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INTRODUCTION

At Hanford, intermediate-level, liquid wastes with radioactivity concentrations in the range 0.1 to 10 Ci/l[1] are stored underground in carbon steel-lined, reinforced concrete tanks. More than 100 such tanks with capacities ranging from 1.9×10^6 to 3.8×10^6 liters each have been used to store the wastes arising from more than 25 years of irradiated fuels reprocessing.

Since the beginning of nuclear operations at Hanford, finding improved technology for confining radioactive wastes has been a primary objective of research and development programs.

As the volume of accumulated liquid wastes grew, emphasis on the development of waste management technology was increased. These efforts culminated in a comprehensive program to decrease the radioactive content in plant effluents and the construction of waste processing facilities to convert high-level wastes to solids. This program, which is now

[1] Hanford classifies wastes in this category as a low-heating, high-level waste. The radioactivity range of the waste falls within Category 4, intermediate-level wastes, as defined by the IAEA.

operational, includes the conversion of the high salt, intermediate-level, liquid wastes to salt cakes for storage in the existing underground tanks; the conversion is accomplished by thermally concentrating the wastes until the salts crystallize in the tanks when the wastes are cooled. The wastes are then to be dried to remove excess moisture. Very high-level wastes are processed to remove the long-lived heat emitters, strontium-90 and cesium-137, so that the residual wastes can be converted to salt cakes following decay of short-lived nuclides. When facilities are available in 1974, the isolated strontium and cesium will be converted to dry fluoride and chloride salts, respectively, and packaged separately in containers of high integrity for interim cooling and storage in water-filled basins. The design concepts of the packaging facilities were described by Shaw previously.[1]

Godfrey, Hanson and Smith[2] in a companion paper prepared for this symposium, describe the in-tank solidification (ITS) process whereby the intermediate-level liquid wastes are converted to salt cakes. In this paper, we discuss the factors which were considered in selecting the ITS program at Hanford, the three phases involved in creating and storing the salt cakes, and preliminary conclusions regarding the safety of the approach for long-term storage of the wastes. Research and development programs being performed to verify these preliminary conclusions are reviewed also.

IN-TANK SOLIDIFICATION PROGRAM

The ITS program was selected over other waste solidification processes for use at Hanford after carefully considering many factors.

- The underground storage tanks were nearing the end of their useful life as reliable liquid containment vessels. A method was needed to reduce the mobility of the radioactive wastes rapidly.

- Since national criteria and techniques for long-term storage of radioactive wastes had not yet evolved, the chosen waste treatment and storage method should not preclude future actions, if desired.

- The varied composition of the wastes which arose from the many different plants and processes employed at Hanford, would require a conversion process operable for all waste types.

- About 230,000 tonnes of contaminated salts were contained in the stored liquid wastes. The conversion process must be suitable for large-scale operation to solidify all the wastes within a few years.

- The wastes contained high concentrations of soluble sodium salts (carbonates, nitrates and nitrites) which could not be readily converted to an insoluble form without a large increase in volume when compared to soluble solid forms.

- The characteristics of the Hanford site are favorable for the storage of a solidified waste. The climate is semiarid, with essentially no percolation of water from natural precipitation to the water table more than 60 meters below ground surface.

- The existing tanks could reasonably be used for storing the contaminated salts provided the tanks need not be regarded as containment vessels.

The deciding factors for selecting the ITS program were:

- Most of the water could be removed from the salt wastes rapidly, thereby achieving prompt improvement in storage safety.

- Storage of salt cakes in tanks was judged to be safe for the near-term future, and such storage would not preclude future actions, i.e., the wastes would be retrievable.

The salt cake formation-and-storage program is being implemented in three phases. In the <u>first</u> phase, water is removed by evaporation, and the salts allowed to crystallize from the concentrate upon cooling. Salt cakes are formed in the tanks. In the <u>second</u> phase, the salt cakes are allowed to cool for several years while temperatures, salt stability and concentration of gases produced by radiolysis and/or chemical decomposition are monitored. With sufficient information to assure long-term stability, the salt cakes are to be prepared for extended storage by adding material to fill the void spaces in the tanks and by stabilizing the ground surface. The <u>third</u> phase involves long-term storage with occasional surveillance. As discussed later, storage of salt cakes at Hanford with minimal surveillance is believed to be safe, at least for decades and perhaps for centuries.

