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Research Article

Soil organic matter and carbonates facing radioactive metal pollution

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Abstract

Pollution from uranium, thorium, and radium poses significant environmental and public health risks. This study examines two regions with distinct soil types: carbonated and marshy soils. Soil samples were selected based on carbonate (28.83% and 9.79%) and organic matter content (22.6% and 1.8%). The research aims to evaluate the role of carbonates and organic matter in retaining radioactive elements and to identify which metal (thorium, uranium, or radium) is most retained. The methodology included physicochemical soil analysis and sorption experiments, varying pH and organic carbon (humic acid and biochar). Results show that thorium has the highest adsorption affinity due to the stability of its solid compounds. Uranium and radium exhibited lower retention, with behavior varying by soil type. The study also compared recalcitrant and active organic matter, revealing that both carbonates and organic matter significantly enhance soil depollution. However, acidic organic materials, unlike basic ones, do not improve metal retention. Instead, they promote metal breakdown, increasing bioavailability and plant uptake. These findings suggest that biochar application could effectively remediate soils contaminated with radioactive elements, offering a sustainable agricultural solution. This approach reduces pollution while improving soil health and crop safety.

Keywords: Pollution, Biochar, Radioactive, Retention, Carbonates

Introduction

The sources of uranium, thorium, and radium pollution include both natural sources, such as rocks and mineral deposits, and anthropogenic activities, such as mining, nuclear industry operations, radioactive waste management, and occupational accidents. Given that the Djelfa region houses a nuclear facility, this study is particularly relevant for evaluating potential risks in the event of an explosion or radioactive release (Gundersen et al.,2010).

The accumulation of these pollutants in the environment is a growing concern, exacerbated by human activities. The widespread and persistent nature of this pollution amplifies its adverse effects. Uranium, thorium, and radium, particularly their most abundant isotopes (^{238}U, ^{232}Th, and ^{226}Ra), pose significant risks to human and environmental health due to their radioactive properties. These elements emit alpha, beta, and gamma radiation, which can damage living tissues and disrupt ecosystems.

The processes of sorption and desorption of metals in soils play a critical role in regulating their environmental behavior. Sorption refers to the attachment of metal ions, such as uranium, thorium, and radium, to solid soil particles, thereby reducing their mobility. This phenomenon is influenced by various physicochemical factors, including soil pH, redox potential, and mineral composition (Trang, 2010). These mechanisms directly affect the availability of these metals in the environment and their potential to migrate to other compartments, such as groundwater or food chains. The primary objective of this study is to evaluate the retention capacity of uranium, thorium, and radium in soils, with a focus on their interactions with organic matter and carbonates, in the Djelfa region.

Materials and methods

Study area

The wilaya of Djelfa is in the center of Algeria and is bordered to the north by the Atlas Tellien, to the south by the Atlas Saharien, and to the east by the Atlas Presaharien region. It is located around 300 kilometers south of the capital and has an area of 32,256,35 square kilometers. Its latitude ranges from 33° to 35° North, while its longitude varies from 2° to 5° East. The wilaya is distinguished by its highest point, which is 1613 meters above sea level, and its lowest point, which is only 150 meters above sea level (Ouanouki,2012). The échantillonnage was carried out using a stratified method. The first zone dealt with calcareous soils (Fig.1). Following the prelevé and analysis of 20 calcaires sol échantillons, we chose the one with the highest carbonate content (28.82%) and the one with the lowest (9.7%). Next, we looked at the second zone, which is made up of marécageux soils, one of which has a higher percentage of organic matter (22,6 %) and another that is lower (1,8%).

