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A REVIEW OF STUDIES ON SOIL-WASTE RELATIONSHIPS ON THE
HANFORD RESERVATION FROM 1944 to 1967

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A REVIEW OF STUDIES ON SOIL-WASTE RELATIONSHIPS ON THE HANFORD RESERVATION FROM 1944 to 1967

INTRODUCTION

Hanford chemical processing plants have been in operation for more than twenty years, and during this period have disposed large volumes of waste liquids to the Hanford sediments. Because the groundwater and sediments at this major nuclear facility have been closely monitored and investigated, a unique opportunity exists to assess the effects of these disposal practices on the Hanford soil environment. This paper discusses the evaluation of the reactions of radionuclides with the Hanford "soils."

In addition other pertinent research at Hanford and other sites are reviewed and summarized, and those areas which have not been adequately investigated are defined and additional studies suggested. It is expected that this and other reviews will be very useful in future waste management planning.

WASTES AND DISPOSAL PRACTICES

Any facility established by man will have some effect upon its environs, and standards as to what is an acceptable level of environmental change need to be established. In the case of the Hanford Project, maximum permissible concentrations (MPC) of radioactive materials for drinking water and air established by the National Bureau of Standards are the standards against which the operational practices have been judged in the past (National Bureau of Standards Handbook 69). AEC Manual, Chapter 0524, Annex 1, Table 2, Column 2 is the present operating guide for Hanford. The standards prescribed by the AEC are essentially 10% of the MPCs of Handbook 69 with one exception; ⁹⁰Sr, which is 1/3 of the MPC listed in Handbook 69. This review is concerned with the large volumes of liquid wastes which have been discharged to ground.

Liquid wastes are divided into high, intermediate, and low level categories according to their content of radioactive emitters. At Hanford, low level wastes are those which contain mixed fission products in liquids at levels less than 5×10^{-5} μCi of β emitters per milliliter.⁽²⁾ These wastes are discharged to natural, low depressions in the ground (swamps) and allowed to percolate to the groundwater. Low level wastes are below the MPC upon discharge and further decontamination is obtained during reaction of the wastes with the sediments during percolation. Intermediate level wastes which contain mixed fission products in liquids at levels of 5×10^{-5} to $100 \mu\text{Ci}$ of β emitters per milliliter are disposed to cribs. Cribs are underground structures from which the solution percolates through the sediment (soil) to the groundwater. Enroute to the groundwater the solution components react with the sediment and much of the radioactive material is sorbed and remains in the sediment. Limited amounts of special intermediate level wastes are also disposed to the ground in trenches on a specific retention basis. Specific retention is the volume of waste liquid that can be disposed to the soil and held against the force of gravity by the molecular attraction between soil particles and the surface tension of water.⁽³⁾ Wastes so disposed will not reach the water table if no additional water is added to the soil profile. Thus, this waste is retained within the pores of the sediment, and retention does not depend upon any sorption reaction for decontamination. Wastes that fall into the specific retention category contain radionuclides that are usually complexed and would thus fail to react with the sediments. High level liquid wastes which contain mixed fission products at levels where the gross β emitters are at levels greater than $100 \mu\text{Ci}$ per ml. These high level wastes are currently stored in underground tanks.

The operating practice at Hanford in the past has been to discharge certain intermediate level radioactive wastes to cribs until the concentration of any nuclide with a half life greater than three years reaches a concentration equal to the AEC guide for drinking water.⁽⁵⁾

Monitoring wells are drilled through the sediments adjacent to each crib to detect the presence of radionuclides in the groundwater and to insure that the AEC drinking water guide is not exceeded.

Disposal of radioactive wastes to the ground is not in reality disposal, but rather storage to allow time for natural radioactive decay to render the radionuclides innocuous. Sorption of radionuclides by the Hanford sediments allows time for this decay to occur.

WASTE DISPOSAL ENVIRONMENT

To insure safe ground disposal, the texture, structure, depth, and mineralogy of the sediments as well as the areal climate need to be considered. The ground disposal portion of the Hanford project is located on a giant bar-shaped plateau of unconsolidated and semi-consolidated glaciofluvial sediments (Figure 1). The plateau rises 200-300 feet above the Columbia River which forms the area's northern and eastern boundary. The sediments can be subdivided into: 1) the thick glaciofluvial sediments; 2) a thin "Palouse Soil"; 3) the Ringold Formation (Figure 2). These sediments are underlain by the Columbia River basalts having a known thickness of greater than 10,500 feet.⁽⁶⁾

