

Fate of Cesium and Strontium in Soil-to-Plant System (Overview)

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Abstract: Radioactive contaminated soils can present an immediate hazard to human health as well as a chronic hazard to the environment. Knowledge of soil-specific contaminant sorption-desorption characteristics is useful for the simulation and prediction of contaminant fate, transport and diffusion in soil-water-plant systems.

The objectives of this overview paper were to explore the relative importance of soil type and some soil properties in the assessment of their environmental significance. And to explore the sorption of Cs and Sr on soils developed under dry climatic conditions.

In general, the behaviour of radionuclides deposited on the soil surface is controlled by their retention time in the top soil. For example, the slow movement and low migration rate of radionuclides in the soil results in a long radionuclide residence time at the plant-rooting zone and, hence, increases the probability of radionuclide uptake by the plant. Cesium (^{137}Cs) and strontium (^{90}Sr) are of special concern for soil contamination during a long half-life. The sorption of Cs and Sr in soils may be affected by a number of factors, such as the quantity and quality of the clay fraction, pH, O.M, EC, SAR and ECC.

Keywords: Sorption: soil retention, Metal-Kd, isotherm models, migration.

1 Introduction

The problem of restoring land contaminated by radioactive fallout has become more vital with the appearance of nuclear explosions and accidental releases of radionuclides. Radionuclides reach humans and other living organisms by a number of routes after they are released into the environment from nuclear installations.

Soils can serve as drains for radioactive pollution depending on their nature. The development of pollution varies with the different properties of the soil, especially those involving soil retention characteristics and the mobility of pollutants. Soil sorption-desorption is extremely important for environmental and agricultural concerns. Radioactive contaminated soils can present an immediate hazard to human health as well as a chronic hazard to the environment [1].

Cesium (^{137}Cs) and strontium (^{90}Sr) are of special concern for soil contamination during a long half-life (30 years for Cs and 28 years for Sr). The sorption and transfer of radionuclides in soil could be influenced by many factors, e.g., pH, chemical species and the nature of radionuclides in the soil solution as well as chemical and physical nature

of the soil [2].

Sorption-desorption behavior at the interface between the particulate matter and the soil solution is extremely important for transport, bioavailability and other inorganic materials in soils [3]. Soil has the capacity to limit the movement of Cs and Sr in the environment by controlling the simultaneous sorption-desorption processes. Therefore, it is essential to understand the kinetics and sorption mechanisms ^{137}Cs and ^{90}Sr on colloidal soil surfaces [4].

2 Physical, Chemical and Radiologic Characteristics of Cs and Sr.

Caesium is an alkaline metal that has similar physical and chemical properties to rubidium and potassium. This metal is very reactive and very pyrophoric. Besides spontaneously igniting in air, it reacts explosively with water even at low temperatures [5]. The common cesium radionuclides are ^{137}Cs and ^{134}Cs [6]. With relatively long half-lives of 30 and 2 years, respectively, these radionuclides stay in the soil and are introduced into the food chain by plant assimilation. It is readily transported in plants as a potassium chemical analog [6].

As a result of its beta decay (at ^{137}mBa), ^{137}Cs is a powerful gamma radiation transmitter. Its half-life makes it the principal medium-lived fission product along with ^{90}Sr both

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are responsible for the radioactivity of spent nuclear fuel after several years of cooling up to several hundred years after use.

The interest in ^{137}Cs as a component of global fallout lies primarily in its potential as a continuing source of radioactivity for animals and humans. For example, ^{137}Cs and ^{90}Sr currently produce the greatest source of radioactive material in the region surrounding the Chernobyl disaster.

Strontium has a high chemical reactivity. While natural strontium is stable, the synthetic isotope ^{90}Sr is present in radioactive fallout and has a half-life of 28.90 years. Strontium is more reactive in water than Ca, with which it reacts to generate strontium hydroxide and hydrogen gas. It burns into the air to produce both strontium oxide and strontium nitride. However, it does not react with nitrogen below 380°C at room temperature and will form oxide only spontaneously [7]. Because strontium has a similar atomic radius to calcium, it easily replaces Ca in minerals [7].

