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CHARACTERIZATION AND IMMOBILIZATION OF CESIUM-137 IN SOIL AT LOS ALAMOS NATIONAL LABORATORY

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Characterization and Immobilization of Cesium -137 in Soil at Los Alamos National Laboratory

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ABSTRACT

At Los Alamos National Laboratory, cesium-137 (^{137}Cs) is a major contaminant in soils of Technical Area 21(TA-21) and is mainly associated with soil particles ≤ 2.00 mm. Cesium-137 was not leached by synthetic groundwater or acid rainwater. Soil erosion is a primary mechanism of ^{137}Cs transport in TA-21. The methodology that controls soil particle runoff can prevent the transport of ^{137}Cs .

INTRODUCTION

Technical Area 21 (TA-21) at Los Alamos National Laboratory (LANL) is situated on DP Mesa and was used primarily for research and production of plutonium metal from 1946 to 1978. Solid Waste Management Unit (SWMU) 21-011(k) is located on the south wall of DP Canyon. The soils in SWMU 21-011(k) contain multiple contaminants which originated from discharges of acid-waste treatment facilities and spills from holding tanks. The initial radiological survey in 1992 and 1993 showed that americium-241, cesium-137, plutonium-239+240, and strontium-90 exceeded both the Baseline Concentrations and their respective soil Screening Action Levels (SAL). The radioactive levels of ^{137}Cs in surface soil range from 1,578 to 119,362 Bq/kg soil, which is 8 to 632 times as much as its SAL (188.7 Bq/kg) (LA-UR-94-228, 1994).

Cesium-137 interacts strongly with micaceous clay minerals (Maule and Dudas, 1989) and organic matter in soils (Comans and Hockley, 1992). It was reported (Robbins et al., 1992; Wieland et al., 1993) that about 90% of Chernobyl ^{137}Cs fallout in the sediments of a German lake and of a Swiss lake was strongly bonded with clay minerals and was significantly transferred on non exchangeable sites of clay minerals. Irreversible sorption of ^{137}Cs on soil clay becomes significant over time scales of weeks to months. In this case, Cs migrates to interlayer sites of clay minerals, from which it is not easily released (Comans and Hockley, 1992). An appreciable quantity of ^{137}Cs was unlikely to be leached from contaminated soils, even under prolonged high rainfall (Kirk and Staunton, 1989). An average vertical migration rate of ^{137}Cs in an unsaturated soil layer was 1.0 mm yr^{-1} when an average rate of movement of soil water was 2500 mm yr^{-1} during forty years after the explosion of the atomic bomb at Nagasaki, Japan in 1945 (Mahara, 1993). More than 95% of ^{137}Cs deposited as fallout was above a depth of 10 cm in the soil, and no ^{137}Cs was detected below 40 cm.

This study was focused on ^{137}Cs because ^{137}Cs is a major contaminant in TA-21. Understanding the characterization and reaction of ^{137}Cs in soil helps us to search proper methods for controlling transport of ^{137}Cs and stabilizing it. The objectives of the research were to investigate the characterization of ^{137}Cs in the soils; to examine the leachability of ^{137}Cs in soil using solutions of synthetic groundwater, synthetic acid rainwater, ammonium acetate, and citrate-bicarbonate-dithionite (CBD); and to test a potential approach for stabilization of ^{137}Cs by using a sand barrier and clay minerals of illite and zeolites. Five samples, including 1) relatively high level radioactive contaminated soil (Hot Spot), 2) surface soil (Surface), 3) wet sediment (Sediment), 4) drainage channel water, and 5) pine bark, were studied.

MATERIALS AND METHODS

Sampling, Soil Properties and Radiological Analysis

Five samples were taken from TA-21-011(k) area. Relatively high level radioactive contaminated soil was collected from a discharge point (Hot Spot) close to the top of DP Canyon wall. The surface sample was collected from soil (0 to 15 cm deep) close to the bottom of DP Canyon. The wet sediment was collected from the sediment (0 to 15 cm deep) at the outfall of a drainage channel which passes through the hot spot area and drained into the canyon. The water sample was separated from the wet sediment sample by centrifuging. The bark sample was collected from a Ponderosa pine [*Pinus ponderosa* (Dougl. ex

Laws)] tree at approximately 1-m height from the ground, which is growing close to the bottom of DP Canyon. The soil samples were air dried in a fume hood, passed through a 4-mm U.S. standard sieve and stored for use. Soil samples at particle-size fraction of 4.0 -2.0 mm, 2.0 - 0.053 mm and < 0.053 mm were prepared by passing 100 g of soil (≤ 4.0 mm) consecutively through a 2.0-mm and a 0.053-mm sieves. These samples were used for determination of ^{137}Cs distribution in soil particle size fractions. The pine bark sample was dried in an oven at 60°C for 72 h, ground and passed through a 0.5-mm standard sieve.

