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Behavior of ^{134}Cs, ^{90}Sr, and ^{238}Pu in different Syrian soils

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The experiment aimed to evaluate the vertical migration of ^{134}Cs, ^{90}Sr and ^{238}Pu in the main types of Syrian soils; entisol, inceptisol, alluvial (rock outcrops) and gypsiferous soils, using soil columns through which the aqueous solution of the radionuclides percolated. The results show that the vertical migration of the studied radionuclides through the soil profile depend on the radionuclide and the soil type. More than 97% of ^{134}Cs and ^{238}Pu concentrated in the upper 2 cm of the entisol, inceptisol, and alluvial soils, whereas only 46.2% to 68.6% of the ^{90}Sr was retained in the upper 2 cm of these soils. The vertical migration of the studied radionuclides in the gypsiferous soil was different from the other soils. The distribution of the radionuclides in the gypsiferous soil was irregular through the soil profile and reached the deeper layer of the soil. This may be due to its physical characteristics; poor structure stability, high permeability and low retention capacity.

Introduction

The distribution of radionuclides in the soil profile after their release and deposition on the soil surface is essential for reliable assessment of the external gamma dose from the soil,[1] as well as of plants' root uptake. Thus the migration behavior, especially of the long-lived radionuclides in various types of soils is necessary.[2]

The harmfulness of deposited radionuclides on the soil surface depends on their concentration which is influenced by their migration rates. Thus their resuspension will be more harmful if they remain on the surface, affecting humans by direct irradiation, inhalation and ingestion of contaminated crops.[3] If they percolate deeper, they may be assimilated by plant roots, and even may contaminate the sub-surface water.

Experimental data from different region in the world[4–6] show that the rate of radionuclide migration depends on soil type. Both organic and inorganic soil components show an affinity for cesium limiting its down-word migration.[7,8] Some inorganic components of the soil, especially clay, have a remarkable capacity for adsorbing and retaining cesium.[1] Migration parameters of plutonium isotopes are proposed to be the same as cesium,[9,10] while relative migration of strontium through soil profile is more than that of cesium.[11,12]

The behavior of radionuclides through soil profile has been extensively studied,[13–17] however, these investigations were almost exclusively convened to temperate and humid regions. There is scarce information about radionuclides migration through soil profile in arid and semi-arid regions,[18] especially for gypsiferous soils, where no data are available.

The purpose of the present study is to obtain information on the vertical migration of ^{134}Cs, ^{90}Sr and ^{238}Pu in the various types of Syrian soils especially gypsiferous soil which occupies 21% of Syrian area.[19]

Experimental

Soil types

Four main types of Syrian soils were selected;[20]

(1) Inceptisol (Calcixerollic Vertic Xerochrepts) is the major agricultural soil in Syria.

(2) Entisol (Lithic Xerorthents) is situated in the coastal mountain regions, and represents the forest soil in Syria. The climate of the coastal mountain regions is of eastern Mediterranean type, marked by a mild, rainy winter and a prolonged (about 6 months) dry and hot summer.

(3) Gypsiferious soil (Hypergypsic Gypsiorthids) is distributed mainly in the arid zone of Syria with less than 200 mm of annual rainfall.

(4) Alluvial soil (Rock Outcrops) is located in AL-Ghab around Orients River.

Soil collection

Soil samples of each studied soils were collected from an area of one square meter at a depth of 0–2, 2–5, 5–10, 10–20, and 20–35 cm. The soil samples were air dried, passed through 0.5 mm sieve, and mixed again to measure ^{134}Cs, ^{238}Pu and ^{90}Sr concentrations. Soil characteristics were determined for a bulk soil sample to a depth of 0–35 cm.

Soil analysis

Soil pH was determined in soil $CaCl_2$ solution 1:2.5, particle size was determined by a method described by BLACK et al.[21] The organic matter, cation-exchange capacity (CEC), exchangeable cations were analyzed according to PAGE et al.[22]

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Experimental design

Eight plastic cylinders made of Polypropylene with 10.5 cm inner diameter and 60 cm long were installed vertically on a wooden stand, a Perspex disc (Polymethyl methacrylate) of 1 mm diameter mesh and 2 mm thick was fixed at the bottom inner side of the cylinder, a layer of fiberglass was put on the disc to avoid any loss of the fine particles.