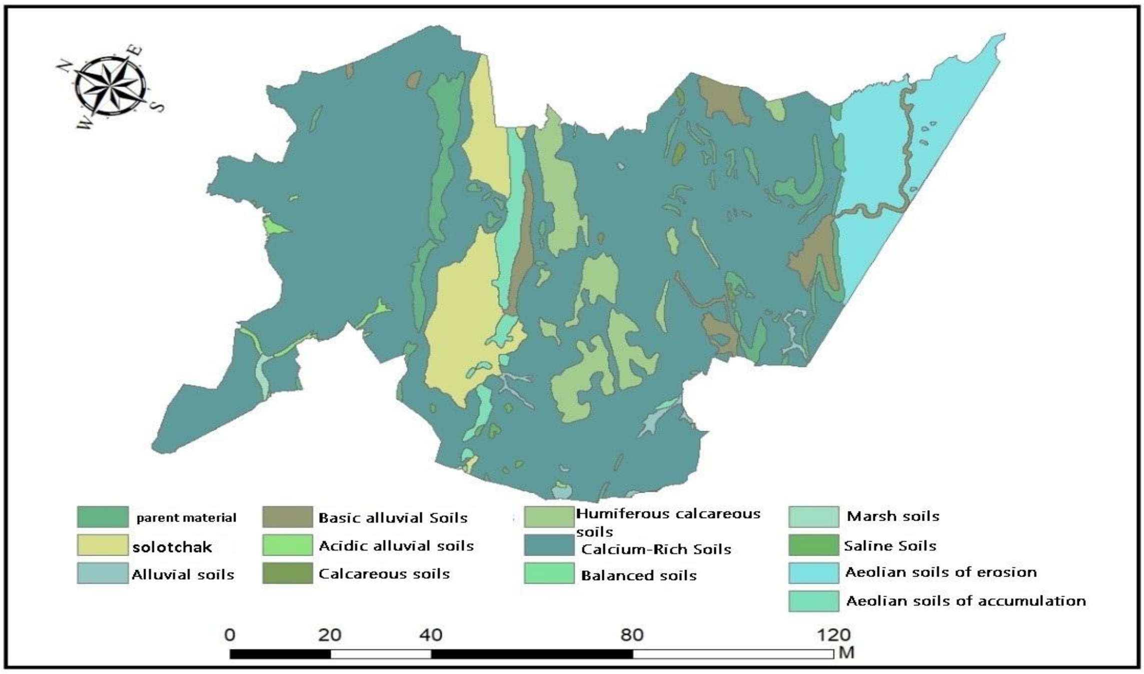

Figure 1. Pédologique Carte, Djelfa Region .

Physicochemical analysis of soils

The techniques described by Aubert (1978). The pH (H_2O) was measured using a pH meter with a volume-to-solids ratio of 1:5, while electrical conductivity was determined using a conductimeter with the same volume-to-solids ratio. Granulometric analysis was performed using the sedimentation method with Robinson’s pipette. Organic matter content was quantified using the Walkley-Black method (1934), which involves oxidizing organic matter with chromic acid in the presence of sulfuric acid, followed by titration of the excess chromic acid with a ferrous ammonium sulfate solution. The amount of oxidized organic matter is proportional to the reduction of dichromate. Additionally, cation exchange capacity (CEC) was measured using ammonium acetate at pH 7 (Metson method), and carbonate content was determined using the Bernard calcimeter (Bernard,1904).

Adsorption Analysis

Adsorption is the process by which molecules or ions from a liquid phase accumulate on the surface of a solid phase (Hanjing et al., 2023). Studies have shown that soils naturally adsorb substances, with their efficiency depending on the prevailing chemical and physical conditions (Rassaei, 2023). The adsorption study was conducted using 0.5 g of soil and 50 mL of a metal solution (U, Th, or Ra) at a concentration of 1 mg/L. The mixture was shaken at 350 revolutions per minute (rpm) for one hour (except for kinetic experiments), centrifuged at 3500 rpm, and filtered through 0.45 µm filters. All experiments were performed at a constant temperature of 23°C. The retention rate was calculated using the following equation (Zerroug, 2024):

$$R(\%) = ((C0–Cf) / C0) \times 100$$

With C_0 is initial concentration of polluant metal (mg/L) and Cf is final chromium concentration (mg/L)

Several parameters affect the adsorption process of metal ions on substrates. The pH plays a crucial role by influencing the speciation of metals, with higher pH values promoting the formation of oxyhydroxides and enhancing immobilization. Temperature, organic matter impacts the mobility of metals and alters the adsorption process.

Results

Table 1 summarizes the physicochemical parameters studied in the soils.

Table 1.The physicochemical properties of the four soil samples.