Texture, Structure, and Depth of Hanford Sediments

The upper glaciofluvial sediments are 100-300 feet thick and overlay the "Palouse Soil" and Ringold Formation. These sediments are composed largely of unconsolidated coarse sands, gravels, and boulders combined with lesser and variable amounts of silts and clays,⁽⁷⁾ and are relatively permeable allowing disposal of large volumes of liquid per unit time.⁽⁸⁾ The "Palouse Soil" underlays the glaciofluvial sediment in the western portion of the disposal area. This sediment is composed of some fine sand, a large content of silt, and some clay, and is less permeable than the overlying glaciofluvial sediments.⁽⁹⁾ Underlying the "Palouse Soil" in the west and glaciofluvial sediments in the east is the Ringold Formation. The Ringold Formation can be subdivided into upper, middle, and lower zones. The lower zone consists of significant quantities of a blue-green

FIGURE 1. Map of the Hanford Project

FIGURE 2. Geologic Cross Section of the Hanford Project

clayey-silt and is locally known as the blue-clay zone;⁽⁶⁾ the middle zone is mainly comprised of coarse materials and locally referred to as the conglomerate zone and the upper zone consists of tan to buff colored silts called the silt zone. There is a marked contrast in permeability between the Ringold Formation and the glaciofluvial sediments.⁽⁸⁾

The water table in the eastern and northern parts of the reservation is generally near the boundary between the more permeable glaciofluvial and less permeable Ringold sediments. Liquid wastes discharged to the percolate ground through the glaciofluvial sediments, the "Palouse Soil," and the Ringold sediments, respectively. Upon entry into the groundwater, wastes move slowly toward the Columbia River.⁽¹⁰⁾

Mineralogy of the Sediments

Mineralogy of the glaciofluvial, "Palouse Soil," and Ringold sediments is based on a limited number of samplings and is not completely defined.^(11, 12) Silt and clay fractions are generally considered to be the active (reactive) portion of any soil based on their relatively large surface area, exchange capacity, and mineralogical composition. The silt fraction of the glaciofluvial sediments, "Palouse Soil" and Ringold sediments are composed of quartz, feldspar, and mica with quartz and feldspar predominating. The clay fraction of the glaciofluvial sediments is somewhat variable and contains quartz, feldspar, montmorillonite, chlorite, vermiculite, and mica. The clay fraction of the "Palouse Soil" contains calcite, mica, chlorite, montmorillonite, quartz, and a trace of feldspar. Montmorillonite dominates the clay fraction of the Ringold sediments with traces of quartz, feldspar, mica, kaolinite, and chlorite. Montmorillonite has a high exchange capacity and is present in all of the sediments. The presence of quartz and feldspar in all the silt fractions and abundance in some of the clay fractions are an indication that the sediments have undergone relatively minor chemical weathering.⁽¹³⁾ The physical, chemical, and mineralogical characteristics of three representative Hanford soils were determined as a part of this study. These data are presented in Appendix A.

Climate

The Hanford climate is one of hot summers, cool winters, low precipitation (6.2 in./year), moderate windiness, and low relative humidity (54%).⁽¹⁴⁾ The relatively warm climate, wind, low rainfall, and low relative humidity limit the percolation of precipitation and result in minimal leaching of radioactive wastes from the vadose zone.

Thus, the Hanford project is a desirable site for ground disposal of radioactive wastes due to the deep-permeable sediments, great depth to groundwater, adequate sorption capacity, favorable climate, and remoteness (both in time and space) from locations of potential radionuclide uptake.

WASTE DISPOSAL ZONES

The Hanford sediments can in general be divided into three zones on the basis of their desirability for the disposal (storage) of radioactive wastes. The three zones are: 1) the vadose (unsaturated) zone above a depth of 20 feet (upper vadose zone); 2) the vadose zone below 20 feet (lower vadose zone); 3) the saturated zone. The upper vadose zone is usually least desirable for the disposal of radioactive wastes because of the relatively high probability of plant or animal (organism) contact either directly or after possible dissemination by wind, water, and food chains. Thus, only low level wastes are deliberately disposed to this zone. The lower vadose zone is the most desirable waste disposal zone. As long as radionuclides disposed to this intermediate zone do not move there is a minimal chance of organism contact. Much of the research effort at Hanford has thus been devoted to obtaining the maximum soil retention of radioactive wastes and to the prediction of movement of wastes in this zone. The saturated zone (below the water table) is the second most desirable zone for radioactive waste disposal. Organism contact will normally not occur with wastes in this zone prior to the movement of the groundwater to the Columbia River which is a distance of over 10 miles, requiring several years travel time.⁽¹⁰⁾ The general arrangement of the remainder of the discussion section of this report will be based on these three waste disposal zones.