Phase I of this program is operational, and Phase II is being started. Each of the three phases is described in more detail below.

Phase I - Solidification of Liquid Wastes

The process and facilities used to solidify the liquid wastes are described by Godfrey, Hanson and Smith[2] and by Kirkman and Godfrey.[3] This phase was begun in 1965, and the three currently operating evaporators have removed more than 125×10^6 liters of water while forming about 10×10^6 liters of salt cake; another 10×10^6 liters of salt cake would probably form if the heated solutions were allowed to cool. Operating costs range from $0.03 to $0.08 per liter of water removed, depending upon which of the three evaporators is considered. The electric immersion heater evaporator[2,3] is the most efficient, while the hot-air sparger evaporator[2,3] is the least efficient. A fourth evaporator is planned to provide sufficient capacity to solidify all intermediate-level liquid wastes by the 1976 goal date.

Phase II - Cooling and Monitoring of Solidified Wastes

As each tank receives its quota of salt cake, steps will be taken to assure its safety for extended storage. The solidified wastes will be allowed to cool by dissipating the sensible heat through the surrounding soil. Air may be blown over the surface of the wastes to remove excess moisture and hasten cooling. Alternatively, moisture may be removed by adding a desiccant. During this cooling and drying period, temperatures in the salt cake and concentrations of gases formed by radiolysis or chemical action will be monitored. Based upon the data obtained, projections will be made as to temperature profiles and rates of gas formation for each tank as a function of time. When enough data have been obtained to project future conditions with confidence, the tank dome will be filled with sand or grout, the tank will be disconnected from all operating systems, and the ground surface will be stabilized with a cover of rock and gravel to prevent wind erosion. Stabilization of the tanks and ground surface is tentatively scheduled to begin in the 1980's.

After completing Phase II, a tank farm consisting of up to 18 tanks would appear as sketched in Figure 1. The tanks would be isolated from the surface and from one another. More than 9 meters of sand, soil and rock would cover the salt cake. The minimum distance from the bottom of the tanks to the water table would be 45 meters.

Phase III - Long-Term Storage of Solidified Wastes

The third phase would involve long-term storage with occasional surveillance to ensure that safe storage conditions are maintained. Wells which are located around the tanks and penetrate the groundwater table would be monitored periodically to audit the continuing safety of the system.

ALTERNATIVE STORAGE MODES

Alternative approaches to the storage of the radioactive salt cakes are under study also, on the premise that some reason may be identified for wanting to avoid long-term storage near the ground surface. It appears possible, for example, to remove and transport the radioactive salts to a salt mine for long-term storage. The magnitude of the move (230,000 tonnes of salt, plus shielding to be transported at least 1500 kilometers) and the risk of accidental contamination spread argue against this alternative. A second possibility involves the mining of caverns in the basalt perhaps one half to one kilometer beneath ground surface at Hanford. Layers of competent rock up to sixty meters thick exist at these depths, and these locations are effectively isolated from near-surface groundwaters. If found to be feasible, the wastes could be transferred to the caverns as slurries or as packaged solids. Test wells are being drilled to evaluate the geology and hydrology of the region at depth.

LONG-TERM SAFETY CONSIDERATIONS

The safety of storing radioactive salt cakes in underground tanks depends primarily on the characteristics of the natural storage environment, on the characteristics of the material to be stored, and on the ability of man to prevent the intentional or accidental movement of the radionuclides.

Site Characteristics

Characteristics of the site which are pertinent to safe long-term storage of wastes include those of climate, water and radionuclide migration patterns, geology, seismicity, and geography.

