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Influence of soil conditions on the distribution coefficients of ^{226}Ra in natural soils

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HIGHLIGHTS

- The operationally defined method for determining the K_d value must depend on its potential use.
- The K_d value is dependent on the structural characteristics of the soil.
- For K_d determination, the incubation time should be longer than 7 days and the moisture levels about 50% saturation.
- The K_d value of small-sized edaphic particles depends on the speed of centrifugation.

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ABSTRACT

In order to clarify some of the assumptions and approximations about the use of the distribution coefficient K_d for ^{226}Ra in soils, a systematic study has been performed using centrifugation to extract the soil solution. The separated fractions of the soil solution have different kinetics with respect to the sorption process in the soil, which may in turn condition the final chemical composition and even the speciation of the radionuclides in solution. In the experimental design of this study three factors were considered: the moisture level in the incubation process, incubation time and the speed of centrifugation. Also, three levels were chosen for each factor. In order to analyze the influence of the structural characteristics of the soil, this study was performed with three textural fractions: coarse sand, fine sand, and silt and clay, obtained from an only soil. Also, the soil was naturally enriched with radionuclides of the ^{238}U series. An analysis of variance (ANOVA) was performed in order to assess the influence of the factors studied on the distribution coefficient of ^{226}Ra. The results indicate that different behaviors can be observed depending on the structural characteristic of the soil. In the case of particle size, the soil with the largest grain size showed that the incubation process parameters influence the equilibrium level achieved, while in the case of the smallest edaphic particles, radium is not homogeneously distributed in the soil solution and the K_d value is dependent on the speed of centrifugation.

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1. Introduction

The interest in risk assessment models concerning the distribution and transport of radionuclides in the environment originated long time ago, from 60′ of previous century, due to open atmospheric nuclear probes, and has significantly increased since the Fukushima nuclear disaster. This interest includes not only artificial radioactivity (Fujiwara, 2016; Lee et al., 2017; Povinec et al., 2017) but also naturally occurring radionuclides whose concentration in the biosphere has been altered by industrial activities (NORM, TENORM) (Eitrheim et al., 2016; Michalik, 2017). Hence, the application of these models using reliable data or parameters is of particular importance in order to minimize uncertain results.

Food consumption is a common way in which humans can become exposed to radionuclides. The radionuclides can enter into the food chain through the soil-plant-animal continuum or through irrigation water or direct consumption (Vandenhove et al., 2007). Therefore, the soil-to-plant transfer process is expected to largely determine the radiological risk associated with the food chain. In order to evaluate this risk, it is not only important to determine the

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behavior of the radionuclides in the transfer path, but also to understand the mechanisms and processes that determine the availability and incorporation of radionuclides in plants (Maity et al., 2013).

The transfer of radionuclides from soil to plants is a very complex process and is greatly influenced by the physicochemical properties of the soil itself (Absalom et al., 2001; Blanco Rodríguez et al., 2017; Hegazy et al., 2013; Roivainen et al., 2012; Vandenhove and Van Hees, 2007). Soil can be considered as a dynamic system that experiences a continuous exchange of water and other chemicals that disturb the natural tendency to attain thermodynamic equilibrium. This process involves both the hydrodynamic characteristics of the soil and its capacity to mitigate the chemical changes that are induced. Based on this premise it can be assumed that soil is able to naturally buffer the impact of external influences, and therefore able to regulate the tendency to come to equilibrium. It can be considered that the transfer process begins inside the soil, acting as a repository of radionuclides and heavy metals. Likewise, it is in soil where the processes that establish the availability of these elements are regulated (Higley and Bytwerk, 2007). In order to understand how soil can behave, it is necessary to determine its hydrodynamic, physicochemical, and chemical properties, as well as the effects of the surrounding biota (Maity et al., 2013).

The availability of a radionuclide for its assimilation by a plant strongly depends on the absorption-desorption process associated with the surface reactivity of the solid phase in contact with the soil solution, which is described by means of the distribution coefficient K_d ($L \cdot kg^{-1}$). When applied to radionuclides, K_d represents the partition of the radionuclide between the solid phase in the soil (C_s, $Bq \cdot kg^{-1}$) and its solution (C_{ss}, $Bq \cdot L^{-1}$).

Although the utility of K_d is commonly accepted and is frequently used in risk assessment models as an input parameter, K_d values must be carefully chosen according to specific aims, owing to the many different K_d values that can be found in the databases. The EPA and IAEA recommend that site-specific measurements should be taken when possible; otherwise the use of the geometric mean and geometric standard deviation from published database is suggested according to the classification of soils in four groups (IAEA, 2010).