Sample of calcareous soil type	Sample of marsh soil type
Soil1: The most calcareous soil pH:8.22 C.E (µs/cm):1.53 C.E.C (cmol+/kg) :10.02 Carbonates % :28.83 Organic matter % :1.7 Grain size: Sandy clay	**Soil 3:** The most organic matter content pH:6.8 C.E (µs/cm):194 C.E.C (cmol+/kg):11.33 Carbonates % :8.04 Organic matter % :22.6 Grain size: Clay loam
Soil 2: The least calcareous soil pH:7.43 C.E (µs/cm):136 C.E.C (cmol+/kg):11.4 Carbonates % :9.79 Organic matter % : 1.9 Grain size: Sandy clay	**Soil 4:** The Least organic matter content pH: 7.11 C.E (µs/cm):189 C.E.C (cmol+/kg):9.80 Carbonates %: 9.81 Organic matter %: 1.8 Grain size: Sandy clay

It is observed that soil 1 has the highest pH and the highest carbonate content compared to soil 3. Additionally, soil 3 has the highest organic matter content and the lowest pH compared to soil 4.

The sorption of thorium, radium, and uranium:

The sorption of uranium, radium, and thorium: The purpose of this section is to determine the factors influencing the rate at which radioactive metals (^{232}Th, ^{226}Ra, and ^{238}U) are absorbed by the sols. The experiments will involve exposing the soils to the three pollutants and tracking changes in their concentrations over time. To better understand the interactions between radioactive metals and various soil matrices, the results obtained will enable the identification of the major chemical mechanism of sorption.

Kinetic study

The following figures illustrate the adsorption kinetics of the studied metals, highlighting the evolution of their retention over time.

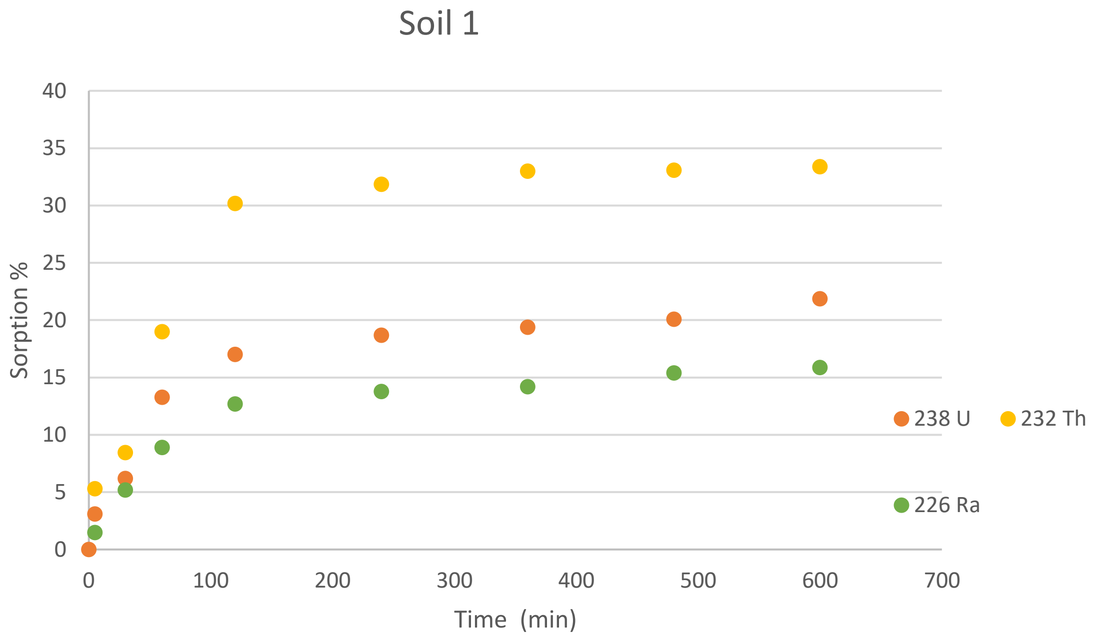

Figure 2 . Adsorption kinetics of ^{232}Th,^{238}U, and ^{226}R for soil 1.