Climate

The Hanford climate can be characterized as semiarid and desert-like. The average annual precipitation for the past fifty-six years is 15.5 cm. Figure 2 shows the annual precipitation data for the period 1913 to 1969, plotted on log-probability paper. The data indicate that the likelihood of exceeding 45 cm of precipitation is only once in 10,000 years. Climatological and geologic evidence suggests that the area has enjoyed a semiarid climate for the last 10,000 to 20,000 years (since the last ice age). Precipitation can be expected to remain low as long as the Cascade Mountains to the west remain.

Temperatures in an average year exceed 32° C on about 60 days and are below 0° C on about 116 days. During a recent three-year period, the average wind velocity at 60 meters above the ground was equal to or greater than 8 kilometers per hour 87 percent of the time. Moderate to high winds are frequent throughout the year.

Moisture and Radionuclide Migration in Soil

A primary deterrent to movement of the radionuclides from the salt cake is the lack of water movement as a liquid in the storage zone. The water table ranges from 60 to 100 meters below ground surface where the tanks are located. Groundwater recharge occurs mainly in the highlands to the west of Hanford. Because of the semi-arid climate, the soil above the water table is very dry. The local topography effectively precludes the possibility of flooding from torrential rains or blockage of the Columbia River.

Current data indicate that no natural water percolates to the groundwater in the subject areas. Studies have shown that tritium in precipitation has not migrated from the ground surface more than 6 meters into the soil since nuclear weapons testing in the atmosphere began about 17 years ago. This conclusion is illustrated in Figure 3 where the tritium concentration in soil water is shown as a function of depth. Similar studies in the Spokane area about 250 kilometers to the north indicate that, under similar conditions, percolation of rainwater to the groundwater does not occur with 50 cm of rain per year. Other studies indicate that the net migration of water is upwards from the water table to the atmosphere. This water must be migrating in the vapor phase so that transportation of radionuclides is not possible.

A detailed study of groundwater flow beneath the waste tanks has culminated in the development of a computer model capable of predicting groundwater flow paths and travel times to the Columbia River. Changes in water table elevations for postulated groundwater recharge mechanisms can be determined also.

Paralleling the studies to determine moisture movement are studies to characterize the factors which influence the movement of radionuclides in the soils at Hanford. Most of the technology has been obtained through 25 years of experience in disposing low-level wastes to ground. A paper presented at a previous IAEA symposium describes some of this technology.[4] Continuing field and laboratory studies are in progress to develop a

3-dimensional waste transport model capable of predicting the distribution and movement of radionuclides with water flowing through the soils as a function of time. This model will be useful in determining the long-term movement of radionuclides from the salt cakes in the improbable event that water is able to penetrate the storage sites.

Geology

Hanford lies near the center of the Columbia River Basin, which was formed millions of years ago by many sequential flows of basalt, a dense, hard rock formed by the solidification of volcanic lava. More than 100 basalt flows were identified[5] in an exploratory well more than three kilometers deep near the edge of the Hanford site. The individual layers of basalt are from one to sixty meters thick and are interspersed with extensive deposits of volcanic ash, tuffs, sands, silts and clays.

Overlying the basalt are layers of semi-consolidated sediments (the Ringold formation) and unconsolidated sands, gravels and silts. The semi-consolidated sediments were deposited at a time when the region was inundated by a large lake. The unsorted sands and gravels were carried into the area by glacial floodwaters during the close of the Ice Age 10,000 to 20,000 years ago, and sands and silts have since been deposited by wind action. The tanks containing the salt cakes are located in the sands and silts above the Ringold formation and the water table.

Seismology

The area is in a region of moderate seismicity, verging on minor seismicity.[6] The ground has not been subjected to more than weak ground shaking during historical times. The possibility of surface ground rupture due to upward propagation or displacement along any fault in the area is believed to be remote.[7]

A program to estimate the magnitude of the maximum potential earthquake for the Hanford region, and to characterize earthquake-induced ground motion in the rocks and sediments is underway. A system of geophones has been placed around the Hanford site to determine precise locations and focal depths of microearthquakes. Geological and geophysical studies of purported fault zones are in progress in order to prove or disprove the existence of faulting. Results of these studies should aid in estimating the probability of experiencing an earthquake of sufficient magnitude to expose salt cake to the atmosphere.