SUMMARY

The literature regarding effects of the liquid waste disposal operations at Hanford on the soil environment was summarized. This summary is divided into studies pertaining to: 1) the vadose zone below a depth of twenty feet (lower vadose zone); 2) the saturated zone; 3) the vadose zone above twenty feet (upper vadose zone). The division is based upon the relative desirability of these three zones for nuclear waste disposal (storage).

Liquid nuclear wastes have been safely discharged to the lower vadose zone for over 20 years. Migration of these wastes from the lower vadose zone by the mechanisms of diffusion, leaching, and particulate transport is discussed. It is concluded that: 1) little movement of sorbed radionuclides by the above mechanisms within ten half-lives of even the long-lived radionuclides occurs; 2) the sorbed radionuclides can only be removed from this zone by a cataclysmic occurrence in which the sediment in the zone and the entrained wastes are both physically removed.

Field studies show that essentially all of the long-lived radionuclides disposed to the ground have been sorbed and remain in the upper levels of the lower vadose zone. Areas of research relating to the lower vadose zone requiring further study include the completion and testing of a quantitative transport model, and the determination of sorption due to the precipitation of SrCO_3 in the presence of CaCO_3 .

The saturated zone is the second most desirable zone for radioactive waste disposal. Since water disposed to the saturated zone takes several years to travel from the disposal areas to the Columbia River a finite time period for radioactive decay exists. Travel times considerably longer than that of the water itself can be anticipated for radionuclides which are sediment-sorbed such as ^{90}Sr and ^{137}Cs . The saturated zone is a less desirable zone for waste disposal than the lower vadose zone because finite travel times for each nuclide do exist therein. Completion of the development of a quantitative transport model for the prediction of water and radionuclide travel times and flow paths is desirable.

Radioactive wastes accidentally released to the vadose zone above twenty feet (upper vadose zone) are carefully monitored and controlled, since this is the zone where possible organism contact is most probable. Radionuclides are not normally discharged to the sediments in this zone. The expectation is disposal of radioactive wastes containing mixed fission products having an activity of $<5 \times 10^{-5}$ μC of gross β per milliliter. Even at this low standard, solution and sediment concentration of radionuclides may occur due to colloid dispersal, hydrolysis, and sediment sorption. The solution concentration of radionuclides in swamps is routinely monitored, but the sediment activity is not presently monitored except along the shoreline. It may be necessary to implement a monitoring program to determine the level of sediment radioactivity as a guide to future management practices.

Research on the soil environment of the upper vadose zone has generally been of a survey nature and limited to those zones where accidental spills have occurred. Extensive monitoring and survey programs are presently conducted. The monitoring program is designed to detect buildup or reconcentration of long-lived radionuclides such as ^{90}Sr in the surface soil or in plants.

A decade old study of the possible buildup of ^{90}Sr at Hanford and its environs due to the operation of the separation plants gave negative results, but an updating and addition to this study are suggested. In general, radioactive wastes have been more closely monitored in comparison to non-radioactive wastes.⁽¹⁵⁾

In summary, due to the suitable climate, deep permeable sediments with adequate sorption capacities, and great depth to groundwater, the Hanford project offers a nearly ideal environment for ground disposal of radioactive wastes. This somewhat unique setting offers substantial economies in the production and processing of radioactive fuels. However, it should be understood that sediment capacity for disposal is not infinite, and the best method for the disposal of all wastes is a function of both the environment and waste composition.

DISCUSSION

VADOSE ZONE BELOW 20 FEET - LOWER VADOSE ZONE

A large portion of the Hanford soil research has been related to the lower vadose zone. This research has tended to verify that the lower vadose zone is the most desirable of the three disposal zones for radioactive waste storage. Large volumes of intermediate level radioactive wastes have been routinely disposed to cribs and the radionuclides in these wastes have been retained in the lower vadose zone.⁽¹⁶⁾

Effects of Ground Disposal at Hanford on the Lower Vadose Zone

Ground disposal at Hanford has been conducted to accomplish the maximum retention of long-lived radionuclides in the lower vadose zone. The retention of a large proportion of the ⁹⁰Sr, ¹³⁷Cs, Pu, rare earths, and transition metals has been achieved in this zone. Only traces of some of these long-lived radionuclides have been detected moving into the saturated zone.⁽¹⁷⁾ A large percentage of the long-lived radionuclides is sorbed in the upper portion of the lower vadose which is high above the groundwater.⁽¹⁸⁾

Associated Research Relating to Retention in the Lower Vadose Zone

Since the lower vadose zone is the zone in which most long-lived radionuclides are stored, a large amount of research effort at Hanford has been devoted to the study of retention in this zone. Related waste disposal research can be divided into studies designed to: 1) attain maximum retention of radioactive wastes; 2) predict breakthrough of radioactive wastes into the groundwater; 3) evaluate possible removal mechanisms of previously sorbed radionuclides.

