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Characterization, Transport and Remedial Options for Radioactive Strontium and Cesium Contaminated Sites.

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ABSTRACT

Radioactive strontium (Sr) and radioactive cesium (Cs) are contaminants of major concern because of their ease of entry into biological systems due to their similarity to calcium and potassium respectively. Large areas of the Ukraine are contaminated with these radionuclides as a result of the Chernobyl accident in 1986. Areas both at Los Alamos in the US and at the Semipalatinsk Test Site in Kazakhstan are also contaminated with these radionuclides. Both cesium and strontium are soluble in water but their mobility in natural environments is retarded by their strong interactions with soil organic materials, clay minerals and iron oxides

Here we examine a site at Los Alamos National Laboratory contaminated with ^{137} Cs and ^{90}Sr. Experimental results show that the distribution coefficients for Cs are much greater than those for Sr. This results in much faster dispersal of Sr through water-borne transport pathways. Further Sr is transported by sorption onto colloids providing an additional transport mechanism.

Remedial options for contaminated sites require detailed studies of sorption processes of the local soils. Apart from the drastic measure of removing contaminated soils to a controlled area, movement of contaminants can be controlled by the presence of media that will prevent both particulate movement and also immobilize soluble species. Immobilization occurs through precipitation, sorption and ion exchangers.

Introduction

Radioactive strontium (Sr) and radioactive cesium (Cs) enter the environment through above ground nuclear weapons tests, nuclear accidents and the improper disposal of waste from processing of nuclear fuel rods. They are fission products of great concern because of their ease of uptake into biological organisms due to their similarity to calcium and potassium respectively. Cesium–137 has a half-life of 30 years and Sr–90 one of 28.5 years. The migration of these radionuclides in soils is an important consideration since soils have a close relationship to human life. In addition, together they are responsible for 98% of the thermal energy and 97% of the penetrating radiation in high level radioactive waste.

Large areas of the Ukraine are contaminated with these radionuclides as a result of the Chernobyl accident in 1986 [1]. Of the total release of ~ 50 MCi, 70% of the activity fell on Belarus in 17,000 km^2 of a forested area of Gomel Oblast. In May 1996, it was estimated that 795,200 Ci of ^{137}Cs and 172,900 Ci of ^{90}Sr remained. Areas both at Los Alamos [2] in the US and at the Semipalatinsk Test in Kazakhstan [3], are also contaminated with these radionuclides. Strontium–90 is the third most frequently occurring radionuclide in ground water at the US

Department of Energy facilities.[4] Further, sorption of radionuclides is an important parameter in the design of nuclear waste repositories.

As a case study in characterization and remediation of Cs and Sr contaminated site, we describe here a canyon at Los Alamos National Laboratory. In addition further remedial options will be discussed.

Description and Characterization of Site

At Los Alamos, New Mexico, DP Canyon was subject to radioactive contaminant releases from 1945 to 1978 from the Plutonium Processing Facility that was used for research and production of plutonium [5]. Radioactive effluent was discharged down the steep slope of the adjacent canyon to the north, DP Canyon, and resulted in extensive contamination. Surveys (including an aerial radiation survey) identified a plume of radionuclides including ^{137}Cs, ^{241}Am, ^{239}Pu and ^{90}Sr. These are stored in the thin soils down the side of the canyon and also in the sediments of the stream channel in the canyon bottom. The highest contamination levels, (found to be due to ^{137}Cs) were found on the steep slope of the canyon wall directly below the discharge point [5]. This is illustrated in Figure 1. This site is named PRS 21-011(k).

