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Estimation and mapping of uranium content of geological units in France

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ABSTRACT

In France, natural radiation accounts for most of the population exposure to ionizing radiation. The Institute for Radiological Protection and Nuclear Safety (IRSN) carries out studies to evaluate the variability of natural radioactivity over the French territory. In this framework, the present study consisted in the evaluation of uranium concentrations in bedrocks. The objective was to provide estimate of uranium content of each geological unit defined in the geological map of France (1:1,000,000). The methodology was based on the interpretation of existing geochemical data (results of whole rock sample analysis) and the knowledge of petrology and lithology of the geological units, which allowed obtaining a first estimate of the uranium content of rocks. Then, this first estimate was improved thanks to some additional information. For example, some particular or regional sedimentary rocks which could present uranium contents higher than those generally observed for these lithologies, were identified. Moreover, databases on mining provided information on the location of uranium and coal/lignite mines and thus indicated the location of particular uranium-rich rocks. The geological units, defined from their boundaries extracted from the geological map of France (1:1,000,000), were finally classified into 5 categories based on their mean uranium content. The map obtained provided useful data for establishing the geogenic radon map of France, but also for mapping countrywide exposure to terrestrial radiation and for the evaluation of background levels of natural radioactivity used for impact assessment of anthropogenic activities.

1. Introduction

Natural radioactive sources account for most of the population exposure to ionizing radiation (Billon et al., 2005; Rapport IRSN/2015-00001). Over the past few years, several studies have been carried out by the Institute for Radiological Protection and Nuclear Safety (IRSN) to evaluate the variability of natural radioactivity over the French territory. The present study consisted in the evaluation of uranium concentrations in bedrocks. The objective was firstly to provide useful data for the geogenic radon potential map of France (Ielsch et al., 2010, 2014). Radon levels in the environment can show important spatial variations on a regional or local scale. It has been recognized that this lateral variability is primarily attributed to geological parameters which control the radon source-term and/or the radon migration in the ground (Appleton et al., 2015; Bossew, 2015; Drolet et al., 2013, 2014; García-Talavera et al., 2013; Gruber et al., 2013; Hahna et al., 2015; Ielsch et al., 2001, 2002, 2010; Kropat et al., 2014; Scheib et al., 2013; Smethurst et al., 2008; Tondeur et al., 2015; Zhukovsky et al., 2012). Therefore, the knowledge of uranium and/or radium contents in rocks is of primary importance to determine the radon source term.

Moreover, the present study was also carried out in order to provide complementary information in the framework of the evaluation of the exposure of the population to terrestrial radiation (Warnery et al., 2015; Rapport IRSN/2015-00001) and the evaluation of background levels of natural radioactivity for impact studies of anthropogenic activities or post-accidental situations.

This study was conducted to provide an estimate of uranium content of each geological unit defined in the geological map of France. Geology of France is complex and shows a great variety of lithologic units of magmatic (plutonic and volcanic), sedimentary

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and metamorphic origin, of variable age and chemistry. This complexity required the development of a method allowing classifying these units according to their lithological nature, to their chemical-mineralogical characteristics, to their petrogenetic origin and to the link between these parameters and the uranium content of these rocks. The work was particularly detailed for the magmatic plutonic rocks, very common in France and representing the major rock category with highly variable uranium contents and some of the highest uranium backgrounds, and which present a complex typology. As a result of its large ionic radius and high valency, uranium is not incorporated into the structure of the main rock forming silicates, which results in its incompatible behavior in silicate magmas. Consequently, during partial melting and crystal fractionation, uranium is preferentially fractionated into silicate melts (Cuney, 1982; Peiffert et al., 1994, 1996). The most felsic melts producing granites and rhyolites, tend to be the most enriched in uranium. Uranium deposits may derive directly and dominantly from magmatic processes (extreme fractional crystallization of peralkaline rocks or partial melting of U-rich crustal protoliths), but are generally related to uranium leaching in some specific granites which have higher uranium contents (Cuney and Friedrich, 1987; Moreau et al., 1966; Cuney, 2014).