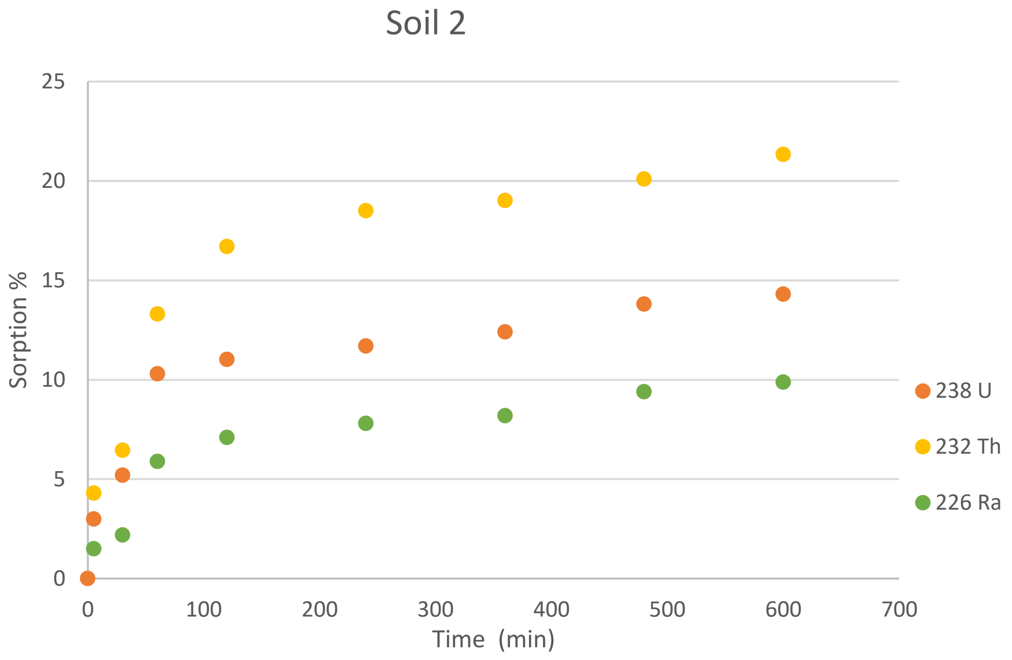

Figure 3 . Adsorption kinetics of ^{232}Th,^{238}U,and ^{226}R for soil 2.

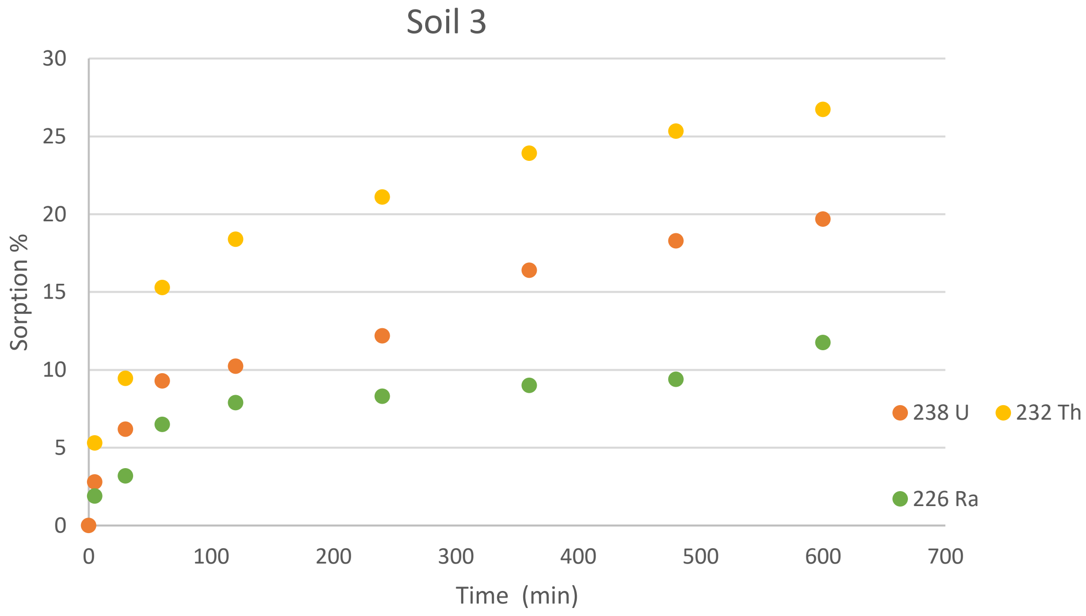

Figure 4 . Adsorption kinetics of ^{232}Th,^{238}U and ^{226}R for soil 3.