Geography

Hanford is located in a region of low population density. The salt cake storage sites are more than 40 kilometers from the nearest population center (Richland, Kennewick and Pasco, Washington) more than 45 meters above the water table, and 9 to 24 kilometers from the Columbia River along the present groundwater flow paths.

CHARACTERISTICS OF SOLIDIFIED WASTES

The salt cake is principally composed of soluble sodium carbonates, nitrates and nitrites. No evidence of chemical or radiolytic instability has been noted in about eight years in a 3.8 million liter tank filled with a radioactive salt cake quite similar to those now being created. The radionuclides, excepting cesium-137, are not readily removed from the alkaline waste by leaching with water. Methods to reduce the solubility of the radionuclides are being developed.

SAFETY ANALYSIS

The objective for long-term storage of the radioactive salt cakes is to isolate, and confine as long as necessary, the radionuclides to the extent that man and his environment will in no way be exposed to deleterious quantities.

The conversion of the waste liquids to salt cakes is the first step in achieving this objective. In this immobilized state, the migration of radionuclides would require the influence of some force outside the waste itself. Results of a preliminary safety analysis[8] indicate that there is no identifiable natural force that would cause the movement of the immobilized radionuclides to the atmosphere or to the groundwater. In the storage site, the wastes would not be contacted by indigenous plants, animals, insects, or other such life forms. Catastrophic flooding is precluded by the terrain, and a doubling or tripling of the rainfall rate would not be sufficient to produce a net downward migration of water.

In the future, environmental conditions could change. Such changes would likely develop over a period of years, however, and actions could then be taken to retrieve, reprocess and/or relocate the salt cakes before a significant hazard could develop.

All available data indicate that, so long as man restricts land use and maintains minimal surveillance, the storage of salt cakes in underground tanks at Hanford is safe. We believe that this storage mode is safe at least for decades, and possibly for centuries.