3 Cesium and Sr in Soils

The amount of cesium and Sr in the crust is in general very small. Therefore, Cs is a natural element that has an average concentration of 3 mg kg^{-1} in rocks, whereas in soils the average is 4 mg kg^{-1} . However, the soil can be enriched with artificial radioactive cesium through nuclear weapons testing and accidents in nuclear power plants. Recently, large amounts of cesium were released into terrestrial and marine ecosystems when the Fukushima Daiichi nuclear power plant was damaged by a tsunami in 2011 [6, 8].

Cesium is a monovalent cation that adsorbs where organic matter and clayey minerals are negative. Radiocesium selectivity compared with other ubiquitous cations in soils is low for negatively charged functional groups of organic matter. Nevertheless, is elevated for interlayer locations with 2:1 amino-silicate clay minerals. Caesium has a very low hydration energy, so the electrostatic attraction of Cs ions of clay particles is great. Consequently, ions are preferably served by the clayey fraction [9]. Sorption is highly dependent on phyllo-silicate clay.

Several hot spots in Japan were investigated with respect to the activity concentrations of β^- -emitting ^{90}Sr and β^-/γ -emitting ^{134}Cs and ^{137}Cs in soil and vegetation samples [10]. They indicated that the ^{137}Cs activity levels were partly as high as in the $\text{kBq}\cdot\text{g}^{-1}$ range, the ^{90}Sr contamination levels of any sample did not exceed the $\text{Bq}\cdot\text{g}^{-1}$ range. Radiocesium contamination could be clearly attributed to the Fukushima nuclear accident by its ratio fingerprint ($^{134}\text{Cs}/^{137}\text{Cs}$). They reported that short-lived ^{89}Sr could no longer be determined, the source of the ^{90}Sr theoretically could, in part, also be fallout from the nuclear explosions of the 20th century or previous nuclear accidents. The dataset (although limited in number of samples) suggests an inherent co-existence of ^{137}Cs and ^{90}Sr in contamination due to the Fukushima nuclear accident. This

is very important in the current food monitoring campaigns. Which currently assumes that the activity concentrations of β^- -emitting ^{90}Sr are not greater than 10% of the level of γ -emitting ^{137}Cs (which can be measured quickly).

It is well documented that in many soils Cs is closely bound by clay minerals. Even in sandy soils, fixation appeared to extend for approximately 3 years after contamination [11]. Radiocesium is a serious threat to populations living in a contaminated environment.

Natural strontium belongs to a microelement and comprises a mixture of four stable isotopes: ^{84}Sr (0.56%), ^{86}Sr (9.96%), ^{87}Sr (7.02%) and ^{88}Sr (82.0%). The chemical properties of strontium are very similar to those of baryum, calcium, which can form the same salts and base. But the heavier strontium forms less mobile hydroxide, which leads to its accumulation in soil and plant tissues and live organisms. This is supported by the high level of this element in sediments of a variety of genera. Algae contain 26-140 mg per 100 g dry matter, whereas herbs contain about 2-3 mg/100 g d.m. The most common strontium salts of marine sediment are carbonate and phosphate. In all Geo-chemical and biochemical processes, calcium and strontium are associated. Their diet (Ca/Sr) in soils formed in bedrock and alluvial sediment inevitably determines the strontium content of plants. For this reason, it should be mentioned that soils and plants may be polluted with ^{90}Sr radioactive substances precipitated after nuclear explosions or accidents involving nuclear objects. In any case, when assessing soil qualities as an agricultural object, account must be taken of the total strontium content and its relationship with calcium.