The pH of soil samples ranged from 7.7 to 8.2, which was measured on soil: water of 1:1 ratio. The Hot Spot and Sediment soils consist of 10% fine gravel, 50% or more of sand, 25% silt, and 10% clay, and low organic carbon content, ranged from 6 to 23 g/kg soil. Organic carbon contents of the soils were determined by dichromate oxidation method (Nelson and Sommers, 1982).

The gamma activity of soils and pine bark were measured as follows: five grams of air-dried soil were placed in a counting container and analyzed by gamma- spectroscopy. Measured activities of ^{137}Cs were decay corrected and, hence, activities reported in this paper correspond to initial activities on March 7, 1995 (sampling time, $t = 0$).

Column Leaching

Hot Spot and Sediment soils were leached using solutions of synthetic groundwater (pH 8.0)(Elless et al., 1994), synthetic acid rainwater (pH 4.1), 0.5 M ammonium acetate (NH_4OAc , pH 7.0), and a mixture of 0.5 M sodium citrate-0.1 M sodium bicarbonate-0.1 M sodium dithionite (CBD, pH 7.2), respectively.

The glass funnels contained porous stone as base, with an 80-ml capacity, and were utilized as columns to perform the leaching process. Seventy grams of soil at particles of ≤ 4.0 mm were loaded in each column. The soil column was slowly saturated by introducing solution from a single tube at the center-top of the column with an application rate of 0.005 cm/min during the first 2 hours. Upon appearance of effluent at the bottom of the column, the application rate of leach solution was adjusted to 0.01 cm/min and remained constant during the 10-day period. A duplication was made from each treatment and a total 16 of the columns were conducted at room temperature. After leaching the columns for 10 days, the soils in the columns were removed, air dried and crushed to ≤ 2.0 mm. The gamma activities of the air-dried soils were analyzed using gamma spectroscopy.

A Sand Barrier

A *Plexiglas* column, with a length of 15 cm and an inside diameter of 9 cm, was used to construct a "sand barrier" which consisted of 5 layers: the top layer of 2-cm height fine gravel (>2 mm), the second layer of 1.5-cm height coarse sand (0.5 to 2.0 mm), the third layer of 7-cm height fine sand (<0.5 mm), the fourth layer of 1.5-cm height coarse sand, and, finally, a fifth layer of 2-cm height fine gravel layer at the bottom of the column. A screen (<4.0 mm mesh) was placed on the top and the bottom of the sand barrier (Fig. 1). Before the sand barrier was constructed, the gravels and the Ottawa sands were washed with deionized water several times and air dried. After the barrier was built up, the sand barrier was moistened by passing through about one liter of deionized water.

Thirty-eight grams of Hot Spot soil (particles of < 2.0 mm) were added to 500 ml of fresh synthetic groundwater ($\sim 7.6\%$ turbidity). The mixture was stirred vigorously for 30 min. and was completely suspended. A subsample was taken for initial radioactivity determination when the suspension was still

stirring. Following this, 400 ml of the suspension was poured on top of the sand barrier. All filtered water was collected at the bottom of the column and mixed thoroughly. The liquid was taken for radioactivity determination. The sand barrier was allowed to stand for 6 to 8 hours. After that, each layer was removed from the column and air dried. The radioactivity of each layer was determined by the same method used for radiological analysis of the air-dried soils.

Sorption and Desorption of Cesium-137.