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LIST OF FIGURES

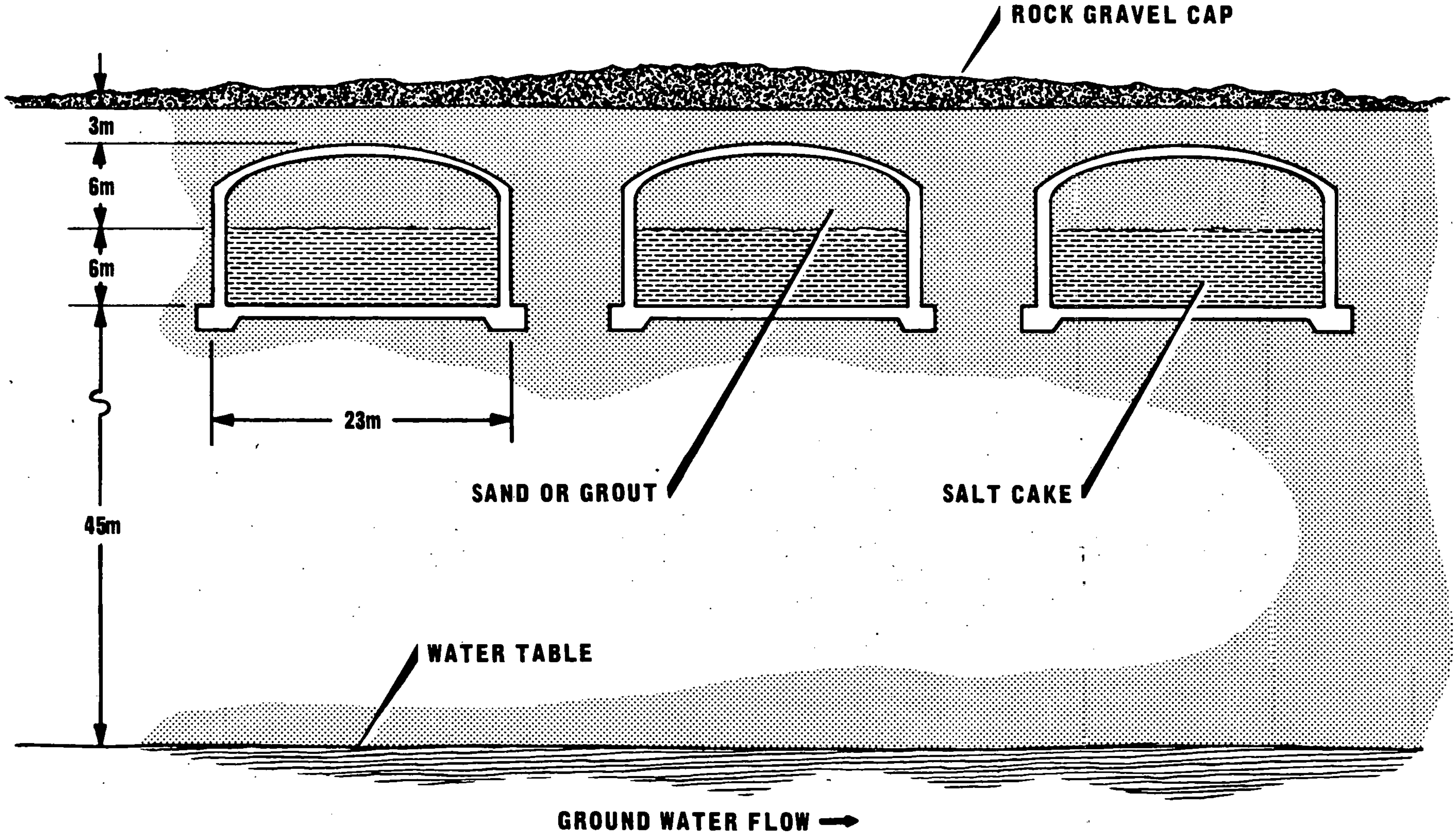

Figure 1. Long-Term Storage of Radioactive Wastes as Salt Cakes in Tanks