The release of radioactive strontium isotopes to the environment began in the middle of the past century. The relationship between strontium and isotopes varies by nature because one of the strontium isotopes (^{87}Sr) is formed by the radioactive disintegration of the natural element rubidium (^{87}Rb). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are primarily used as water-to-rock interaction tracers [12]. The main sources of Sr in groundwater are atmospheric inputs, dissolution of Sr-bearing minerals, and anthropogenic inputs [13].

The adult human body contains about 0.3 grams of strontium. Virtually all strontium content is localized in the skeleton, whereas all other organs contain only 3.3 mg of strontium.

^{90}Sr is a pure beta emitter with a maximum energy of 0.54 MeV. Upon decay, it forms a daughter radionuclide ^{90}Y with a half-life of 64 h. The biological half-life for ^{90}Sr in the human body is estimated to be around 18 years. Because of its chemical similarity with calcium, it accumulates in the bones and radiates the bone marrow, resulting in high radiotoxicity. Another radiologically significant strontium radioisotope is beta-emitting ^{89}Sr ($T_{1/2} = 51$ days). However, due to the shorter half-life, it poses radiological importance only for relatively short periods (about 1 year) until more complete decay.

Sr appears on average of 370 mg kg⁻¹ in rocks and 240 mg kg⁻¹ in soils, [14]. The strontium content of soils is strongly controlled by the original rocks and climate. It has an average surface soil concentration of about 250 mg kg⁻¹ [15]. However, in some limestone soils and industrial regions, Sr levels higher than 1000 mg kg⁻¹ have been reported [16]. Soils in the San Joaquin Valley in central California contain Sr at average concentrations of 236-246 mg kg⁻¹ [17]. The median concentrations of Sr in Japanese agricultural soils vary from 32 to 130 mg kg⁻¹, respectively, in Acrisols and Gleysols [18].

Strontium is moderately mobile in soils, and the predominant cation, Sr²⁺, may be adsorbed as hydrated by clay minerals and Fe oxides and hydroxides. In calcareous soils, Sr may precipitate as strontianite (SrCO₃) and become less mobile. Its distribution in soil profiles follows the general patterns of circulation of soil solutions. Thus, in acidic soils, they are likely to be leached along the profile, while in calcareous soils, Sr can be concentrated in the upper horizons [19].

4 Factors affecting Cs/Sr retention in soil.

Soil is a complex system where soil minerals, organic matter and micro-organisms interact with each other in a variety of ways. The quantities, qualities and properties of these components vary considerably depending on the site. Therefore, it is very likely that the interactions of radionuclides with these soil components and the predominant retention mechanisms of radionuclides induced by these interactions in the surface layers of the soil. There are a number of factors that may affect the mobility and bioavailability of ¹³⁷Cs in soils. These factors, beside the clay mineralogy, include soil physicochemical properties such as the pH, cation-exchange capacity (CEC), presence of exchangeable cations, organic matter content; and soil structure [20], soil microbial activities and local climatic and ecological conditions [21, 22].

Normally, the fraction of a radionuclide found in the soil solution is considered to be a very small fraction of the total quantity. Consequently, the transfer depends on the ability of the solid phase to restore levels of radionuclides in the soil solution after adsorption or elution. Furthermore, this capacity depends on the fraction of the available radionuclide (pool of total radionuclide in the soil that participates in solid-liquid partitioning) and also on the distribution coefficient (soil solution-solid phase). The first, which quantifies the potential of soils to fix radionuclides, depends upon the time (aging process). The second is related to the sorption selectivity of the solid phase and the composition of the soil solution, particularly of competitive sorption species.

With respect to radiocaesium behavior, soil-chemical availability is essentially a function of radionuclide partitioning, between regular ion exchange complex (REC) and the specific sites on the frayed edges of clay layers (FES), in the solid phase. K and NH₄ levels in solid and liquid phases are of paramount importance as they are

competitive ions of Cs.

On the other hand, the key factors of radiostromium retention in soils are the cation exchange capacity and the composition of the REC, especially Ca and Mg levels, which are Sr-competitive species. Because radiostromium sorption and desorption is largely controlled by ion exchange processes.