Laboratory sorption and desorption experiments were conducted with illite and zeolite resins of A-51[®], IE-95[®], and EP-9174[®] to evaluate the sorption and desorption of ¹³⁷Cs on these clay minerals. The particle size of the clays was less than 0.5 mm. Carrier-free ¹³⁷CsCl (non-radioactive Cs was not used as a carrier) was completely dissolved in fresh synthetic groundwater (pH 8.0). The initial activity of ¹³⁷Cs in the solution was 0.05 mCi/L. The clays and ¹³⁷Cs labeled groundwater at a ratio of 1:30 were transferred into 50-ml centrifuge tubes and continuously shaken at a constant temperature of 20 ± 1°C. Sorption was allowed to proceed from 6 hours to 480 hours. The samples were taken after 6, 24, 120, 240, and 480 hours, and subsequently centrifuged at 17,000 g for one hour to separate the liquids from solids.

Desorption studies were performed with the solids obtained from the sorption experiment. The solids were sequentially extracted with cesium-free fresh synthetic groundwater for the same periods of time used in the sorption experiment. The extracts were separated from solids by centrifuging.

The gamma activities of the liquids, collected from both sorption and desorption experiments, were analyzed using gamma spectroscopy. Measured activities of ¹³⁷Cs were decay corrected and, hence, activities reported here correspond to initial activities at the time that the sorption experiment was started.

RESULTS AND DISCUSSION

Cesium-137 Distribution

The highest level of ¹³⁷Cs occurred in the Hot Spot soil and the quantity of ¹³⁷Cs in the air-dried soil was 13,722 Bq/kg, which was 3.5 times as much as in the Sediment from the outfall of the drainage channel (4,155 Bq ¹³⁷Cs/kg), and was 25 fold as much as in Surface sample (152 Bq/kg) collected from the location close to the bottom of DP Canyon (Fig. 2). The results show that ¹³⁷Cs is a major contaminant in TA-21-011(k) area and ¹³⁷Cs in Hot Spot soil is 73 times as much as its SAL and 22 times of its SAL in the Sediment.

After water was separated from the wet sediment by centrifuging, ¹³⁷Cs in the water could not be detected by the gamma spectroscopy. Furthermore, ¹³⁷Cs in pine bark could not be detected by the gamma spectroscopy (Fig. 2). The results suggest that ¹³⁷Cs was neither released from the soil particles into water nor accumulated in the pine bark. The level of ¹³⁷Cs in the soil close to the bottom of the canyon was low, perhaps ¹³⁷Cs did not migrate to the roots of pine tree.

Wind can cause soil erosion and is one mechanism of ¹³⁷Cs transport. However, ¹³⁷Cs concentration in the sediments from outfall of the drainage channel were much greater than in the surface soil from the bottom of the canyon. Cesium-137 in the sediments may come from runoff (erosion) of soil particles in Hot Spot area, moving along the drainage channel to DP Canyon.

Soils on the top of DP Canyon are often eroded with summer storm runoff down to the canyon. In Los Alamos, about 36% of the annual precipitation falls from storms during July and August. It was reported (Purtymun, 1974) that during the summer of 1967, rainfall resulted in 23 runoff events that carried about 88,000 kg of suspended sediments and transported down to the canyon in about 36,800 m³ of water. With this large amount of sediment runoff, about 2.59 x 10⁶ Bq of gross alpha emitter and about 4.1 x 10⁵ to 1.1 x 10⁷ Bq of gross beta were transported with suspended sediments to the bottom of the Canyon (Purtymun, 1974). Cesium-137 is still found further down the Canyon.

As shown in Figure 3, radioactivity of ¹³⁷Cs decreased linearly as the soil particle size increased. The relationship between ¹³⁷Cs activity and particle size follows an equation of $Y=19,602.9 - 2,158.4X$ with correlation coefficient (r) of -0.9998, where Y is the activity of ¹³⁷Cs in Bq/kg, X is the soil particle size in mm. According to the percentage of each particle size fraction, ¹³⁷Cs concentration in each fraction was calculated using the data shown in the Figure 3. In 1 kg of air-dried Hot Spot soil, the fine gravel (11.5 wt. %) contained 1,255 Bq of ¹³⁷Cs, the sand fraction (48.8 wt. %) contained 7,503 Bq of ¹³⁷Cs, and silt-clay fraction (37.4 wt. %) contained 7,266 Bq of ¹³⁷Cs. These results suggest that ¹³⁷Cs in the soil from TA-21-011(k) is mainly associated with the sand and silt-clay fractions. The radioactivity of a whole unfractionated soil (≤ 4.0 mm) from Hot Spot (13,722 Bq/kg) was 17% less than the sum of the radioactivity from its fine gravel, sand, and silt-clay fractions (16,024 Bq/kg). This is likely due to vibrations encountered by the sample vial containing the whole soil sample during transportation and handling, where the smaller soil fractions would tend to settle to the bottom of the vial. The larger, and less radioactive, soil fractions (*i.e.* the fine gravel) would shield the gamma counter, located above the sample vial, from the finer, and more radioactive, soil fractions.