Maximum Retention Studies

In considering the disposal to the ground of radionuclides it is necessary to consider: 1) the sorption reactions of waste components with Hanford sediments; 2) composition of waste solutions disposed to the sediments; 3) the hydrodynamic flow of the waste solutions. An evaluation

and integration of data on the waste solutions and sediments relating to the above three categories allows one to predict disposal conditions which will improve the retention of radioactive wastes.

Sorption reactions - When radioactive waste solutions are percolated through sediments, radionuclides react with and are retained to a large extent by the sediments. The sorption of radionuclides can be divided into five categories of reactions: 1) ion exchange; 2) precipitation; 3) replacement; 4) other sorption reactions; 5) no sorption. The relative importance of the above reactions to the sorption of any given waste solution is a function of both the waste composition and mineralogical composition of the sediments. For example, when a waste containing both rare earth and cationic components is disposed to sediments having both cation exchange capacity and free CaCO_3 , ion exchange may be the principal sorption reaction for the cationic component; while an increase in pH due to the reaction with CaCO_3 may cause the precipitation of the rare earths due to hydrolysis. Thus, it is difficult to generalize as to what sorption reaction is the most important with reference to any given waste or component of that waste.

Ion exchange - Many solids, when placed in contact with solution, exchange ions with similar ionic species in solution. This mechanism is known as ion exchange. In ion exchange the solid (exchanger) phase remains basically unchanged during the exchange reaction, and exchange is stoichiometric. Ion exchange can be divided into cation exchange and anion exchange depending on whether cations or anions are exchanged with the solution phase. In the Hanford sediments cation exchange is the major sorption reaction, while anion exchange is minor to nonexistent.^(19,20,21) Layer silicates and zeolites of the silt and clay size fractions of sediments have relatively high cation exchange capacities due to: 1) the isomorphous substitution of lower valence for higher valence cations; 2) broken bonds at surfaces and particle edges; 3) dissociation of structural OH^- groups (22,23). Much of the exchange capacity of layer silicates and zeolites is due to isomorphous substitution with a lesser portion due to the other two mechanisms.^(24,25, 26,27,28) The exchange capacity due to isomorphous substitution is constant

at all pH values; while the exchange capacity due to dissociation of OH^- is pH dependent. The higher the pH the greater is the cation exchange capacity due to dissociation of OH^- .

Anion exchange in sediments is due to the protonation of free iron and aluminum hydrous oxides at a pH below 7.0.⁽²⁴⁾ The sediments of the Hanford project are generally basic in reaction (pH > 7.0).⁽⁷⁾ Thus, the sediments of the Hanford project would be expected to have limited anion exchange capacities as was found by Hajek.⁽²¹⁾

Precipitation - When the solubility product of any compound is exceeded in a solution, the compound precipitates. The rare-earths' and transition metals' hydroxides and hydrous oxides are relatively insoluble, and appreciable ionic concentrations of these elements can be maintained only in acid solutions. As a general example consider the reaction:

If the above hydroxide is extremely insoluble, the reaction will tend to be displaced to the right side of the equation, and any reaction that tends to make the solution more basic (remove H^+ ions) will drive the reaction further to the right. When an acid waste solution enters a calcareous soil, the free CaCO_3 in the soil tends to neutralize the waste, and precipitation of radionuclides which form insoluble hydroxides will occur.^(29,5)

Replacement - Replacement is a special case of precipitation. In a replacement reaction at least one of the components of the newly precipitated phase is supplied by a previous solid phase, and the newly precipitated phase precipitates (replaces) in place of the previous solid phase. The solubility product of the new phase would maintain the concentration of the component supplied by the previous solid phase at a lower concentration in the replacing solution than would the previous solid phase. Thus, the new solid phase is more stable in the replacing solution than the original solid phase.^(30,31,25,32)

Other sorption reactions - Sorption reactions are generally divided into chemical and physical sorption. These are usually differentiated on

the basis of whether the sorption increases (chemical sorption) or decreases (physical sorption) with increasing temperature. Some research workers have empirically measured sorption reactions that would seem to be neither an ion exchange nor a precipitation reaction, but the exact sorption mechanism was not determined.^(33,34,35,36,37) These other sorption reactions are best considered as a separate category of reactions.