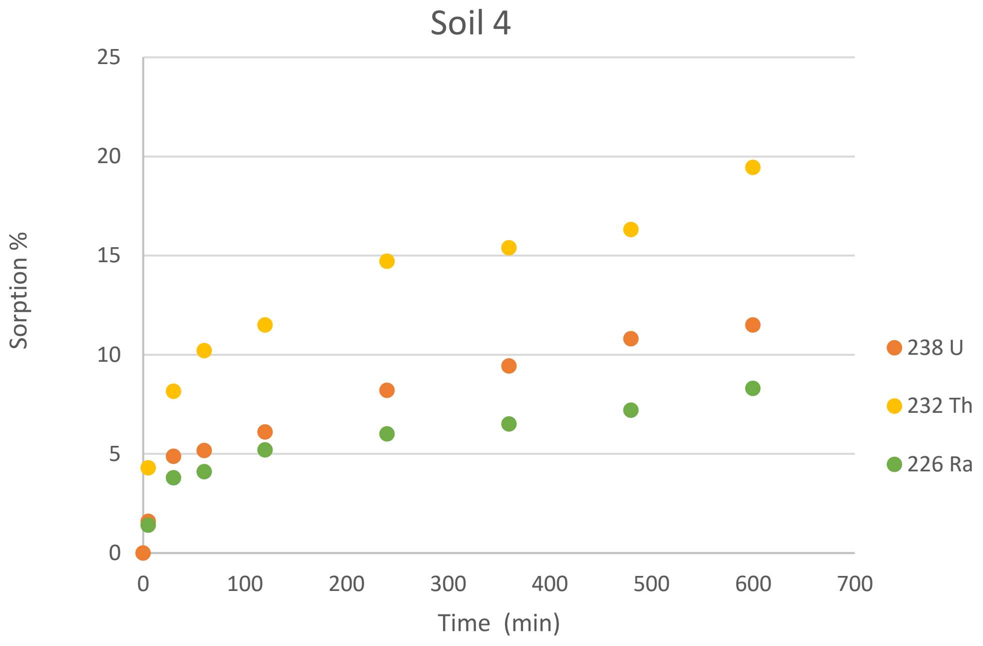

Figure 5. Adsorption kinetics of ^{232}Th,^{238}U,and ^{226}R for soil 4.

The effect of pH on metal retention

To investigate how pH affects retention, we have, We used soil 4 as the standard soil, which has the lowest content of organic matter and carbonates. Its pH was adjusted in the laboratory from 2 to 12 using a 0.1 M CaCO3 solution and a 0.1 M sulfuric acid (H_2SO_4) solution.

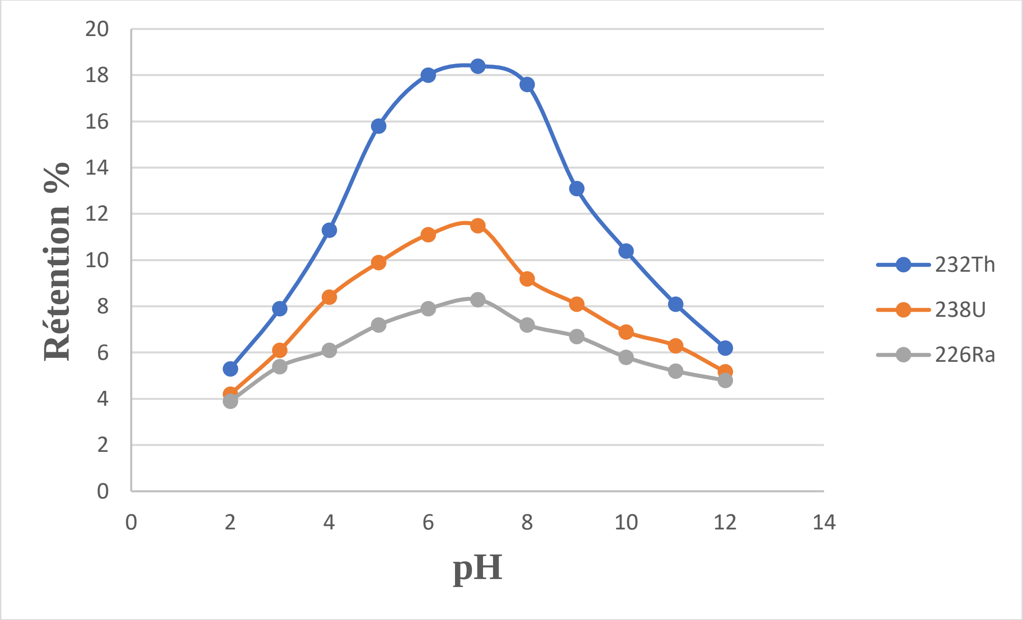

Figure 6. Variation of Thorium, Uranium, and Radium Retention as a Function of pH.