The silt and clay fractions of Los Alamos soil are largely montmorillonite and illite which are weathering products of the tuff (Staritzky, 1949). Montmorillonites and illites have strong ability to hold monovalent cations such as Cs⁺, K⁺, NH₄⁺ and Rb⁺ against the effects of leaching by rainfall (Borchardt, 1977; Fanning and Keramidas, 1977). These cations are weakly hydrated and lose their shell of hydration more easily than other cations, thus they may enter interlayer of illite preferentially. Once within the interlayer these ions may become "fixed." When this happens, the Cs ions are not exchangeable. Therefore, ¹³⁷Cs bound to these clay minerals is not leached by rainfall. Our data also showed that ¹³⁷Cs was not leached by synthetic groundwater and synthetic acid rainwater.

Sand-size (0.053 to 2 mm) fraction of Los Alamos soil consists of largely quartz and alkali feldspar crystals and crystal fragments and small amounts of rock fragments of tuff, pumice, rhyolite, and some mafic minerals (Purtymun, 1974). The crystals and rock fragments of tuff, pumice have large surface areas to adsorb ¹³⁷Cs.

Column Leaching of Cesium-137

In general, the leachability of ¹³⁷Cs from the soil was low. Cesium-137 was not leached from the soil using either synthetic groundwater or synthetic acid rainwater after 10 days of leaching. However, using NH₄OAc, about 9% of ¹³⁷Cs in Hot Spot soil and 30% of ¹³⁷Cs in Sediment was removed, while about 11% to 15% of ¹³⁷Cs was removed from the soil using CBD (Fig. 4).

Ammonium acetate solution was used to examine how much ^{137}Cs ions in the soil exchanges with NH_4^+ ions and so removed from the soils (Simard, 1993). Ammonium ions have the same hydrate size as Cs ions, while other cations (e.g. Ca^{2+} , Mg^{2+}) have larger hydrate size than Cs^+ or NH_4^+ ions (Bohn et al., 1985). Thus, NH_4^+ ions may move in to the exchange sites to replace Cs^+ ions. However, when Cs^+ ions are fixed in the interlayer of illite or montmorillonite, even NH_4^+ ions could not displace Cs^+ ions from the interlayer. Cesium ions may be removed from the external surface of the minerals by NH_4^+ ions.

The CBD solution was used to remove ^{137}Cs -bonded to Fe-oxide without disrupting clay structure (Mehra and Jackson, 1960). Free iron oxides, occur as discrete particles, nodules or as coatings on the soil particles. Free hematite (Fe_2O_3), goethite (FeOOH) and amorphous iron oxides are selectively dissolved by CBD (Mehra and Jackson, 1960). Therefore, the complex-reducing reagent of CBD removes mainly coated oxides bonded ^{137}Cs on the minerals during the leaching process. However, the percentage of ^{137}Cs removed was not directly related to the total Fe content in the soil. It is assumed that $\text{Fe}(\text{OH})_3$ is representative empirical formulas for the Fe oxide (Tessier et al., 1979), and ^{137}Cs bond to Fe oxide originates only from Fe oxide. Solution of CBD may remove large amounts of Fe with small amounts of ^{137}Cs , depending on the soil properties and the fractions with which ^{137}Cs associated.

Sand Barrier and Clay Sorbents

The initial concentration of ^{137}Cs in the suspension was 15,475 Bq/kg per liter synthetic groundwater. After 400 ml of the suspension was passed through the sand barrier, the ^{137}Cs in the filtered water could not be detected (Fig. 5). However, the ^{137}Cs level was 528 Bq/kg fine gravel at the top layer of the sand barrier, and 5,177 Bq/kg coarse sand at the second layer of the barrier, and no ^{137}Cs was found below the second layer of the barrier (Fig. 5). The radioactivity balance from the sand barrier was 92.2% of recovery for all ^{137}Cs labeled soil particles. The error of 7.8% was due to gamma spectroscopy and experimental error. The result indicates that the sand barrier blocks the fine particles with which ^{137}Cs was mainly associated in the TA-21-011(k) soils.