The soil was filled in the cylinders in the same order as in the field and each layer was compacted to its field bulk density, then a Perspex disc similar to the bottom one was fixed on the top surface of the soil to avoid soil suspension and to ensure an even distribution of water on the soil surface. All soil columns were covered with plastic sheet to minimize water evaporation. Plastic beakers were placed under the columns to receive any possible effluents, and all columns were duplicated.

Soil labeling and irrigation

The soil in each column was wetted with one liter of water (near saturation), to insure easy distribution of the radioactive solution. A 100 ml of water solution containing 550 Bq of ^{90}Sr, 1000 Bq of ^{134}Cs, and 90 Bq of ^{238}Pu were added to each column. Soil irrigation was carried out once a month and for six months. The amounts of added water to the different soil types were calculated according to the average annual rainfall in the regions of the collected soils.

Radioactivity measurements

All measurements of soil radioactivity were done at the end of the experiment (8 months after the first irrigation). Soil columns were cut into soil slices of 0–2, 2–5, 5–10, 10–20 and 20–35 cm depths. The soil samples were oven dried at 105 °C to a constant weight, pounded with a wooden hammer and sieved to pass a 0.5-mm aperture. Cesium-134 was counted directly by gamma-spectrometry using HPGe detector (with active volume 100 cm^3 and 26% efficiency), where the amplifier output of the detector was processed using 4096-channels MCA system. The activity of ^{134}Cs has been calculated from energy peaks 604 keV using the same geometry for all samples.[23] The LLD for ^{134}Cs was 2 Bq/kg for overnight counting.

Strontium-90 measurements were done after digestion with concentrated HNO_3 and addition of ^{85}Sr as a tracer, and then the ^{90}Sr was precipitated as strontium nitrate in concentrated nitric acid medium. The ^{90}Y equilibrated as yttrium oxalate and counted using proportional counter. The recovery yield was determined by counting ^{85}Sr using gamma-spectroscopy.[23] A proportional counter (sample detector diameter of 5.7 cm and its efficiency: alpha≥18% ^{241}Am, beta≥42% ^{90}Sr) has been used to determine ^{90}Sr activity, and the background was measured for blank source and counted for 100 minutes. The efficiency was determined by counting a reference source for the same time and the same windows taking the self absorption in consideration, the LLD for ^{90}Sr was 3 Bq/kg.

Plutonium-238 was measured by leaching with acid after tracing the samples with ^{236}Pu for determination of chemical yield, and then sample solution purified from actinides using ion-exchange resin, then plutonium isotopes co-precipitated with neodymium fluoride and alpha-activity measured by alpha-spectroscopy.[24] The LLD was less than 1 Bq/kg.

Quality control test of radioactivity determination was performed using reference materials provided by the International Atomic Energy Agency, IAEA. Quality control results are presented in Table 1.

Results and discussion

Soil characteristics

The main characteristics of the bulk soil (0–35 cm) are presented in Table 2. The textures of the studied soils were of silty loam to sandy loam, the pH ranged between 7.31 and 8.14 which is a characteristic of the region. Cation-exchange capacity ranged between 7.39 meq/100 g for the gypsiferous soil and 52.2 meq/100 g for the inceptisol. The organic matter content was low (0.99%) in the inceptisol to medium (5.28%) in the entisol. The entisol and alluvial soils have higher exchangeable Ca (24.2–29.5 meq/100 g) than the inceptisol and gypsiferous soil (about 9.5 meq/100 g), on the other hand the exchangeable K was higher in the inceptisol (3.96 meq/100 g) than that in the other studied soils (0.11–1.21 meq/100 g).