Effect of organic carbon on metal retention

We examine the impact of organic matter on retention in this section. For this, we used two types of organic matter that contained acidic groups: (humic acid), in varying concentrations from 2% to 10%, and an organic matter rich in carboxylic groups(Biochar) also applied in concentrations ranging from 2% to 10% . Sol 4 was utilized as the normal sol. However, prior to the experiment, the naturally occurring organic matter in this soil was removed by chemical oxydation with hydrogen peroxide in order to obtain a reference environment free of organic matter.

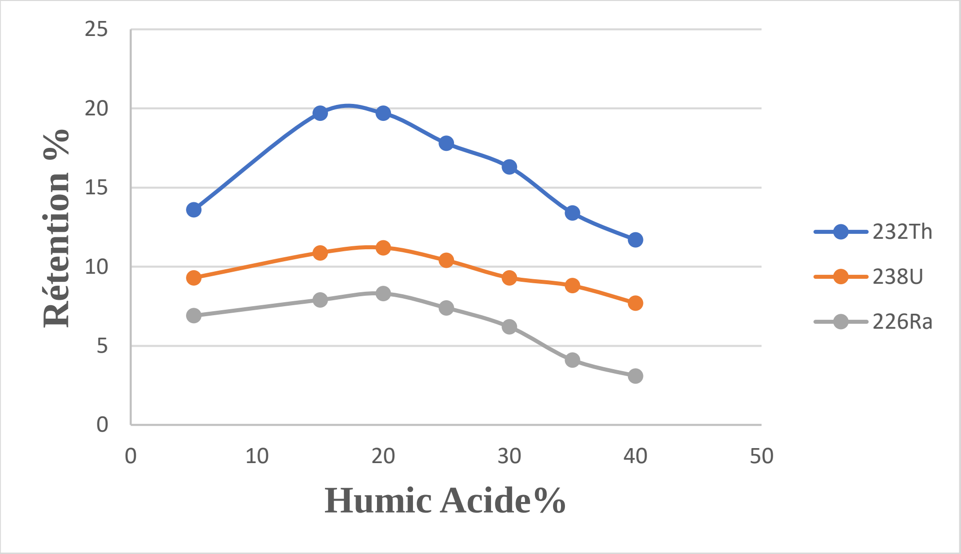

Figure 7. Variation of Thorium, Uranium, and Radium Retention as a Function of Humic Acid Concentration %

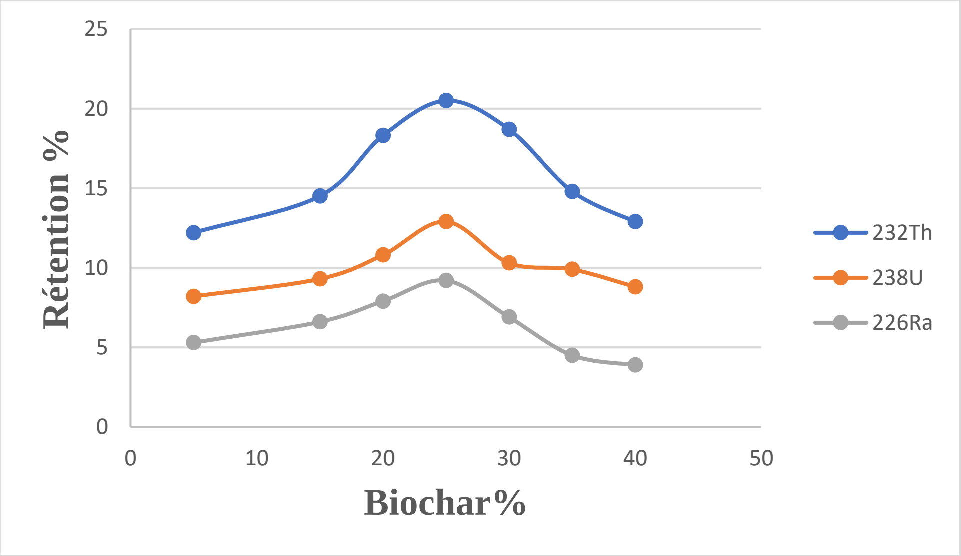

Figure 8. Variation of Thorium, Uranium and Radium Retention as a Function of Biochar Concentration %