4.1 Effect of clay content in soil on Cs and Sr retention

Adsorption capacities are caused by a net negative charge on the structure of clay minerals. This negative load allows the clays to adsorb positively loaded species. Their adsorption properties are also related to their high surface area and high porosity. Al-Rayyes, [23] reports that montmorillonite is an excellent sorbent because of its high specific surface, chemical and mechanical stability, stratified structure and high cation exchange capacity (CEC). Sparks [24] reported that the elevated CEC for montmorillonite is caused by substantial isomorphic substitution and the presence of fully expanded interlayers that promote cationic exchange. Mirkhani, et.al [25] (2012) indicated that the greatest amount of illite and vermiculite clay minerals in soils contributed significantly to the greater affinity of Cs in soils.

The adsorption behavior of Cs and Sr on two selected calcareous soils (clay and silty clay loam) with contrasting physical and chemical characteristics was studied in different concentrations of Cs and Sr by a batch method [25]. The chemical and physical properties of the soils have been determined and the principal clay minerals of the soils have been identified by XRD. Sorption characteristics Cs and Sr were determined at 22.5°C when 0.01 M CaCl₂ was present. They indicated that Freundlich isotherms were found to be suitable for experimental data from adsorbed Cs and Sr.

The study of the cesium adsorption on Ca-montmorillonite in the presence of ions of other metals revealed that ion exchange was a predominant mechanism of the Cs+ adsorption on montmorillonite in the range of nanomolar concentrations [26,27].

Thus, clays minerals play an important role in the environment by acting as a natural scavenger of pollutants by taking up cations and anions either through ion exchange or adsorption or both. As a result, the clay invariably contains exchangeable cations and anions that remain on the surface. The prominent cations and anions found on clay surface are Ca²⁺, Mg²⁺, H⁺, K⁺, NH₄⁺, Na⁺, and SO₄²⁻, Cl⁻, PO₄³⁻, NO₃⁻. These ions can be exchanged with other ions rather easily without affecting the clay mineral structure. The large specific area, chemical and mechanical stability, layered structure, high cation exchange capacity (CEC), etc., have made clays excellent adsorbent materials [28].

In most soil types, strontium is more mobile than caesium. The reason is that caesium is selectively fixed in clay minerals, particularly illite minerals. Whereas strontium is

either complex bound in organic or oxide compounds, or bound to exchange sites on clays and organic material. Exceptions to this general rule are soils with a high organic content, where ^{137}Cs may be quite mobile [29]. However, the ^{90}Sr is likely to be more strongly associated with the exchange complex due to the strong cationic exchange capacity of organic matter [30]. Generally, coarse-grained soils have a lower tendency to adsorb Cs&Sr than fine-grained soils. The fine-grained soil fraction contains large surface reactivities and large surface areas. Such as clay minerals, iron and manganese oxyhydroxide, humic acids, and other and displays improved adsorption properties.

4.2 Effect of soil organic matter on Cs and Sr retention

Soil organic matter directly and indirectly influences the behavior of radionuclides in mineral soils. Soil organic matter presents a variety of functional groups with high CEC values, which improves the retention capacity of metal ions, primarily through surface completion, ion exchange and surface precipitation. Xu *et al.*, [31] indicated that the high functionality, humic acids (HS) can strongly complex metal ions and thereby modify the retention, adsorption, migration and, bioavailability of metal ions in the natural environment. Using batch techniques, Tan *et al.*, [32] investigated the influence of HS on metal ion adsorption and its species on clay minerals and oxides is thereby studied extensively. The results indicate that HS enhances cations' adsorption at low pH values, but reduces the adsorption at high pH values.