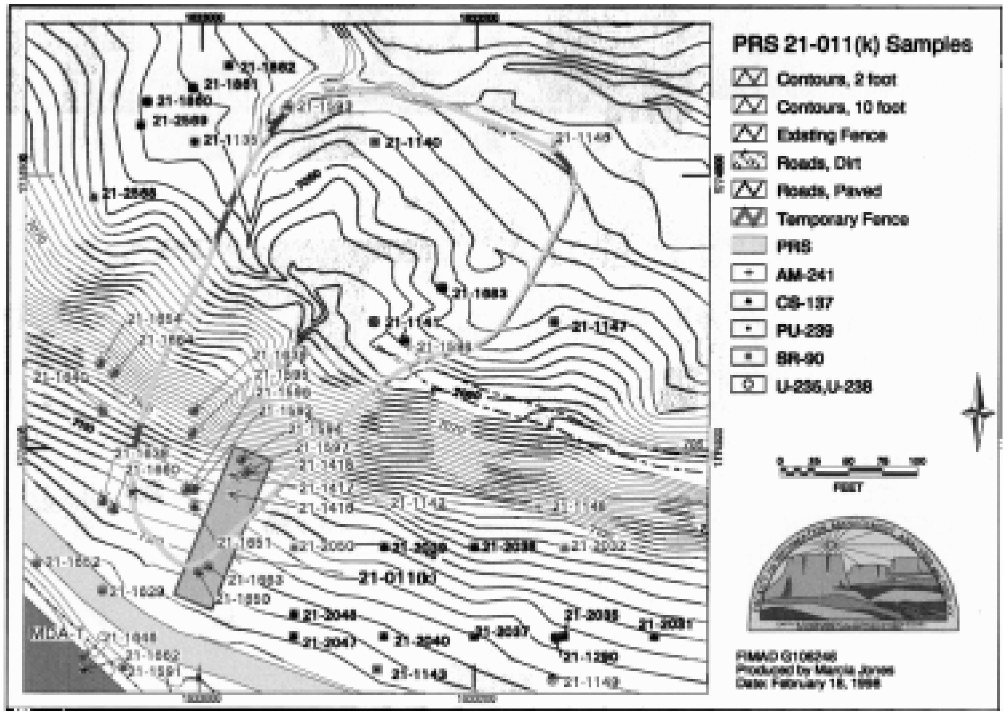

Figure 1. Map showing the location of PRS 21-011(k). Sample points and contours are shown together with the temporary fence enclosing the area of where the dose was calculated to be greater than 15millirem/y.

Transport mechanisms

Both cesium and strontium are soluble in water. However their mobility in natural environments is retarded by their strong interactions with soil organic materials, iron oxides and clay minerals. In general the sorption distribution coefficients (Kd_S, ml g^{-1}) are calculated using the following equation:

$$Kd_S = [(A_oV_o - A_fV_f)/M]/A_f \qquad [1]$$

where A_o is the initial radioactivity of ^{137}Cs or ^{90}Sr per milliliter solution, V_0 is the initial volume (ml) of ^{137}Cs or ^{90}Sr solution in contact with solid samples, A_f is the final radioactivity of per milliliter solution after sorption, V_f is the final volume ($m\ell$) of the solution after sorption, and M (g) is the mass of solids or colloids used in the sorption process.

Experimental results from soils and sediments in Los Alamos County [6] are shown in Table 1. These reveal that the distribution coefficients for Cs are much greater than those for Sr resulting in much faster dispersal of Sr through water-borne transport pathways while Cs is mainly transported with sediments and soils.

Table 1.
Distribution Coefficients for Cs and Sr [6]

	Cs	Sr
surface soils	Kd_S = 160 -1400 ml/g	Kd_S = 15 -70 ml/g
sediments	Kd_S = 80 -1000 ml/g	Kd_S = 10 -40 ml/g
desorption	4-9 %	8 - 30%

It was found that the radioactivity associated with Cs decreased linearly as the soils particle size increased, suggesting that Cs is mainly associated with the silt-clay fractions [5]. The silt and clay fractions of Los Alamos soils are largely montmorillonite and illite formed by weathering of tuff (the volcanic base for Los Alamos County). Monovalent cations such as Cs enter the interlayer of illite preferentially and become irreversibly bound. Thus Cs bound to these clay minerals is not leached by rainfall, as was experimentally verified [5].