Discussion

The sorption study, presented in Figures 2, 3, 4, and 5, was conducted in two distinct steps. The first step involves the rapid occupation of easily accessible fixation sites, which occurs almost immediately upon agitation of the soil suspension. This phase is characterized by the fast adsorption of radioactive elements. The second, more gradual step involves the occupation of less accessible fixation sites in the soil, which take longer to become saturated. As demonstrated by Record (1997), equilibrium is typically reached within approximately two hours, indicating that the majority of sorption sites are occupied and the sorption dynamics stabilize.

The results shown in Figures 2, 3, 4, and 5 reveal that the adsorption capacity of the three pollutants (Ra, U, and Th) increases over time. After approximately two hours of contact for all soil types, sorption equilibrium is achieved, marking the point at which the concentrations of Ra, U, and Th stabilize. Among the studied soils, Soil 1 exhibited the highest sorption percentage and maximum adsorption capacity for the three radioactive elements. This was followed by Soil 3 and then Soil 2, while Soil 4 demonstrated the lowest adsorption capacity. The sorption kinetics of the three metal elements on the four soils indicate a strong affinity for radioactive metals in carbonated soil. Notably, thorium (Th) showed the highest retention affinity on the most carbonate-rich soil (Soil 1) and on the soil with the highest organic matter content (Soil 3), compared to uranium (U) and radium (Ra).

The differences in adsorption behavior among thorium (Th), radium (Ra), and uranium (U) can be attributed to their varying affinities for carbonated structures. Thorium forms stable compounds with carbonate ions in solution, making it more readily adsorbed than uranium and radium. Although radium (^{226}Ra) can interact with carbonates, it is generally less favored due to its lower solubility and reduced ability to form carbonate complexes. In contrast, uranium (^{238}U) exhibits a stronger affinity for carbonates, as it readily forms complexes with carbonate ions (CO_3^{2-}), facilitating its incorporation into carbonated structures. Uranium is also more soluble in oxidizing environments and is frequently found in carbonated solutions as carbonate complexes, unlike radium (IAEA, 2001).

Figure 6 shows an increase in retention on a pH range of 2 to 7, with the ideal range between pH 6 and 7. Beyond this range, between pH 7 and pH 12, retention decreases. For the three metals under study, this trend is evident, with thorium showing a particularly notable retention

An improvement in thorium (^{232}Th) retention in the pH range of 2 to 7, with the ideal pH range being between 6 and 7. Beyond this range, retention decreases between pH 7 and pH 12. This trend is evident for the three metal elements under study; thorium exhibits particularly notable retention across the pH range because it may bind more steadily to carbonates in alkaline settings. In environments with high pH, thorium forms compounds with carbonates, such as $Th(CO_3)_4^{2-}$. These complexes, which are more stable than those of radium, make thorium less soluble and encourage its retention on solid soil surfaces, especially in the pH range of 6 to 7. However, these compounds may become more soluble above pH 7, which would decrease thorium retention(Echevarria et al.,2001; Melson et al.,2012).

Regarding uranium (^{238}U), because of its solubility in oxidant environments and its ability to form stable complexes with carbonate ions, such as the complex $(UO_2)_2CO_3^{2-}$, uranium (238U) tends to bind strongly to carbonates in alkaline settings. This combination makes uranium more soluble, which increases its mobility in the environment, especially at higher pH values (above 7). Because of its ability to combine with carbonates, uranium is particularly sensitive to changes in the pH of the soil, with retention being enhanced at moderate pH values and decreased at higher pH values due to the increased solubility of uranium-carbonate complexes (Bish et al .,2005). The breakdown of carbonates into bicarbonates decreases the formation of complexes with uranium at low pH values. This makes uranium more soluble and reduces its retention on carbonated soil surfaces compared to thorium (Harmsen et al. ,1980)(Meslon et al.,2012).

Although the complexation is typically less marked, the ability of radium (^{226}Ra) to form complexes with carbonates is comparable to uranium. Indeed, under alkaline conditions, radium can form complexes with carbonates, although these complexes are often less stable than those of uranium. Although radium can form complexes with carbonates in the form of $RaCO_3^+$, they are less stable and more prone to dissociation than those formed with uranium. A high pH can cause radium to form complexes with carbonates, although the solubility of these complexes can also reduce radium's retention.