Previous regional studies carried out in France, on the influence of uranium geochemistry of bedrocks and radon exhalation at the soil surface, conducted to propose a classification of lithologies of the study areas according to their uranium contents (Ielsch, 2000, Ielsch et al., 2001; Ielsch and Torrenti, 2004). In particular, the classification proposed for granitoids was based on the chemical affinity of magmatic rocks and their Th/U ratio and uranium content. This first approach provided a basis for the present study. The methodology used for this study is based on the interpretation of geological and geochemical data and on the expertise and the current knowledge gained in the field of geochemistry of uranium (Cuney, 2009, 2010, 2013, 2014; Cuney and Kyser, 2008; Leroy and George-Aniel, 1992; Madore et al., 2000; Pagel,1982; Pagel et al., 2005; Poty et al., 1986; Stussi and Cuney, 1993; Turpin et al., 1990; Vigneresse et al., 1989).

2. Material and method

This work is based on the compilation and analysis of available geological and geochemical data from maps, databases (results of rock samples analysis), results of research, studies etc. The knowledge of petrology, litho-geochemistry of geological units and of the behavior of uranium during rocks genesis also brought a necessary expertise to adjust the estimates. The methodology was divided in three steps: 1/first estimation of the uranium content of rocks from the interpretation of geochemical data (results of rock sample analysis) and the knowledge of petrology/litho-chemistry, 2/identification of particular rocks, which could present a uranium content higher than the one generally observed for these lithologies, and location of uranium and coal/lignite mines to adjust the first estimation 3/synthesis and final classification of the geological units according to their uranium content.

2.1. First estimation of the uranium contents of geological units

This step allowed to attribute a mean uranium content and a range of uranium contents to each geological unit and to propose a first classification of the rocks.

2.1.1. Geological map

The mapping was based on the geological map of France 1:1,000,000 (Chantraine, 2003; BRGM, vector). Some complementary information on geological units was provided by this map as lithology (main, secondary etc.), age, main geochemical affinity, litho-tectonic region. A number of inaccuracies on the attribution of the geochemical affinity of certain plutonic (granitoids in particular) and volcanic units with respect to the classification used in the present study was noticed and then corrected (section 2.1.3). The geological map supplied relatively detailed information for magmatic and metamorphic rocks, however this was not systematically the case for sedimentary units. For the latter, the units correspond to the grouping of several sub-units which may correspond to very different lithologies with different uranium contents. Data from larger scale geological maps were therefore used.

2.1.2. Analysis of geochemical databases

The first geochemical data used were results of the analysis of major and trace elements of whole rock samples directly available for the geological units. These results came from the Artemise database of CRPG-CREGU[1] and other compilations from previous research (Ielsch, 2000; Ielsch et al., 2001, 2002, 2004, 2010) that represented 5092 records of analyzed rock samples that could be precisely georeferenced (X,Y coordinates). The databases contained information on the location of rock samples, their texture and mineralogy, the corresponding lithology and geological unit, bibliographic data, and major and trace elements contents. Moreover, some additional data (hundreds of records) could not be georeferenced but could be associated to a geological unit according to the information available on the location, the lithology or the name of the unit. Direct geochemical data was available for most of plutonic, metamorphic and volcanic rocks of the Central Massif, the Armorican Massif, the Alps, the Pyrenees, Corsica and the Vosges. However, for the sedimentary rocks, only few results were available, essentially for detritic sedimentary rocks (sandstones, greywackes etc.). For carbonates (limestones, dolomites, malms), which constitute large parts of the territory (mostly the Paris basin, Aquitain Basin and South-East of France), the data was very rare.

The results compiled for each region allowed assigning a range of uranium contents and a mean uranium content (in ppm or mg/kg) to each geological unit of the geological map for which direct data was available. Outlying values which do not reflect the global trend of the other measurements were identified and studied carefully. Some values, judged as not representative of the unit, were removed.

The results were also used to create a synthetic database on uranium contents of the main lithologies observed in France. This database was used when no data, or not enough representative data (most sedimentary rocks, specially carbonates), was available for a given geological unit, to attribute a range of uranium contents and a mean uranium content by extrapolation of results obtained for similar lithologies of similar age observed in other regions. The database was in some cases completed by data from the literature particularly for sedimentary rocks for which uranium content was not often available (Clark et al., 1966; Rogers and Adams, 1969; Bhatia and Crook, 1986; Gascoyne, 1992).

2.1.3. Petrology and uranium geochemistry expertise

The exploitation of the geochemical data was based on the expertise and the current knowledge gained in the field of geochemistry of uranium. This expertise was used to propose a classification of geological units according to their lithology, petrology, geochemistry and the behavior of uranium during petrogenetic processes. The upper continental crust presents a

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