No sorption - Some radionuclides fail to react with sediments to any significant degree, and percolate to the groundwater along with the waste solution. When a nonreacting, long-lived radionuclide is present in a waste at a concentration greater than 10% of MPC, the waste is disposed on a specific retention basis. Such radionuclides are present in wastes either as uncharged or anionic complexes.^(38,39,21,40)

Radionuclides in liquid wastes - Radionuclides which normally need to be considered in waste disposal include ^{90}Sr , ^{137}Cs , Pu, ^{60}Co , ^{106}Ru , transition metals, rare earths, and ^3H . In many wastes ^{90}Sr is the radionuclide which limits ground disposal to Hanford sediments due to its long radioactive and biological half-lives and resulting low MPC.^(41,39,42,11) In sediments containing small amounts of mica, ^{137}Cs could become the limiting nuclide; however, ^{137}Cs has a much shorter biological half-life than ^{90}Sr , and therefore, a higher MPC. Much of the sediment of the Hanford project has a moderate mica content and exhibits high ^{137}Cs sorption.

Plutonium is an α -emitter and as such, a biological hazard.⁽⁴³⁾ At pH above 2 it is essentially completely sorbed by the Hanford sediments, probably due to hydrolysis;^(44,45,5) however, in one laboratory study the presence of a high-salt acid waste with an appreciable organic component rendered plutonium difficult to sorb, possibly due to its being in a complexed form.^(46,40)

The transition metals also generally form insoluble hydroxides and are precipitated from solution at a moderate pH due to hydrolysis. As an example, ^{95}Zr was found to be essentially totally sorbed at a pH above 2 by the Hanford sediments.⁽⁵⁾ Several of the transition elements including Ru and ^{60}Co do, however, form soluble anionic and uncharged complexes which

are poorly sorbed by the Hanford sediments. Complexed ^{60}Co was, in the past, the limiting radionuclide in concentrations greater than 10% of the MPC. In such wastes ^{60}Co made specific retention disposal necessary.⁽³⁸⁾ Complexed ^{106}Ru has also reached the groundwater but has half-life of less than three years (1.0 year) and, thus, ample time exists for decay prior to the waste entering the Columbia River.⁽¹⁰⁾

The rare earth nuclides are generally precipitated as insoluble hydroxides at the Hanford sediment pH. As an example, virtually complete uptake of Ce was found at pH 6.0 in the Hanford sediment.⁽⁵⁾ However, complexed forms may occur and not be removed in the sediment when an appreciable organic component is present in a waste.

Tritium (^3H) has only recently been reported as a fission product.⁽⁴⁷⁾ Tritium occurs in the forms of HTO and T_2O (replacing ^1H in the water molecule). In the above study approximately 25% of the ^3H was reported in the gas phase. At Hanford 47-64% of the theoretical production was discharged to the ground via cribs and ponds and the remainder discharged as a gas.^(48,49)

Since ^3H is present as tritiated water, no sorption occurs in the vadose zone, and ^3H moves with the wastewater to the groundwater. Tritium concentration at Hanford is controlled by : 1) decay; 2) dilution; 3) hydrodynamic dispersion; 4) controlled discharge.⁽⁴⁹⁾

Groundwater contamination with ^3H is presently approximately five miles from the Columbia River at its MPC.⁽⁵⁰⁾ "It is estimated . . . that the yearly rate of fission product tritium addition to the river should not increase the present tritium background concentration by more than 50 percent".⁽¹⁰⁾

Thus, in the Hanford sediment ^{90}Sr is the radionuclide which most often limits ground disposal. In certain other sediments containing low mica contents and with high concentration ^{137}Cs wastes, ^{137}Cs could limit liquid waste disposal.⁽⁵¹⁾ In a few special wastes containing complexing agents, ^{60}Co and Pu may limit ground disposal.

Due to the likelihood of ^{90}Sr and ^{137}Cs limiting the disposal of most wastes, a more detailed review of the sorption of these nuclides will be made.

Strontium-90 sorption - Strontium is an alkaline earth metal, which usually exists in solution as a divalent cation. Strontium-90 is generally considered to be sorbed in sediments by cation exchange reactions.^(52,53,54) Certain experimental findings, however, suggest that ion exchange may not be the sole sorption reaction of Sr.

McHenry⁽⁵²⁾ found an extreme pH dependence of Sr uptake in a calcareous Hanford sediment, and Sr uptake was enhanced by the addition of $\text{PO}_4^{=}$ to waste solutions. This pH dependence was greater than would be expected from a simple ion exchange reaction. Ames and Hajek^(55,56,57) and Hajek and Ames⁽⁵⁸⁾ in statistical experiments found a large Sr-sorption dependence upon the presence of calcite in calcareous Hanford sediments. Tamura^(34,35,36,37) and Tamura and Struxness^(59,33) found selective sorption of ^{90}Sr on low exchange capacity Fe and Al oxides (hematite, limonite, corundum). Tamura also found that the Sr-sorption capacity of some soils was decreased due to the removal of free hydrous Fe and Al oxides. All of the above findings seem to indicate that Sr can be sorbed by reactions other than ion exchange.