There are several reasons that explain the broad range of K_d values observed, since this parameter is commonly described as representing processes related to equilibrium, that is, when the rate of sorption and desorption is the same. This condition requires that sorption processes are reversible and rapid compared with the flow velocity, which is not easily confirmed in a real scenario (Ireson and Butler, 2009). The variability of K_d values also occurs because K_d is related to several soil properties such as biological activity, redox potential, soil structure, mineral forms, organic matter content, and pH, among others (Hormann and Fischer, 2013; Kumar et al., 2015; Maity et al., 2013; Sheppard, 2011; Sohlenius et al., 2013). Additionally, K_d values are also influenced by the method used to quantify them, which can be one of the many different types of approaches used (Blattner et al., 2000; Schlotter et al., 2012). The variations that exist among these methods often make it difficult to compare the different K_d values calculated (Vandenhove et al., 2009).

In the present study, different experiments were designed to determine the available fraction of ^{226}Ra in soils collected in a mineralized uranium zone located in the west of Spain. In the experimental design different approaches were considered to define the soil solution. These approaches aimed to reduce the large amount of uncertainty associated with the evaluation of the distribution coefficient K_d.

A centrifugation method was tested for extracting a soil solution to be used for further characterization (Geibe et al., 2006; Reatto et al., 2008). The centrifugation of a sample of previously moistened soil generates the soil solution which contains contents that originate from the soil sample. The origin of the water in the soil, associated with the different forces that retain it (such as gravity or capillary or hygroscopic action), depends on the effectiveness of the suction applied to the soil core through the water retention characteristic curve (WRC) (Dexter et al., 2008; Lu, 2016). The hypothesis presented here is that the water origin in the soil will reflect different distribution coefficients, which will affect the definition of radionuclide availability. The speciation of the radionuclides in the soil solution can also be governed by this hydrological origin.

The experiments carried out in this study include various types of initial conditions involving moisture, incubation time, and the effective suction applied (centrifugation speed) to the soil specimen. In addition, the analysis also includes three physical fractions of the soil based on particle size, a parameter that is well recognized as having an important influence on the retention/liberation of radionuclides (Geris et al., 2015; Hormann and Fischer, 2013).

2. Materials and methods

2.1. Sampling and sample preparation

The main soil sample was collected from an area with natural uranium mineralization located in the west of Spain, in the southwest region of Castile and Leon (40°43′N; 6°42′E). The sample, taken from uncultivated area, represents a well-consolidated soil, which is of interest to the proposed study.

In the laboratory, and with the purpose to have samples of different textures, the soil sample was mechanically fractioned by dry sieving into three textural fractions: coarse sand, S1 (0.5–2 mm); fine sand, S2 (0.067–0.5 mm); and silt and clay, S3 (<0.067 mm). In order to ensure accurate separation by grain size, particle aggregates were previously broken up. To do this, a new device consisting of a whetstone lined with rubber located in the orbital shaker (Heidolph, Mod. Rotamax 120) was designed (Prieto et al., 2015). The ^{226}Ra activity concentration ($Bq \cdot kg^{-1}$) in the different fractions were: 86.4 ± 3.1 for S1, 58.1 ± 2.2 for S2, and 98.1 ± 3.5 for S3.

Also, each of the three soil fractions was chemically and physically characterized (see Table SM1 in Supplementary Material).

2.2. Experimental design

The first step in determining the distribution coefficient was the incubation process. For this, the samples (S1, S2 and S3) were kept at a specific degree of saturation during a certain amount of time in order to analyze the effect of the contact time with respect to the true solid-liquid equilibrium state. It was expected that the concentration of radionuclides in the liquid fraction of the soil depended on the moisture level during the incubation period. Also, the incubation time had to be adjusted in order to ensure that equilibrium was achieved. In order to study the possible influence of these two factors on the distribution coefficient values, three conditions were selected for each factor: degree of saturation (moisture level) of 50%, 75% and 100%, and incubation periods of 1, 7 and 30 days.

The soil solution extracts were obtained by centrifugation using a high performance centrifuge and specially designed devices to adapt the rotors to a fixed angle, specific to the requirements of the experiment (Medeiros et al., 2014). For this, an ultracentrifuge (Beckman-Coulter, Avanti 25I) and a fixed-angle rotor (Beckman-Coulter, JLA 16.250) were used.

In the method used, a direct relationship between the speed of

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