the precipitate/sediment fraction. Also, the activity concentrations of ^{210}Pb and ^{210}Po are low in these samples and the activity concentration of ^{226}Ra is dominant. From the analysis of the rock samples, it was found that the layer above the reservoir has a higher uranium and thorium concentration than the layer of the reservoir, which on the other hand contains more radium than the layer above it.

1. Introduction

Deep geothermal energy is a local energy source that is based on the heat generated by the Earth. As the heat is continuously regenerated, geothermal exploitation can be considered as a renewable and, depending on the techniques used, a sustainable energy production system. The hydrothermal resources are the only kind in wide use today, e.g. in a doublet system using a hydrothermal aquifer (Rybach, 2007). In order to explore the deep geothermal potential in Flanders, the Flemish Institute for Technological Research (VITO) started drilling an exploration well in September 2015. The well targeted a limestone reservoir at a depth of about 3 km at the Balmatt site near Mol. The purpose was to demonstrate that it is possible to pump up hot water from the Lower-Carboniferous limestone reservoir and to use the extracted heat for heating purposes and electricity production. If the first and the second well, completing the geothermal doublet, meet expectations, it opens possibilities for the construction of similar geothermal power plants in the Belgian provinces Antwerp and Limburg (Loveless et al., 2015, VITO website).

In the early eighties a well of 1761 m depth was drilled in Merksplas-Beerse. This well proved the geothermal potential of the Carboniferous Limestone Group, but due to the moderate depth compared to that of the Balmatt site, this source could only be used for heating (Vandenberghe et al., 2000). The exploitation of geothermal energy is considered to cause little or no harm to the environment. It is known that geothermal steam and hot water, however, contain naturally occurring gases, chemicals and radionuclides at variable concentrations. The actual concentrations and the potentially related hazards strongly depend on the local geological and hydrogeological conditions. Untill now, little data were available about the concentrations of radionuclides in deep geothermal reservoirs and in the hot fluids produced from them. Hence the radionuclides and the way they may be released in the environment have to be investigated and controlled if necessary. Because the formation water brings these byproducts to the surface and in the installation, it is important to make a radiological characterization for the naturally occurring radioactive material (NORM) (Scheiber et al., 2012; Regenspurg et al., 2014).

In European regulations, the radiation risks linked to the exploitation of geothermal installations have been included in Basic Safety

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Standards for Radiation Protection (EU Directive 2013/59/Euratom), where these installations have been considered as a NORM practice of concern to the Member States. At the Belgian level *"geothermal energy production, including exploration and pumping activities in the development thereof"*, has recently been added as a NORM practice to national radiation protection legislation by the (Federal Agency for Nuclear Control - FANC) FANC-Decree of March 3, 2016. This addition should ensure control of radiation protection issues by the national regulatory body (FANC) in the installations and in the safe disposal or discharge of solid and liquid residues that exceed the exemption criteria (EU Directive 2013/59/Euratom; FANC-besluit van 3 maart, 2016).

In this paper the characterization of the natural radioactivity of the water collected from the Carboniferous Limestone Group at depths between 3200 m and 3600 m, of its salt content and the associated sediment fraction (which was created by the flocculation of iron oxides and hydroxides) is discussed. The radiological characterization of several geological samples obtained from different depths during the drilling is also addressed.

A similar study was performed by Regenspurg et al. (2014) but in their case the study was performed in a sedimentary basin hosting sandstone reservoir in the North German Basin and additionally they present results of the natural radionuclides accumulated in the filter residues (Regenspurg et al., 2014).

2. Samples

During production tests on the geothermal exploration well drilled at the Balmatt site, several formation water samples (called in the text 'original' formation water) were collected in order to analyze their natural radioactivity content and chemical composition. The water was pumped from a fractured limestone reservoir at depths between 3200 m and 3600 m below surface. An open basin was constructed near the drilling site to collect the formation water brought to the surface during the tests. With time, the suspended matter precipitates and creates a sediment layer at the bottom of the basin. Formation water and sediment samples from this basin were also analyzed (referenced in the text as basin water). This water has no fixed composition since water from rain fall may add to the basin.

During the drilling phase, rock samples were collected from different depths. 18 samples from 3100 m (the shale layer) to 3560 m (the reservoir layer) were selected for in-depth analyses.

3. Methods

During the drilling a spectral gamma-ray logging tool (HNGS-tool of Schlumberger) was used in order to identify the rock layers that were rich in radioactivity.