The elucidation of the increased sorption of ^{90}Sr from wastes in which phosphate is added to the waste solutions was made by Ames.^(30,31,60) It was found that solutions containing phosphate replaced calcite (CaCO_3) in the Hanford sediments with the more stable mineral apatite at moderate and high pH. Apatite, $\text{Ca}(\text{F}, \text{Cl}, \text{OH})(\text{PO}_4)_3$, is a calcium phosphate; and Sr, which is chemically analogous to and of a similar size as Ca, was found to substitute for Ca in the apatite structure.

Increased sorption of Sr in calcareous soils in the absence of phosphate is more difficult to explain because of some, seemingly, conflicting evidence. For example, it was found that the CaCO_3 neutralization of wastes decreased the sorption of Sr relative to NaOH neutralization.⁽⁴²⁾ In addition Spitsin et al.⁽⁶¹⁾ reported that Ca was a strong competitor with Sr for ion exchange sites, which has the effect of reducing Sr sorption. Thus, increased solute Ca is not the cause of increased sorption, but to the contrary solute Ca

actually decreases Sr sorption by ion exchange. Increased sorption then must be due to the presence of solid phase calcite.

Halevy and Tzur⁽⁶²⁾ reported the precipitation of SrCO_3 , by CaCO_3 in ion exchange studies. This precipitation (replacement) of CaCO_3 by SrCO_3 may be the key to the understanding of the increased Sr sorption. The increased sorption reported by Ames and Hajek was found in an open flow system (through small soil columns) with a concentrated Na^+ influent. In this open system the concentrated Na^+ probably maintained the solution Ca^{++} concentration at a low level, thus preventing a strong Ca^{++} competition with Sr for ion exchange sites, and a SrCO_3 phase may have precipitated. Thus, precipitation would account for the increased sorption. In batch studies (closed system) with the same sediment, decreased sorption was found probably due to the overwhelming effect of Ca^{++} competition with Sr^{++} for ion exchange sites. The above reaction also explains the strong pH dependence of Sr sorption in a calcareous soil since a low pH would result in the dissolution of CaCO_3 .^(52,19)

Cesium-137 sorption - Cesium is an alkali metal element which usually exists in solution as a monovalent cation. Cesium is certainly sorbed by ion exchange; however, it is also sorbed by a more selective reaction when present in trace concentrations.^(51,63,64,65,66,67) It was demonstrated by Tamura and Jacobs⁽⁶⁵⁾ that mica-like minerals (illite) selectively "fixed" Cs^+ . The above authors further demonstrated that when expandable layer silicates were collapsed to 10 Å mica spacing by K^+ saturation and/or heating, an increased Cs selectivity by the collapsed minerals resulted.^(51,65,63,35) It was suggested that the increased Cs sorption is due to the creation of a small number of selective sites around the edges of the collapsed mica-like structure.

Cs in these selective sites is usually considered to be fixed and not to conform to normal ion exchange equilibria treatments, because the process is assumed to be irreversible.⁽⁶¹⁾ This may not be the case. The reaction may still be reversible, but has different reaction rates depending upon the direction of equilibrium attainment. Equilibrium attainment from a previously Cs-saturated system would be expected to be much slower than

would equilibrium attainment from a previously Cs-unsaturated system. This would be true because the previously Cs-saturated system would be collapsed, which would sterically hinder the exchange. Thus, the Cs-selective reaction may only be a special case of an ion exchange reaction at selective Cs sites.

Experimental methods for sorption measurement - Two general categories of laboratory experiments are used at Hanford to determine the sorption of radionuclides on sedimentary materials: batch and column studies.

Batch experiments are equilibrium experiments in which a known volume of a real or simulated waste solution is shaken with known amounts of sediments until equilibrium is attained. An equilibrium distribution coefficient (K_d) is calculated from the equation

$$K_d = \frac{As/W}{Al/V} \quad [2]$$

Where: As is the equilibrium concentration or activity sorbed on the solid phase, W is the weight of solid phase in grams, Al is the equilibrium concentration or activity of the solution phase, and V is the volume of liquid in milliliters. Thus, K_d is the ratio of the sorbed activity per gram of exchanger to the solution activity per milliliter of solution.⁽⁵⁾

In a two-ion system the K_d can be related to the equilibrium constant for an ion exchange reaction calculated from a mass action model of ion exchange. For example, consider the reaction