The principle of the sand barrier is based on the filtration step for drinking water treatment. A typical sand filter is commonly used to remove suspended particles and colloidal matter in stream water. The sand filter is constructed with a smaller size of sand in the top surface, the larger size of graded gravel in the bottom (Cleasby, 1990). When the suspension was poured into the sand barrier, the soil particles in suspension are transported to the top surface of the barrier by sedimentation and moved to sandy layer by mass flow. The particle removal efficiency depends on the particle concentration in the water, particle density, filtration rate and filter pore diameter (Cleasby, 1990).

The sorption data is plotted in Figure 6 as the percentage of ^{137}Cs sorbed on clays vs. time. The amounts of ^{137}Cs were sorbed by resins of A-51[®], IE-95[®], and EP-9174[®] do not differ significantly from each other, average values of ^{137}Cs adsorbed by zeolite resins are shown in Figure 6. The results show initially rapid adsorption, followed by slower uptake. For zeolite resins, about 95% to 99% of ^{137}Cs in the synthetic groundwater was sorbed after 6 hours with vigorous shaking. The percent ^{137}Cs sorbed did not change significantly during the remainder 474 hours (Fig. 6). The sorption of ^{137}Cs to illite was slower than

to zeolite resins. During the first 6 hours with vigorous shaking, about 76% of ^{137}Cs in the synthetic groundwater was sorbed. A total of 93% ^{137}Cs was adsorbed to illite after 480 hours (Fig. 6).

The desorption results show less than 1% of ^{137}Cs was desorbed from zeolites and about 3% of ^{137}Cs was desorbed from the illite after an additional 20 days with vigorous shaking. This suggests that zeolites and illite are effective sorbent materials. Zeolites sorbed more efficiently than illite.

The unique feature of zeolite group is the presence of a structural channel which provides high CEC (100 to 300 meq/100g) (Zelazny and Calhoun, 1987). The zeolite resins used in this study are basic zeolites. If Cs ions are fixed in the basic zeolites, the structural channel is too narrow to allow other cations exchange with Cs ions (Zelazny and Calhoun, 1987). It was reported that ^{137}Cs was sorbed strongly on zeolite tuffs from Yucca Mountain, Nevada and ^{137}Cs did not release from these tuffs (Thomas, 1987).

Soil erosion is a primary mechanism of ^{137}Cs transport at TA-21. Preventing soil erosion is a possible approach to control the migration of ^{137}Cs . At the top of DP Canyon, ^{137}Cs in Hot Spot area may be fixed using some cover materials because the area of Hot Spot is not large (<100 m²) or deep (< 30 cm). On the slopes of the canyon, ^{137}Cs may be controlled using an erosion control web, such as "Miramat™" which is a three-dimensional web of bonded polypropylene or PVC monofilaments and it can shield the soil particles from rain, wind and surface runoff while it allows natural revegetation to establish (GEOCIV, 1995). At the bottom of the slope, spreading of ^{137}Cs contaminated soil particles may be controlled by a barrier. Laboratory study shows that the sand barrier blocked ^{137}Cs labeled soil particles successfully. Clay minerals of zeolite or illite can be used in the barrier for stabilization of ^{137}Cs .

CONCLUSIONS

At LANL, DP Canyon was subject to radioactive contaminant releases, resulting in the soils containing multiple radioactive contaminants. The results show that ^{137}Cs is a major contaminant in the soil and is mainly associated with silt, clay and sand fractions. Because ^{137}Cs has been "fixed" in the soil, it can not be leached by synthetic groundwater or acid rainwater. Soil erosion is a primary mechanism of ^{137}Cs transport in DP Canyon. It is possible to use a sand barrier to prevent that transport. Zeolites are effective materials for stabilizing ^{137}Cs in the barrier. The results from our studies are in agreement with those reported by previous investigators in Europe, Asia, and North America.

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List of Figures

Figure 1. A sand barrier structure.