Soil organic matter provides a high CEC and is therefore capable of adsorbing ^{137}Cs ; however, this adsorption is not specific to ^{137}Cs and adsorbed ^{137}Cs is easily exchangeable with other cations. In addition to direct influences, an indirect influence of soil organic matter on the ^{137}Cs behavior has also been suggested in that the presence of soil organic matter inhibits the fixation of ^{137}Cs by clay minerals by limiting the access of ^{137}Cs to ^{137}Cs -fixation sites on the clay minerals.

4.3. Effect of soil pH on Cs and Sr retention

As illustrated by Fig. 1, at pH 7 very little sorption occurs [33] whereas the k_d values of Cs as a function of pH of four studied soils. these specific experimental conditions. It can be also supported that the four soils exhibit a similar behavior when the pH is neutral.

Their results showed that the maximum sorption of Cs was observed at pH 8. The high sorption levels of Cs indicate a high fixation capacity of the studied soils for cesium ions in this pH region. According to the values of k_d , it seems that the clay soil (soil 3) was influenced greatly from the changes of the pH. The k_d of clay soil at pH 8 was higher by a factor of 4 compared with the respective k_d of the sandyloam soil (soil 1).

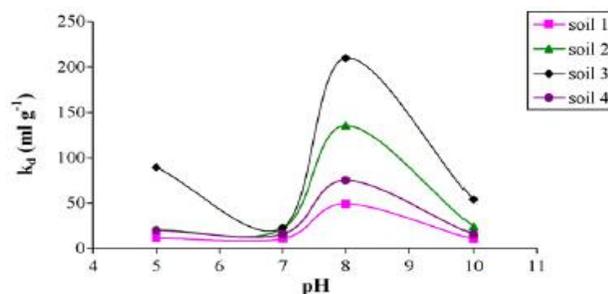


Fig. 1: The k_d values of Cs as a function of pH of four studied soils [33].

The increase in k_d for pH 8 is ascribed to the significant high negative charge density of the clay minerals. As pH increasing, the number of protons on the soil surface decreasing and more electrostatically attractive surface to the cations occur, allowing more Cs^+ to be sorbed.

Soil pH is one of the most important factors control radionuclides sorption to soil. **Faghihian *et al.***, [34] concluded that acidic medium has an inhibitory effect on the uptake process. This can be attributed to the hydronium ions competing with Cs^+ and Sr^{2+} for the exchange sites. In the other hand, the concentration of deprotonated surface sites of iron oxide increases with increasing pH value. Therefore, the adsorption capacity of the composite increased with increasing the pH, and the maximal adsorption was observed at pH of 8. They indicated that the pH dependence of $^{90}\text{Sr}(\text{II})$ sorption can be attributed to three factors: (1) the speciation of $^{90}\text{Sr}(\text{II})$ in aqueous solution; (2) the surface properties of Na-montmorillonite; (3) dissociation of functional groups. **Giannakopoulou [33]** studied the effect of pH on sorption behavior of Cs in four different soils (sandy loam, loam, clay loam and clay). At acid pH levels less cesium was sorbed, due to a greater competition with other cations for available sorption sites. The maximum sorption of Cs was observed at pH 8, where the negative charge density on the surface of the absorbents was the highest. For all soils was observed significantly lower Cs sorption at pH 10.

Shujun *et al.*, [35] reported that, the species of Sr^{2+} is present at a significant concentration in the studied pH range from 2.0 to 11.0, and the $\text{Sr}(\text{OH})^+$ species is negligible. Because of the protonation reaction (i.e., $\equiv \text{SOH}^+ + \text{H}^+ \leftrightarrow \equiv \text{SOH}^{2+}$) on the surfaces of Na-montmorillonite, the surface charge of Na-montmorillonite is positive at low pH. Therefore, the sorption of $^{90}\text{Sr}(\text{II})$ on Na-montmorillonite is unfavorable as a result of the electrostatic repulsion at low pH values. And the ion exchange between $^{90}\text{Sr}(\text{II})$ and H^+/Na^+ on Na-montmorillonite surface can lead to the sorption of $^{90}\text{Sr}(\text{II})$ on Na-montmorillonite. The concentration of deprotonated sites (SO^-) increases with increasing pH because of the surface deprotonation reaction (i.e., $\equiv \text{SOH} \leftrightarrow \equiv \text{SO}^- + \text{H}^+$) at high pH. The deprotonated sites ($\equiv \text{SO}^-$) are more available to retain the positively charged $^{90}\text{Sr}(\text{II})$ ions, and

