Journal of Environmental Radioactivity 175-176 (2017) 34–38

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Radioactivity in wastes generated from shale gas exploration and production – North-Eastern Poland

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A R T I C L E I N F O

Article history:
Received 29 December 2016
Received in revised form
29 March 2017
Accepted 10 April 2017

Keywords:
Shale gas
Hydraulic fracturing
Radioactivity
NORM
Wastes
Poland

A B S T R A C T

In the present study, the K-40, U-238, Ra-226, Pb-210, Ra-228 and Th-228 activity concentrations were measured in 64 samples of wastes generated from shale gas exploration in North-Eastern Poland. The measured samples consist of drill cuttings, solid phase of waste drilling muds, fracking fluids, return fracking fluids and waste proppants. The measured activity concentrations in solid samples vary in a wide range from 116 to around 1100 Bq/kg for K-40, from 14 to 393 Bq/kg for U-238, from 15 to 415 Bq/kg for Ra-226, from 12 to 391 Bq/kg for Pb-210, from a few Bq/kg to 516 Bq/kg for Ra-228 and from a few Bq/kg to 515 Bq/kg for Th-228. Excluding the waste proppants, the measured activity concentrations in solid samples oscillate around their worldwide average values in soil. In the case of the waste proppants, the activity concentrations of radionuclides from uranium and thorium decay series are significantly elevated and equal to several hundreds of Bq/kg but it is connected with the mineralogical composition of proppants. The significant enhancement of Ra-226 and Ra-228 activity concentrations after fracking process was observed in the case of return fracking fluids, but the radium isotopes content in these fluids is comparable with that in waste waters from copper and coal mines in Poland.

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1. Introduction

Shale gas exploration and production, involving the extensive application of well-stimulation techniques (hydraulic fracturing) in horizontal wellbore sections to release gas from rock formations, poses a potential threat to the environment. The hydraulic fracturing process requires the use of very large amounts of water and chemicals necessary for the preparation of fracking fluids and other technological fluids used for borehole drilling (Javadpour, 2009; Karacan et al., 2011; Shabani et al., 2014). Consequently, in the course of this process, a large amount of waste in the form of return fluids and small amounts of proppants are generated. In addition, during the wellbore drilling, wastes are generated in the form of waste drilling fluid and drill cuttings (hard scales). The threat posed by shale gas exploration and production includes soil degradation, local pollution of the ground surface by fuels and technological liquids, contamination of surface water and groundwater, resulting from uncontrolled discharges of wastewater to them (Macuda, 2010).

Beside the above-mentioned hazards to the environment and humans, others may be attributable to the radiation hazard associated with radioactivity from natural radionuclides, widely occurring in the rocks of shale gas deposits and other formations. These radionuclides derive from natural radioactive decay series, mainly from two series: the uranium decay series (the primary radionuclide is U-238) and thorium decay series (the parent radionuclide is Th-232) and potassium K-40. From a radiation protection point of view, the most important radionuclides are radium Ra-226 (the half-live is $T_{1/2} = 1600$ years), radon Rn-222 ($T_{1/2} = 3.8$ days), lead Pb-210 ($T_{1/2} = 22.2$ years) and polonium Po-210 ($T_{1/2} = 138.4$ days) from the uranium series; from the thorium series, in turn, the most important radionuclides are Ra-228 ($T_{1/2} = 5.75$ years), thorium Th-228 ($T_{1/2} = 1.91$ years), Ra-224 ($T_{1/2} = 3.6$ day) and Rn-220 ($T_{1/2} = 56$ s). The third major source of radiation is the radioactive isotope of potassium, K-40, with a natural abundance of 0.0119%. Potassium occurs in significant quantities in the Earth's crust (about 2.5%). In the gas and oil mining industry, the radiation hazard associated with NORM mainly results from the migration of U-238 and Th-232 decay

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http://dx.doi.org/10.1016/j.jenvrad.2017.04.006
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products along with a mixture of oil, gas and water extracted from a wellbore. These products (mainly radium isotopes: Ra-226 and Ra-228) accumulate in sludge and hard scales. The average concentrations of radium isotopes in sludge and hard scales vary in a wide range from 100 Bq/kg to 15,000 kBq/kg. In general, radium concentrations are lower in sludge than in hard scales. The opposite applies to Pb-210, for which the concentration is relatively low in hard scales, but in the case of sludge the Pb-210 activity concentration may reach even 1000 kBq/kg and more (UNSCEAR, 2000; IAEA, 2003; UNSCEAR, 2008; OGP, 2008).

In the literature, there are few papers dealing with the problem of NORM in shale gas exploration and production. For example, the U.S. Geological Survey conducted a study of the activity concentrations of Ra-226 and Ra-228 in waters co-produced with oil and gas from Marcellus Shale. Radium concentrations vary in a range of 1.3 Bq/L to 590 Bq/L for Ra-226 and of 0.08 Bq/L to 44 Bq/L for Ra-228 (Rowan et al., 2011). In turn, the study carried out for radioactivity in samples taken from four reservoirs of fracking fluids located at the Barnett deposit, show that concentrations of natural radionuclides in analysed samples do not exceed 204 Bq/kg for K-40, 89 Bq/kg for Ra-226 and 26 Bq/kg for Ra-228 (Rich and Crosby, 2013).