There is also evidence that Cs is transported by colloids [7]. Snowmelt from the base of the of DP canyon where the effluent was discharged was filtered through successively smaller filters (0.45-µm, 0.2-µm 0.05-µm nylon membranes) which removed all the radioactivity as is illustrated in Figure 2. The results show that most particles ranged from 0.05 to 0.45-µm. The dry weights of the particulates collected are 632.7 mg L^{-1} with no filter, 554.3 mg L^{-1} with 0.45-µm, 521.5 mg L^{-1} with 0.2-µm and 507.2 mg L^{-1} with 0.05-µm. This indicates that the ^{137}Cs is associated with the fine particles including colloids.

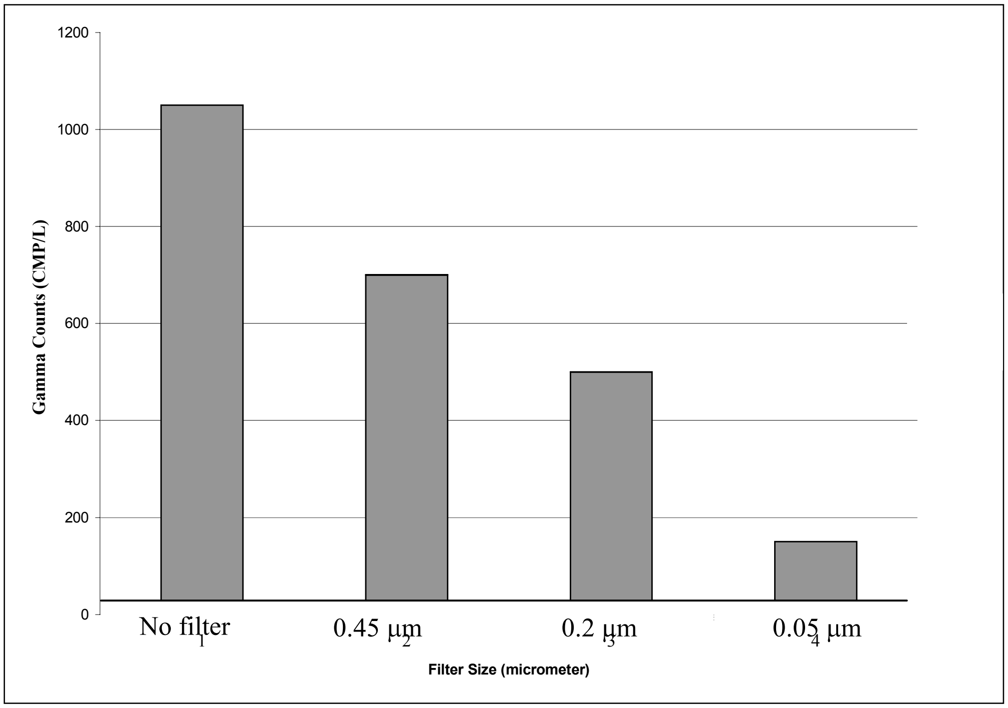

Figure 2. Radioactivity of snow melt water as a function of size of filter.

Conversely, ^{90}Sr desorbs from soils and sediments continuously and has a fast migration rate relative to other contaminants [8]. A radiological survey in 1995 found that as much as 526 pCi L^{-1} of ^{90}Sr exist in shallow alluvial wells in DP Canyon. Further Sr is transported by sorption onto colloids providing an additional transport mechanism. However the presence of soluble cations, such a sodium and calcium will affect the sorption properties. Figure 3 shows the sorption of ^{85}Sr (used as a laboratory surrogate for ^{90}Sr) onto two colloids using deionized water and natural water from DP Canyon. Calcium in DP canyon water competes successfully for the sorption sites when using silica colloid but was unaffected for the smectite (calcium montmorillonite) colloid.