more sorption sites are available for binding $^{90}\text{Sr}(\text{II})$ ions on Na-montmorillonite, thus resulting in an increase of $^{90}\text{Sr}(\text{II})$ sorption.

4.4 Effect of soil solution ionic strength on Cs and Sr retention

A knowledge of the effect of ionic strength on Cs adsorption is essential for the interpretation of many desorption studies. Bouzidi et al., [36] reported that the presence of divalent cation, Ca^{2+} , Mg^{2+} , and also the monovalent cation Na^+ inhibited significantly Cs adsorption. Among these cations, the presence of K^+ strongly constrains the Cs adsorption, especially when potassium concentration exceeds 10^{-3} M. This is in agreement with other studies that Cs adsorption is the result of cation-exchange reactions, in which cations, with similar radius and hydration energy, compete more effectively against Cs^+ ions on the mineral surface [37]. So this explains the behavior of potassium, because K^+ has the same characteristics that Cs^+ (ionic radius of 138 pm in comparison with 167 pm of Cs^+ ions, and similar hydration energy). While Ca^{2+} as well as Na^+ also affects the Cs adsorption to a certain extent, this is due to their high contents in GW solutions. They showed that the ability of the cations to depress the sorption of cesium in Ain Oussera soil can follow this order: $\text{K}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{Na}^+$. Tsai et al., [38] noticed that the Cs adsorption sharply decreases as the ionic strength increases. The presence of divalent cation, Ca^{2+} and Mg^{2+} , inhibit about half of the Cs adsorption. In addition, a significant negative correlation between increasing Na^+ concentration and Cs adsorption is clearly observed. Li et al., [39] studied the effect of ionic strength on the equilibrium adsorption amount. The adsorption of both Sr^{2+} and Ca^{2+} is not sensitive to ionic strength as NaCl concentration changes from 0.5 to 4 mol L^{-1} , suggesting that inner-sphere complexation reactions account for the adsorption and that there is a covalent bonding between the metal ions and the gel particles.

For the adsorption of Sr^{2+} to hydrous ferric oxide (HFO), adsorption was also found to be insensitive to ionic strength. In contrast, for strontium adsorption to hydrous aluminum and manganese oxides, cellulose/alginate ion-exchange membrane, and bacteria *Shewanella alga*, adsorption was found to decrease with increasing ionic strength, suggesting physical adsorption where hydrating waters were not lost upon adsorption and the outer-sphere complexation reactions accounted for the adsorption. Kasar et al., [40] indicated that Cs and Sr sorptions were found to be affected by exchangeable Na, K, Mg, and Al ions, silt + clay (%), and OM(%).

5 Distribution coefficients of Cesium and strontium ions on soils.

Huge amounts of radio-cesium, radio-strontium, and fission products were released into the atmosphere during the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident.

Therefore, it is necessary to determine the distribution coefficient of Cs and Sr in the soil-water system around FDNPP from the viewpoint of their migration. Soil K_d values are frequently used to relate the concentrations of RNs in contaminated soils with those in soil solution in order to assess radiological risk arisen from different radioactive contamination scenarios. Gil-Garcia [41], indicated that the pattern of radio-strontium and radio-cesium sorption-desorption was examined in 30 Spanish soils by the quantification of the distribution coefficients (K_d) with batch tests. The data obtained overlapped with those found in soils from other climatic areas, suggesting identical interaction mechanisms and allowing the extrapolation of parameterizations and prediction models among different scenarios.