Table 1. Quality control results

Sample code	Radionuclide	Reference value, Bq/kg dw	Measured value, Bq/kg dw
Soil-375	^{137}Cs	4677	4513 ± 145
Soil-6	^{137}Cs	53.65	50.7 ± 4.5
IAEA-135	^{137}Cs	1108	1167 ± 42
Soil-375	^{90}Sr	94.6	97.5 ± 9.6
Soil-6	^{90}Sr	30.34	28.2 ± 2
IAEA-135	^{90}Sr	64.5	70 ± 8.3
Soil-6	$^{239+240}$Pu	1.04	0.99 ± 0.08
IAEA-368	$^{239+240}$Pu	31	29 ± 3.4
IAEA-135	$^{239+240}$Pu	213	227 ± 14.7

Table 2. Physical and chemical characteristics of the studied soils

Parameter	Soil type			
	Inceptisol	Entisol	Alluvial	Gypsiferous
pH $CaCl_2$	7.31	7.30	7.82	8.14
Clay, %	23.65	23.90	19.30	18.20
Silt, %	59.50	41.95	43.50	36.45
Sand, %	16.85	34.15	38.20	45.35
CEC, meq/100 g	52.2	43.5	21.3	7.39
OM, %	0.99	5.28	2.14	1.56
Ex. Ca, meq/100 g	9.38	24.2	29.5	9.5
Ex. K, meq/100 g	3.96	0.11	1.21	0.94
Average annual rainfall, mm	300	1390	630	210

Cesium-134 migration

Table 3 shows that more than 98% of the added ^{134}Cs was concentrated in the upper 2 cm for the inceptisol, entisol, and alluvial soil. The migration of ^{134}Cs through the soil profile of the gypsiferous soil was different than the other types of soils, where the maximum ^{134}Cs activity was situated at a depth of 10–20 cm (37%) and 7% of the activity reached the deeper layer of the columns (20–35 cm).

The results obtained for ^{134}Cs migration through the soil profiles of inceptisol, entisol and alluvial soil were in agreement with other studies conducted in different region. PRICE[25] reported that up to 99% of cesium was retained in the top (0–5 cm) soil layer. RAURET et al.[26] found that 81–97% of the added cesium to the loamy soil was absorbed in the top 2 cm layer. The same result was obtained by NIELSON and STANDBERG,[27] where more than 90% of the deposited cesium was retained in the top 2 cm of the soil after two years of Chernobyl accident. Also AL-RAYYES[28] reported that more than 85% of the ^{137}Cs activity was still in the top 5 cm layer of the entisol in Syria after 10 years of Chernobyl accident.

The low ratio of the migrated cesium through the soil profile may be attributed to the strong bound of cesium with clay particles.[29,30] Moreover, KONOPLEV et al.[30] reported that cesium migration through the soil profile could be attributed to the movement of clay particles or as a result of soil mixing.

The migration of ^{134}Cs through the profile of the gypsiferous soil was different than the other studied soils, where the distribution of added cesium was irregular through soil layers. These differences of ^{137}Cs distribution may be due to the physical characteristics of the gypsiferous soil, where the particles of the gypsiferous soils are weakly aggregated as the cohesive forces of attracting single soil particles are very weak. BOYADGIEV[31] noticed that gypsum content over 15% in soil (the studied soil contains about 25% gypsum) tends to give unstable structure and caverns can be formed through mechanical removal of fine soil particles. Also he concluded from his results in the Euphrates basin of Syria, that soils containing more than 10% gypsum are permeable, have poor structural stability and low water retention capacity. SMITH and ROBERTSON[32] observed that soils with more than 25% gypsum, in Iraq, lack plasticity, cohesion and aggregation and become completely unstable in water. Thus the mechanical movement of the gypsum particles with water in gypsiferous soils plays a significant role in the irregular migration of ^{134}Cs in the soil profile.

Strontium-90 migration

The results show that ^{90}Sr migration throughout the soil profile was higher than ^{134}Cs (Table 4), where about 46.2 and 51.7% of the added ^{90}Sr remained in the upper 2 cm of the inceptisol and alluvial soil respectively, whereas a small amount of ^{90}Sr (about 4%) reached the depth of 20–35 cm. These results may be explained by the low content of organic matter in these soils (less than 1%). For the entisol which is of medium content of organic matter (5.2%), the pattern of ^{90}Sr distribution throughout the soil profile was slightly different compared with the inceptisol and alluvial soil, where 68.6% of ^{90}Sr activity concentrated in the top 2 cm, and about 11% was situated at a depth of 2–5 cm, whilst the activity at a depth of 20–35 cm was about 2–3 folds (9.3%) higher than that of the inceptisol and alluvial soil.