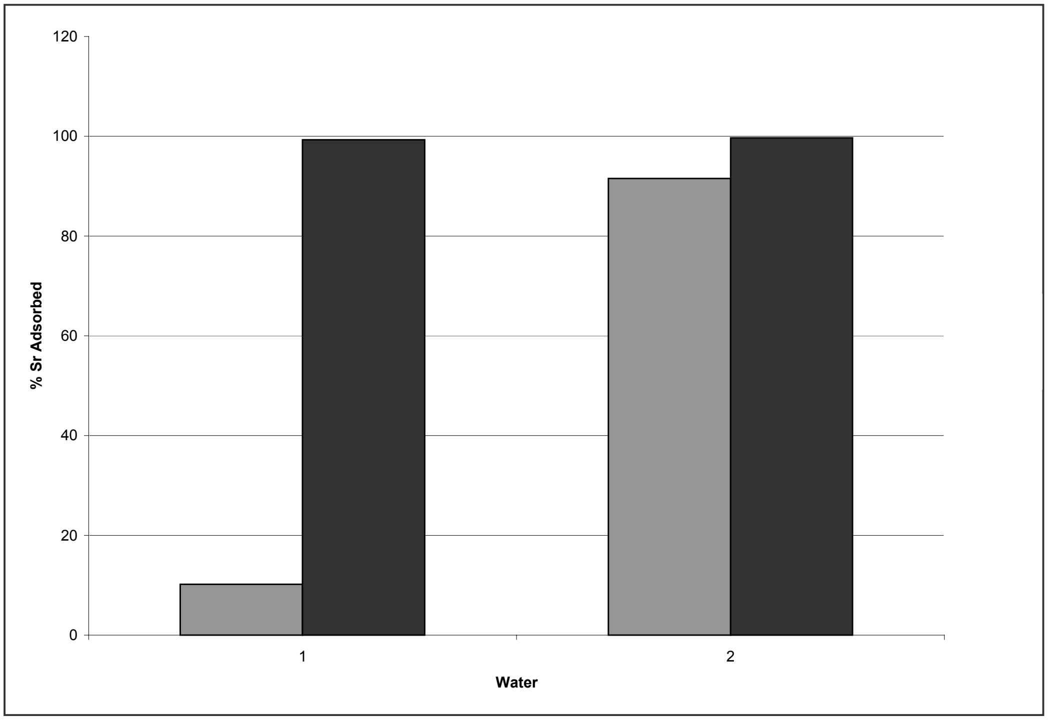

Figure 3. % ^{85}Sr sorbed onto colloids of Silica-ST-ZL (gray) and Smectite (black) after 7 Days. 1 –DP Canyon Water 2- Deionized water

Entry into Biological Systems

Plants

A common shrub that grows in the semi-arid climate of New Mexico is the chamisa (chrysothamnus nauseosus) with roots growing to depths of 200 cm. Table 2 shows the uptake of ^{90}Sr into a chamisa bush situated over a former low–level waste disposal area. Clearly not only does chamisa sorb Sr, it also redistributes it through plant detritus on the ground surface.

Table 2.
^{90}Sr in chamisa in a former liquid waste disposal area [9]

		Sample 1 (pCi/g dry ash)	Sample 2
Chamisa	Top growth	90,500.0	5,930.0
	Root growth	40,600.0	4,630.0
Background	Plant top growth	0.3	
	Root growth	0.0	
Soil	Understory	191.4	37.5
	Interspace	12.6	16.0
Background	Understory	0.1	
	Interspace	1.40	

Animals.

A study of uptake of ^{137}Cs in the wild rodents was undertaken in 1978 [10]. In the stream channel from the outfall that received the effluent waste in DP Canyon. Lithium fluoride thermoluminescent dosimeters (TLD) were implanted subcutaneously. The ^{137}Cs concentrations and absorbed radiation doses are shown in Table 3. The difference in dose reflects the difference in mobility and habitat for each species, the western harvest mouse remaining close to the stream channel.

Table 3.
Mean ^{137}Cs concentrations and mean estimated radiation dose.[10]

Species	Number of mammals	^{137}Cs (pCi/g wet carcass)	Total dose* mrads/day
Western harvest mouse	8	41	26
Deer mouse	4	6.6	8.3
Piton mouse	5	1.0	1.9
Chipmunk	13	1.6	1.4

* Measured subcutaneously with implanted TLD

Remedial Actions Taken

Because DP Canyon is favorite place for outdoor activities, the first priority was to render it safe for hikers and other recreational users. This was accomplished in the summer of 1996 when approximately 300 m^3 of ^{137}Cs-contaminated soil with γ activity >100,000 cpm was removed leaving the remaining site at < 100,000 cpm (~ <400pCi/g). This removal included the hot area shown in Figure 1. The soil was trucked to a low-level radioactive repository at Los Alamos.