Where X represents the ion sorbed on the exchanged phase. The equilibrium constant (k) for this reaction is

$$k = \frac{NaX}{KX} \frac{(K^+)}{(Na^+)} = K_d \frac{(K^+)}{KX} \quad [4]$$

Where the brackets represent the equilibrium activity of ions in solution and X the sorbed activities. In a trace system where Na^+ is present as the trace element, KX is essentially a constant which is equal to the cation exchange capacity at all trace Na^+ concentrations. Thus,

$$k^1 = k KX = K_d(K^+) \quad [5]$$

Solving Equation [5] for K_d gives

$$K_d = \frac{k^1}{(K^+)} \quad [6]$$

The general equation which takes into account the valence and stoichiometry of the general mass action equation gives

$$K_d^x = \frac{k^1}{(Cy)^y} \quad [7]$$

(68,69,70) Thus the K_d is inversely proportional to the concentration of the competing ion in a trace system when mass action principles are valid.

It should be emphasized that the equilibrium K_d which is usually calculated in closed systems is a function of the concentrations of all the competing ions present in that system. Systems containing either different competing ions or different concentrations of the same competing ion will have a different K_d .⁽⁷¹⁾

Column experiments in contrast to batch experiments, are open experiments, in which various influent solutions are pumped or percolated through packed sediment columns. The effluent is sampled to determine the breakthrough of any given radionuclide as a function of the throughput volume.⁽⁵⁾ Column radionuclide distributions can be obtained by either sectioning or surveying the intact columns with a scintillation detector.⁽⁷²⁾

Small columns can be used to calculate a K_d for any ion in a given system.⁽⁷³⁾ In this technique a small volume of soil is placed in sealing tubes with fritted disc bottoms, and trace solutions are passed through the sediment until the system comes to equilibrium. Direct radioanalysis is used to determine the activity of the sorbed trace ion, while solution activity of the trace ion is equal to the influent solution activity. Calculation of the K_d is made using Equation [2].

Hajek and Ames⁽⁵⁸⁾ used the above batch and small-column techniques to compare batch and column K_d . The two K_d did not agree when the influent solution was 3N NaOAc traced with ^{90}Sr and the sediments were calcareous. The column K_d was higher than the batch K_d when the soil to solution ratios

of the batch determination were 50 or less. This is as expected, because Sr must compete with Ca in the closed system due to the solution of CaCO_3 and exchangeable Ca from the sediment. Calcium is a known strong competitor for exchange sites with Sr. In columns (open systems) the exchange became essentially Na^+ saturated at equilibrium. Na^+ is a much weaker competitor with Sr^{++} for ion exchange sites than is Ca^{++} , and an increased Sr^{++} sorption resulted. The K_d for the column was found to increase to a maximum and then slowly decrease to the value of a preleached column at equilibrium. Dissolution of the system CaCO_3 was probably attained by the preleaching. In the column which was not preleached, SrCO_3 probably replaced CaCO_3 which explains the initial increased sorption of Sr^{++} ; however, the SrCO_3 was eventually dissolved by continued leaching and the final K_d was the same as the K_d of the preleached column. This indicates a possible problem in predicting the breakthrough of long columns using short-column data when more than a single sorption reaction is occurring.

Breakthrough Prediction Studies

The ultimate objective of column and batch studies is to be able to predict accurately the breakthrough of long-lived radionuclides into the groundwater from the Hanford ground disposal facilities. Prediction would allow the cessation of disposal to any crib and time for the preparation of new disposal facilities before the groundwater reached 10% of the MPC for any long-lived radionuclide.

Presently, breakthrough is predicted empirically, but it is the object of continuing research to develop a mathematical model for breakthrough prediction. The presently used empirical procedure is to: 1) pack a column with sediment from the disposal site; 2) pump the actual waste solution **upflow** through the sediment column at the anticipated disposal flow rate; 3) collect effluent fractions with an automatic fraction collector; 4) calculate the timing C/Co value (10% of the MPC) for the limiting radionuclide; 5) plot the error function of C/Co (C/Co is the ratio of effluent to influent concentration of the limiting radionuclide) versus the log of the column volumes of effluent; 6) extrapolate the resulting breakthrough

curve to the limiting effluent concentration to obtain the total volume to be disposed. (41,74,75)

The empirical procedure has proved to be conservative because:

1) spreading beneath the crib increases the size of the effective column volume and decreases the rate of solution movement; 2) the slope of the breakthrough curve for long columns is greater than for short columns; thus, the column volumes to breakthrough for the much longer field column are greater than for short laboratory columns; 3) layering in the sediment causes further spreading. (76)

Column parameters - In order to develop a mathematical model for breakthrough prediction, various parameters were investigated at Hanford with empirical column and batch methods. (77) Parameters studied included:

1) column length and diameter; 2) unsaturated versus saturated flow; 3) temperature and accompanying ion effects; 4) organic material addition; 5) flow rate.