3.1. Determination of ^{238}U, ^{234}U, ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{210}Po

3.1.1. Water samples

For determination of ^{210}Pb, ^{210}Po and uranium isotopes, the water samples were prepared by $Fe(OH)_3$ co-precipitation using a 0.5 L sample. For the analysis of ^{210}Pb and ^{210}Po, the separation and measurement by liquid scintillation counting for ^{210}Pb and alpha-particle spectrometry for ^{210}Po were performed as described by Vajda et al. (1997). Uranium isotopes were separated using UTEVA® resin (Horwitz et al., 1992) and measured by alpha-particle spectrometry using PIPS semiconductor detectors and counting times of 250,000 s.

The radioactivity concentrations of the radium isotopes, ^{226}Ra and ^{228}Ra, were determined using gamma-ray spectrometry and also by using the radon emanation technique with Lucas cells for ^{226}Ra. Additionally, the ^{226}Ra activity concentration was determined by use of 3M Empore™ Radium RAD disks; the method is described in detail in Vasile et al. (2016) and Fons et al. (2016).

3.1.2. Sediment and rock samples

The massic activities of ^{238}U, ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{228}Th were determined using gamma-ray spectrometry, using a low-background shield and N-type detector with low-energy range. For several samples, the activity concentration of ^{226}Ra was also determined using the radon-emanation technique.

Additionally, for 5 rock samples, the method described by IAEA-AQ/34, (2014) for sequential separation of ^{210}Pb, ^{210}Po, ^{238}U and ^{234}U was applied using Sr and UTEVA® resins (IAEA-AQ/34, 2014). Polonium - 210 and uranium radioisotopes were measured by alpha-particle spectrometry using PIPS semiconductor detectors and counting times of 250,000 s. Lead-210 was measured using a Quantulus 1220™ with a measurement time of 300 min (Vajda et al., 1997). A blank was prepared in the same way as the real samples using the same reagents and deionized water.

4. Results and discussions

4.1. Formation water sample

The chemical composition of the 'original' formation water is presented in Table 1. This water has a high salt content with a chlorine concentration of 87 g L^{-1}. In geological waters with such high chlorine content, the concentration of radium isotopes is also expected to be high (Fisher, 1998; Albu et al., 1997; Degering et al., 2015). The activity concentrations of ^{226}Ra and ^{228}Ra determined by gamma-ray spectrometry were respectively 95 ± 18 Bq L^{-1} and 8.5 ± 0.7 Bq L^{-1} (uncertainty is expressed with a coverage factor $k = 2$), (Table 2). These uncertainties are determined based on an uncertainty budget, and the main contributions are coming from efficiency calibration, counting statistics of the sample, efficiency transfer, density corrections and summing corrections. A comparable activity concentration of ^{226}Ra was found in formation water of the Merksplas-Beerse geothermal site (~74 Bq L^{-1}), which geologically is retrieved from the same reservoir (Vandenberghe et al., 2000). The respective radioactivity concentrations of ^{210}Pb and ^{210}Po were found to have the much lower values of 0.687 ± 0.037 and 0.036 ± 0.007 Bq L^{-1}. Regenspurg et al. (2014) reported that in the geothermal fluid from the geothermal site in the North German Basin the measured activity concentrations of ^{226}Ra were as high as 72 Bq L^{-1}. Also in their case a high amount of chlorine was determined in the geothermal fluid (4.719 mM) and low values for the uranium and thorium activity concentrations were determined. In their paper, Regenspurg and co-authors explain that the low concentrations for uranium and thorium are due to the reducing conditions which limit their mobility. The formation water in the current study shows activity concentrations of U and Th isotopes to be low as well (see Table 2). The highest activity concentration of the thorium isotopes was found to be that for ^{228}Th (0.360 ± 0.070 Bq L^{-1}), which is to be expected since it

Table 1
Chemical composition of the formation water (the chemical analyses were done by SERVACO).

Sample	Description	Cl^- (mg·L^{-1})	HCO_3 (mg·L^{-1})	SO_4^{2-} (mg·L^{-1})	Ca (mg·L^{-1})	K (mg·L^{-1})	Mg (mg·L^{-1})	Na (mg·L^{-1})	pH	Conductivity (μS/cm)
'Original' formation water	First water brought to the surface	87840	141.6	255.8	7308	3258	483.4	37640	7.3	> 130000