An additional remedial action was taken to stop the spread of ^{137}Cs-contaminated soil by placing straw bales over the area where the soil washes down the canyon wall during the short period when intense thunderstorms occur. These straw bales prevent the movement of particulate matter. The average γ activity was reduced by 59% showing that the straw bales blocked the coarse soil particles but not the fine ones [7].

Possible Remedial Options

Cesium-137 contaminated soil particles with size < 2.00 mm have been shown to be blocked by a 5-layered sand barrier [5]. No detectable ^{137}Cs was found either below the second layer of the barrier or in the leached water. Movement of soluble contaminants, such as ^{90}Sr, can be controlled by the presence of media that will form immobilize ^{90}Sr. Such media are shown in Table 4.

Table 4.
Immobilization mechanisms

Immobilization Mechanism	Example
Precipitation	1. Phosphates such as apatites (e.g. $Sr_5(PO_4)_3(OH)$, log Ks = -51.3) [11] 2. Organic phosphates (phytic acid) [12] 3. Barium Sulfate (coprecipitation) [13]
Ion Exchange and sorption	clays, zeolites, geothite, beidellite, clinoptilolite [14] silico-titanates [15]

Richards Barriers.

A Richards barrier [16] acts in an unsaturated hydrogeologic system to prevent advective flow of water down through a barrier. It incorporates two or more layers with contrasting capillary suction potentials and hydraulic conductivities such as a fine layer of sand with small pore size and high capillary potential and also a coarse layer of gravel with large pore size and low capillary potential. The hydraulic conductivity of the sand results in water draining down the layer boundary. This is illustrated in Figure 4. Such barriers have been found to be effective over thousands of years at prehistoric burial sites in Japan [17].

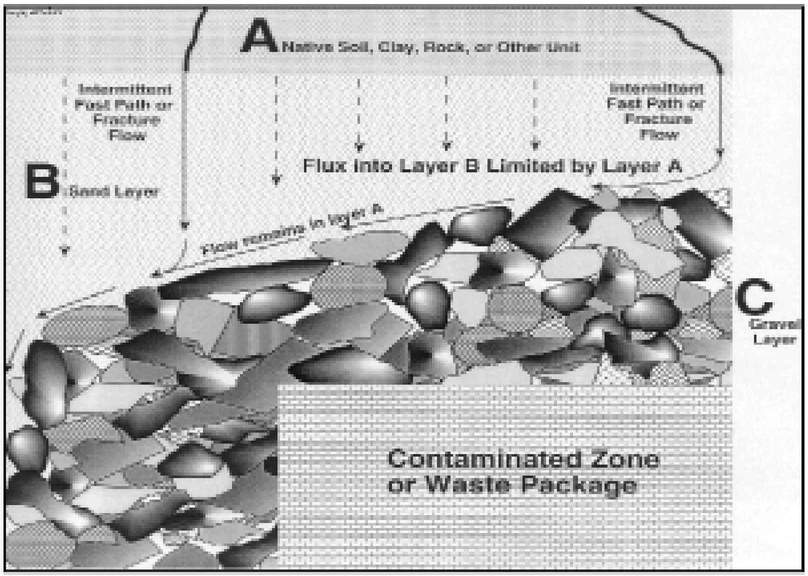

Figure 4. Features of a Richards Barrier (Gravel Barrier) for hydrologic isolation in the vadose zone [16]

Summary

By detailed examination of one site contaminated with ^{137}Cs and ^{90}Sr, it is possible to reach some general conclusions:

- Even when ^{137}Cs and ^{90}Sr originate from the same source they will be unevenly distributed due to different transport mechanisms. This is found at all contaminated sites, including those in the Ukraine.
- ^{137}Cs and ^{90}Sr are both taken up in biological systems
- they can be controlled by barriers and immobilization
- each contaminated site presents a unique combination of hydrology, chemistry and geology

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