Our results are in agreement with other studies carried out in other regions of the world. ANDERSSON et al.[1] and CLINE and CADWELL[33] indicated that in general the rate of strontium migration exceeded that of cesium in both mineral and organic soils, and the mobility of ^{90}Sr decreased as the amount of organic matter increased.[12]

The migration of strontium in the gypsiferous soil was different compared with the other studied soils. Table 4 shows that the activity of strontium did not exceed 12% in the upper 2 cm and 4.8% in the 2–5 cm layer. Strontium-90 concentration increased to 16.2%, 18.1%, and 49.5% in the 5–10 cm, 10–20 cm and 20–35 cm soil depth, respectively. The different migration of ^{90}Sr throughout the profile of the gypsiferous soil could be attributed to its characteristic, i.e., poor structural stability, high permeability and low water retention capacity.

Table 3. Retained (in %) ^{134}Cs in the profile of the different soils

Soil type	Depth, cm				
	0–2	2–5	5–10	10–20	20–35
Inceptisol	99 ± 1	T	T	T	T
Alluvial	98 ± 1	T	T	T	T
Entisol	99 ± 1	T	T	T	T
Gypsiferous	33 ± 1	10 ± 1	14 ± 1	37 ± 1	7 ± 1

T: Trace.

Table 4. Retained (in %) ^{90}Sr in the profile of the different soils

Soil type	Depth, cm				
	0–2	2–5	5–10	10–20	20–35
Inceptisol	51.7 ± 3	29.5 ± 4	11.3 ± 3	3.2 ± 1	4.3 ± 2
Alluvial	46.2 ± 3	39.2 ± 3	8.3 ± 1	2.1 ± 1	4.1 ± 1
Entisol	68.6 ± 4	11.1 ± 2	3.9 ± 2	7.2 ± 2	9.3 ± 2
Gypsiferous	11.9 ± 3	4.8 ± 1	16.2 ± 2	18.1 ± 4	49.5 ± 5

Table 5. Retained (in %) ^{238}Pu in the profile of the different soils

Soil type	Depth, cm				
	0–2	2–5	5–10	10–20	20–35
Inceptisol	97.5 ± 4	0.6 ± 0.2	1.1 ± 0.3	0.75 ± 0.3	0.08 ± 0.04
Alluvial	98.2 ± 7	1.1 ± 0.3	0.2 ± 0.04	0.3 ± 0.1	0.07 ± 0.03
Entisol	98.5 ± 5	0.8 ± 0.2	0.2 ± 0.08	0.3 ± 0.1	0.13 ± 0.1
Gypsiferous	80.5 ± 7	6.5 ± 1	1.4 ± 0.5	11.1 ± 1.6	0.5 ± 0.1

Migration of plutonium-238

Table 5 shows that plutonium behavior throughout the profiles of the studied soils was almost similar to that of ^{134}Cs, where more than 97% of the added plutonium to the inceptisol, entisol and alluvial soil, was retained in the upper 2 cm. These results were in agreement with other studies. PRICE[25] and HARLEY[34] indicated that plutonium migration through the soil profile was slightly higher than that of cesium and lower than strontium, and they found that about 95% of the deposited plutonium was retained in the upper 5 cm layer of the soil, even after 10 years of the fallout deposition.

Similar to ^{134}Cs and ^{90}Sr the distribution of plutonium in the gypsiferous soil was different compared with the other studied soils. Table 5 shows that the maximum activity (81%) of the added plutonium was retained in the top 2 cm of the soil, 7% in the 2–5 cm layer, and only about 0.5% reached the layer of 20–35 cm. As for ^{134}Cs and ^{90}Sr the irregular ^{238}Pu migration throughout the profile of gypsiferous soil may be due to the physical properties of this soil.

Conclusions

During eight months of the experiment, the migration of ^{134}Cs and ^{238}Pu in the inceptisol, entisol and alluvial soil was low, and up to 97% of both radionuclides were retained in the upper 2 cm of the soil, while ^{90}Sr migration exceeded that of ^{134}Cs and ^{238}Pu in the same soil types.

The migration of the ^{134}Cs, ^{238}Pu and ^{90}Sr in the gypsiferous soil were irregular through the profile and reached deeper soil layers. This may be due to the physical characteristics of the gypsiferous soil: poor structural stability, high permeability and low retention capacity. Therefore, more care should be taken when dealing with contaminated gypsiferous soils with radioactive materials, because of the possible translocation of radionuclides throughout the soil profile.

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