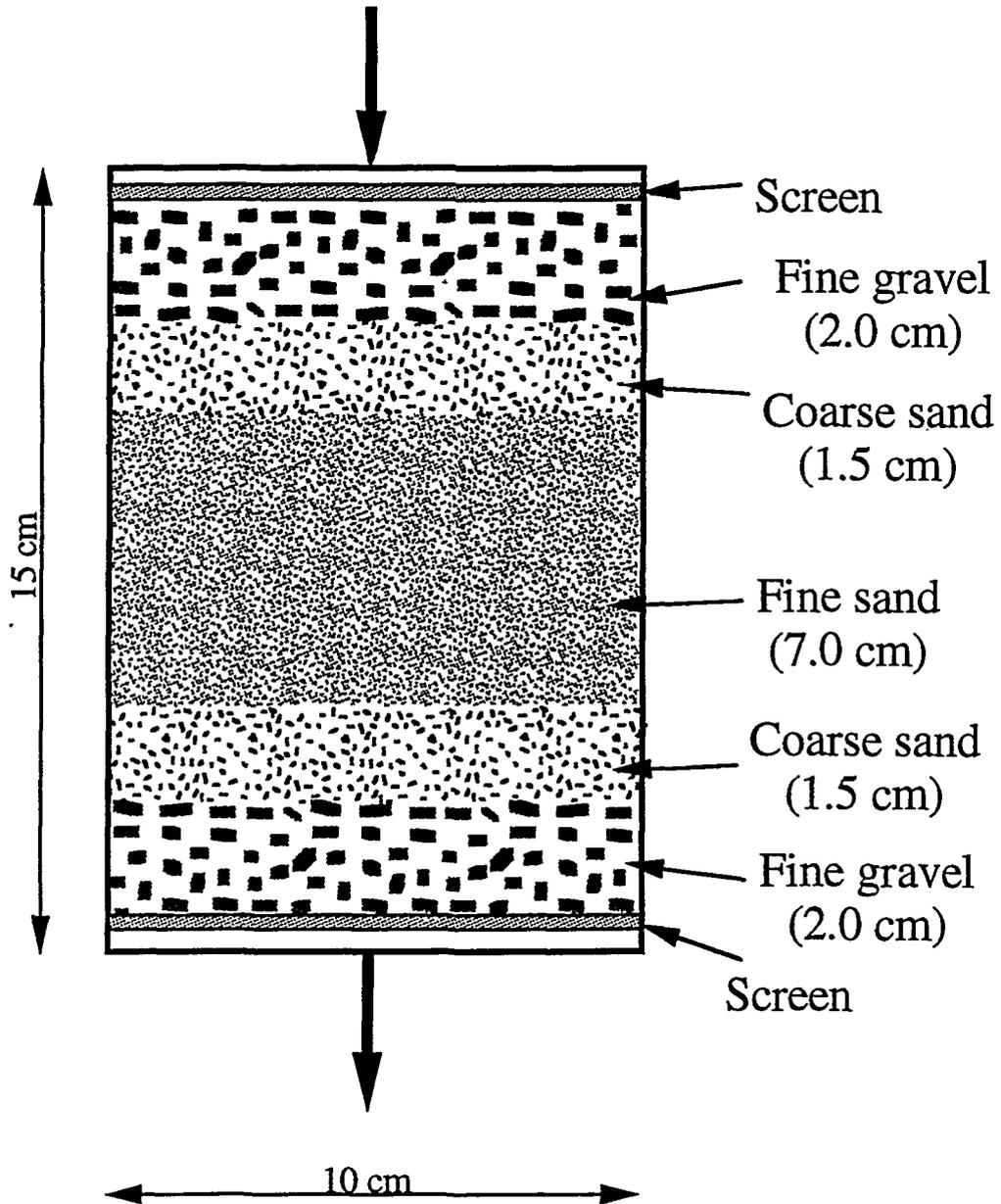
Figure 2. Cesium-137 in air-dry soils, water and pine bark from TA-21-011(k).

Figure 3. Cesium-137 distribution in the different particle-size fractions of the Hot Spot soil from TA-21-011(k).

Figure 4. Cesium-137 in Hot Spot and Sediment soils from TA-21-011(k), pre- & post-leach.

Figure 5. Cesium-137 in liquid and in sand layers of the barrier before and after processes.

Figure 6. Percentage of cesium-137 adsorbed on zeolite and illite.



Sand Barrier