In order to assess radioactive contamination of wastes resulting from shale gas, measurements of the natural radionuclides (K-40, U-238, Ra-226, Pb-210, Ra-228, Th-228) activity concentrations in wastes samples were performed. The samples of solid phase of waste drilling muds, drill cuttings, fracking fluids, return fracking fluids and waste proppants were collected from selected drilling rigs located in North-Eastern Poland (i.e. from Gdańsk Pomerania, across Mazovia to Lublin Region) in two of the most prospective basins of the Baltic Basin and Lublin Trough (Fig. 1). In this paper, the authors only provide information about the types of samples and assign sample codes due to information confidentiality.

2. Sampling and methods

Samples for analysis were taken from selected drilling rigs from August 2012 until June 2014. 64 samples consisting of fracking fluids, return fracking fluids, solid phase of waste drilling muds, waste proppants (these being proppants which flowed back from boreholes together with return fracking fluids after hydraulic fracturing) and drill cuttings were collected. Fluids were drawn directly from the sampling site to glass containers with a volume of 1 L, in turn, solid samples were packed into plastic bags. Samples of fracking fluid were collected before the injection into wellbores, in contrast to other types of samples collected after the processes of drilling and hydraulic fracturing. Samples of return fracking fluids were collected after the set of shale shakers, desanders and centrifuges which separate solid phase from return fracking fluids. After the transport to the laboratory, samples were sealed into beakers. There were two types of beakers used: the aluminium cylindrical beaker of the volume of 121 mL (used for solid samples)

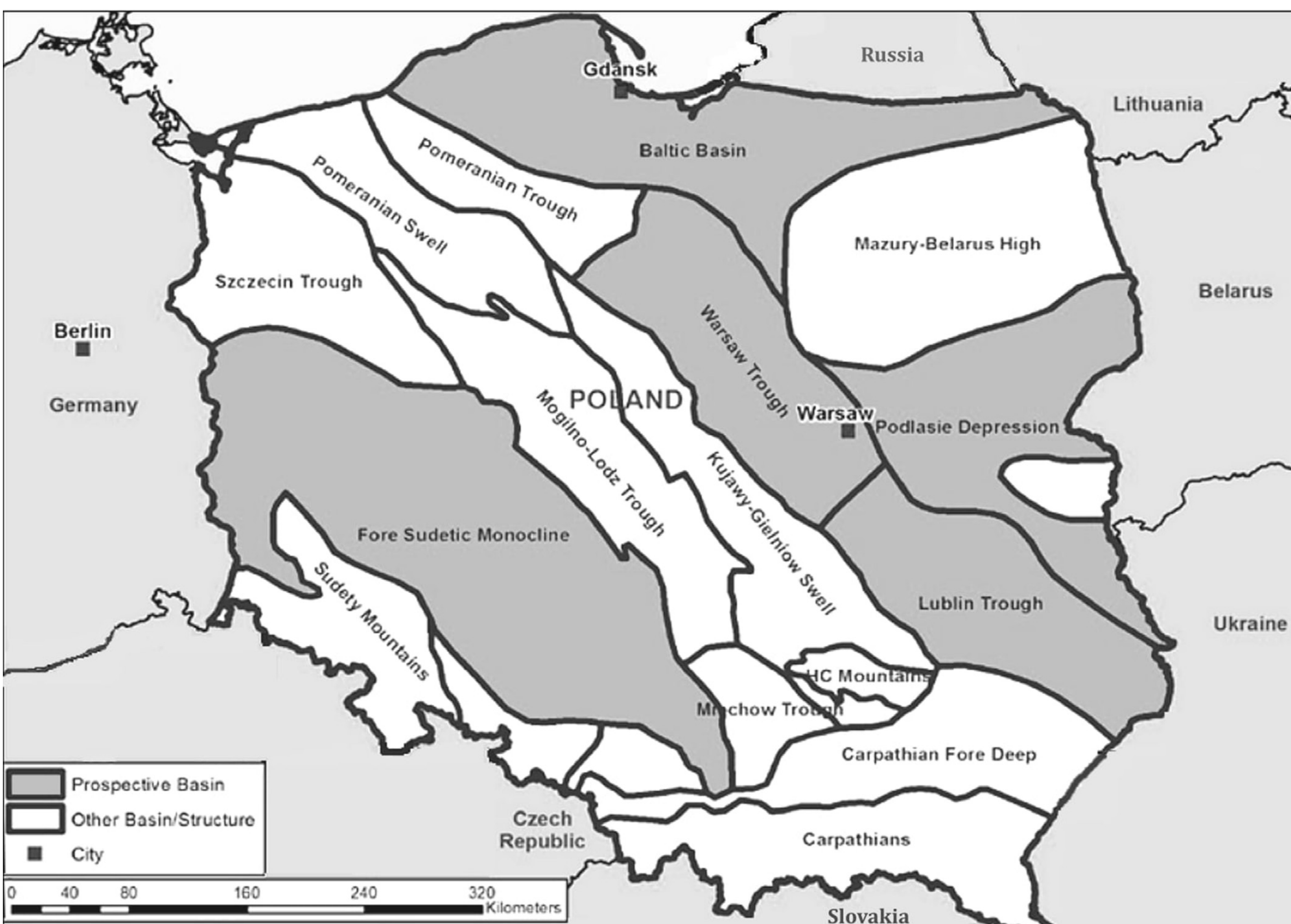

Fig. 1. Location of the most prospective shale basin in Poland (modified from San Leon Energy, 2013).

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