Kasar et. al., [40] indicated that K_d -Cs was found to be in the range of 65 to 2100 L/kg while, Sr- K_d was in the range of 15 to 130 L/kg. High values of K_d reflect more sorption capacity of adsorbent for that nuclide. The concentration of both metals used initially for batch sorption experiments was of the same order of magnitude with co-addition of stable Cs and Sr metal solutions. The interaction of these metal ions with soil surface is responsible for the uptake of metal ions. The wide range of K_d values for Cs in Fukushima is related to the nature of the soil samples. The difference in distribution characteristics can be understood by estimating soil parameters and correlation studies to some extent [42].

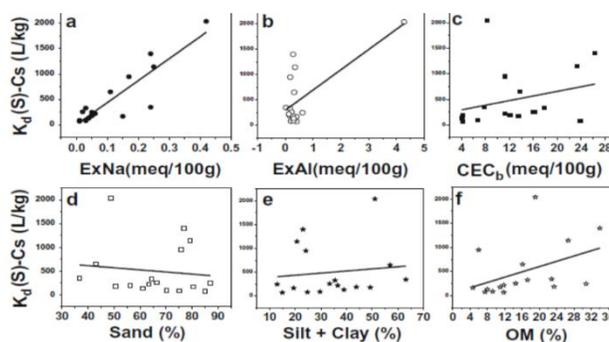


Fig.2: Pearson's correlation of $K_d(\text{S})\text{-Cs}$ with soil parameters. a Exchangeable Na. b Exchangeable Al. c CEC_b . d Sand (%). e Silt + clay (%). f Organic matter (%), [40].

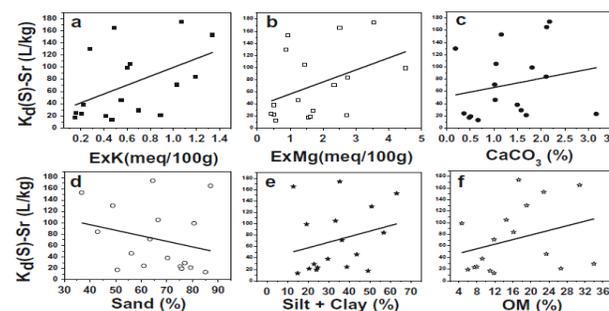


Fig. 3: Pearson's correlation of $K_d(\text{S})\text{-Sr}$ with soil parameters. a Exchangeable K. b Exchangeable Mg. c

CaCO₃ (%). d Sand (%). e Silt + clay (%). F Organic matter (%), [40].

5 Migration or the distribution of cesium and strontium in soil

The harmfulness of deposited radionuclides on the soil surface depends on their concentration that is influenced by their migration rates. If they percolate deeper, they may be assimilated by plant roots and even may contaminate the sub-surface water.

The vertical migration of radio-cesium fallout in the soil was monitored for 1 year at several locations in Fukushima after the nuclear power plant explosion [43]. He determined the vertical gamma ray intensity profiles in boreholes in the soil using a scintillation survey meter with a lead collimator to restrict the incoming radiation, only allowing horizontal detection. The average migration distances of radio-cesium at two time points were accurately determined based on the difference in the depth of the centroids of two gamma ray intensity profiles. The results showed that although the convective velocity of radiocesium was unexpectedly as high as 1/10th of the velocity of the infiltrating rainfall water 2–3 months after the nuclear plant accident, the velocity decreased to 1/100th–1/200th of that of the water after 6–12 months. This indicated that strong fixation of radio-cesium to clay particles occurred during the initial 2–3 months. Radio-cesium uptake by plant roots may have decreased remarkably along with the mobility of radio-cesium in the soil.

Two basic processes control the migration of the fallout in the undisturbed soil column. Firstly, the particles can percolate downwards with rainwater; second, the solvable phase is subject to convection and diffusion with the soil solution and sorption to the soil matter described by the diffusion-convection model equation.

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