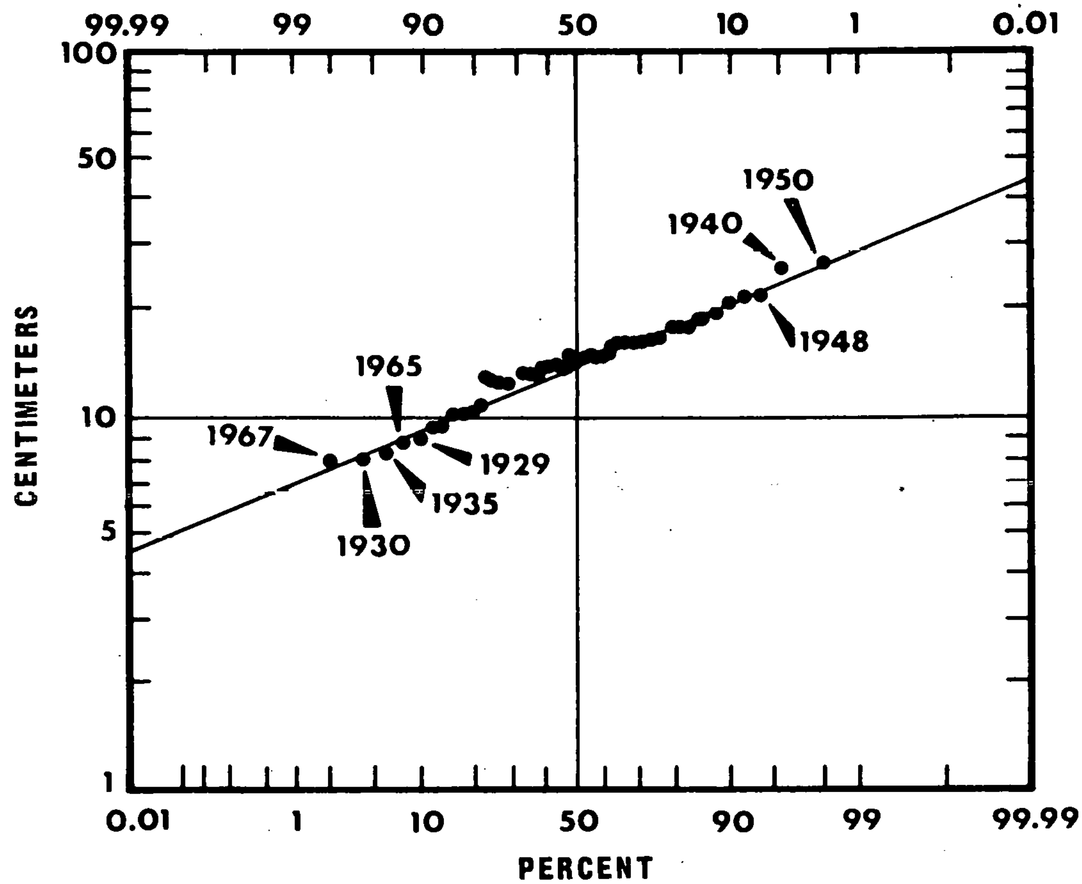

Figure 2. Total Annual Precipitation (1913-1969)

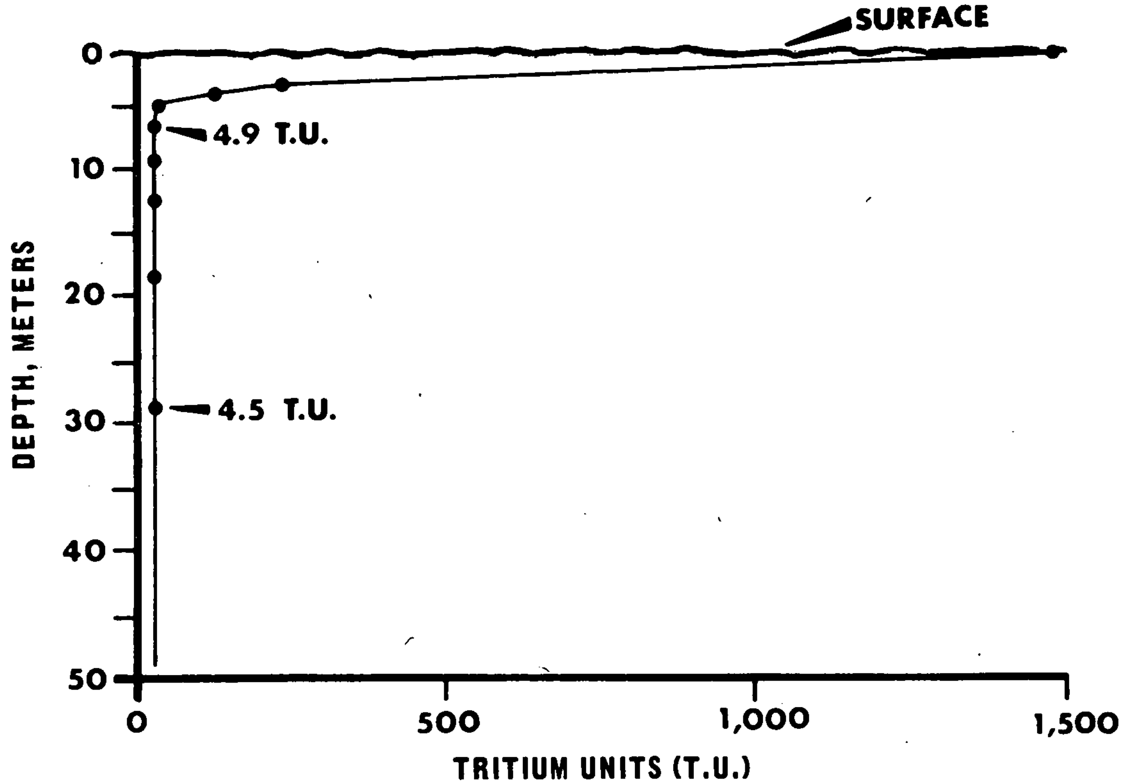

Figure 3. 1969 Tritium Distribution in Soil Moisture