Figure 7 shows the retention of thorium, uranium, and radium as a function of the concentration in humic acid (%), ranging from 5% to 40%. It can be observed that retention is moderate up to the peak point at approximately 20%, then gradually decreases. The initial retention observed may be attributed to other components in the soil. At this stage, the soil is moderately alkaline until it reaches saturation with humic acid, which leads to its acidification and promotes the dissolution of metals. In addition, compared to uranium and radium, thorium exhibits the highest retention rate.

Figure 8 shows the retention of thorium, uranium, and radium as a function of biochar concentration (%) ranging from 5% to 40%. It is observed that retention increases from ٥% to ٢٥% of biochar, then decreases between 25% and ٤٠% for all metals. Additionally, thorium exhibits the highest retention rate compared to uranium and radium.

the organic matter plays a crucial role in the retention of metals. The sort of organic matter present, whether it be basic(recalcitrant organic matter) or acidic (active organic matter) determines the ability to retain heavy metals (Deschamps et al., 2006). Because of their ability to attach to metal ions and create soluble complexes, organic acid matter, such as fulvic and humic acids, promotes the solubility of metals**.** This increases their availability to plants and other organisms by facilitating their mobility in the soil. On the other hand, basic organic matter, such as organic carbonates, tends to encourage metal retention. This is because of its ability to neutralize metal ions and create more stable and less soluble complexes, which decreases their availability for migration in the soil (Guo et al.,2008).

Additionally, it has been demonstrated that as soils become saturated with organic matter, the pH tends to increase, leading to a more alkaline environment. This results in a decrease in metal retention, suggesting that pH plays a significant role in a soil's ability to retain metals. This phenomenon is likely due to competition between anionic species, such as hydroxyl ions (OH^{-}), and metal ions for adsorption sites (Murphy et al., 1999; Strawn, 2021). As discussed in the previous section on pH effects, at higher pH values, hydroxyl ions may bind more strongly to available adsorption sites, reducing the soil's affinity for heavy metals. Thus, soil type and composition are critical factors influencing pollutant retention (Tao et al., 2023 ;Yu et al.,2023). Organic matter in the soil facilitates the adsorption of heavy metals by forming stable complexes, thereby limiting their mobility and bioavailability. However, the retention of metals depends on factors such as the type of metal, soil pH, and the proportion of organic matter. In alkaline environments, organic matter promotes metal precipitation and stabilization, whereas in acidic environments, metals may become more soluble, increasing their availability for uptake by plants and aquatic organisms. Consequently, organic matter plays a vital role in regulating heavy metal behavior in soils, influencing both their retention and mobility in response to environmental conditions (Villar et al., 2001).

A comparison of retention results between the carbonate phase and recalcitrant organic matter reveals similar retention percentages for the three heavy metals studied. This suggests that these two soil fractions play comparable roles in the adsorption and stabilization of contaminants, although their interaction mechanisms with metals may differ.

Conclusion

The results demonstrate that retention and decontamination depend on both the adsorbate-adsorbent interaction and the type of adsorbent, with the bonding mechanism playing a crucial role. In this study, the adsorbents are carbonates and organic matter, and the adsorbates are thorium, uranium, and radium. Thorium exhibits the highest retention due to its strong affinity for both organic matter and carbonates. While retention is generally high in organic matter, it is particularly significant in basic organic matter, which forms more stable complexes. However, the degradation of organic matter can produce stronger acids, such as humic acid, which may reduce metal retention. The three radioactive elements (uranium, radium, and thorium) interact with carbonates differently, depending on pH and the availability of carbonate ions. At lower pH values, metal-carbonate complexes are more stable, promoting retention. At higher pH values, the solubility of these complexes increases, potentially reducing the retention of radioactive elements in soils. Thorium, however, exhibits stronger retention than uranium and radium due to the greater stability of its carbonate complexes under typical soil pH conditions.

Thorium (^{232}Th) is favored by carbonates and organic matter due to its low solubility and strong affinity for solid surfaces. As a Th^{4+} ion, it forms stable complexes with organic acids and carbonates, reducing its mobility in the aqueous phase. In contrast, uranium forms more soluble complexes (e.g., UO_2^{2+}), which decrease its retention, while radium, being more soluble, is less retained than thorium.

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