Column length and diameter - The effect of column length and diameter on the breakthrough curve was studied. (78) Breakthrough was found to be independent of the column diameter but dependent on the column length. Column length affected the slope of the breakthrough curve: the longer the column the steeper the slope of the breakthrough curve.

Saturated-unsaturated flow - The effect of unsaturated flow on the breakthrough curve was studied. (79) No significant difference was found between saturated and unsaturated flow although the data showed considerable scatter, making interpretation difficult.

Temperature and accompanying ions - The effects of temperature and accompanying ions were studied. (80) It was found that when Sr^{+2} was the trace ion and Na^{+} the accompanying ion, increased temperature improved the sorption of Sr^{+2} . The reverse was true when Mg^{+2} and Al^{+3} were the accompanying ions. When Cs^{+} was the trace ion, increased temperature decreased the sorption of Cs^{+} with all accompanying ions.

At a fixed temperature the accompanying ions Al^{+3} and Ca^{+2} were found to be strong competitors for exchange sites and Na^{+} a weak competitor relative to trace Sr^{+2} .

The above thermal data is important to waste disposal at Hanford because some wastes are disposed to the sediments thermally hot. Nelson and Knoll suggested the selectivity changes were the result of hydration changes caused by the increased temperatures.

Organic components - The effect of some detergents, surfactants, and polyelectrolytes on sorption was studied.^(81,82) Some of the above compounds increased sorption and some decreased sorption. This would indicate that the effects of any organic compound on waste disposal must be presently treated empirically and as an individual case.

Flow rate - The effect of flow rate on sorption was studied.⁽⁷⁶⁾ It was found that the slower the flow rate the steeper the slope of the breakthrough curve. Thus, at slow flow rates more radionuclides will be sorbed than at rapid flow rates.

Breakthrough theories - Following the preliminary parameter experiments several theoretical equations were tested to determine if a theoretical basis for breakthrough prediction could be established.

The tested sorption theories can be divided into two general categories; viz., those which consider ion exchange as the controlling cause of dispersion and those which consider ion exchange, diffusional transport, convectional transport, and hydrodynamic dispersion as the causes of dispersion.

Theories in the first category include the plate theories of Glueckauf,⁽⁸³⁾ Michaels,⁽⁸⁴⁾ Dutt,⁽⁸⁵⁾ Thornwaite et al.,⁽⁸⁶⁾ and the rate-plate theories of Vermeulen and Hiester.⁽⁸⁷⁾ The theories of Meyer and Tompkin, Michaels, Glueckauf, and Vermeulen and Hiester, all include two or three parameters; viz., equilibrium parameters (distribution rate) and kinetic parameters.⁽⁸⁸⁾ The theories of Thornwaite et al. and Dutt are plate theories with the height of the plate fixed. Frissel and Poelstra^(89,90) found the fixed plate theories failed to agree well with the column data.

The variable plate theories work well at slow flow rates and in large particle size systems where hydrodynamic dispersion is minimal.^(77,68) At high flow rates and in the presence of small sized particles (silts and clays) it was found that the system parameters of the variable plate theories changed as the column length was varied. For any theory to be useful at Hanford the system parameters must remain constant for all column lengths to allow the extrapolation of short column data to the much longer Hanford project sediment columns.⁽⁷⁷⁾

Ionue and Kaufmann⁽⁹¹⁾ formulated a general differential equation for ion exchange in which terms for the sorptive influence of the solid phase, diffusional dispersion, hydrodynamic dispersion, and convectional transport were included. A completely general solution of this differential equation has not been found; however, if the solution velocities are low enough to maintain a continuous equilibrium between the solution and solid phases, and if the equilibrium parameter (K_d) remains constant a special solution for a one-dimensional system becomes

$$C/C_o = 1/2 (1 - \operatorname{erf} [\frac{x - \bar{V}_x t/a}{2 \sqrt{D_m \bar{V}_x t/A}}]) \quad [8]$$

Where:

$$A = 1 + \rho \frac{(1 - f)}{f} K_d$$

C = effluent concentration of radionuclide

C_o = influent concentration of radionuclide

ρ = bulk density of sediment

f = fraction pore volume

K_d = distribution coefficient

D_m = dispersion coefficient

$\bar{V}_x$ = average solution velocity

x = distance from the influent end of column

t = time from initial disposal

For Equation [8] to be valid: 1) the porosity of the sediment must remain constant; 2) the K_d must remain constant in time and space; 3) the column's solution and solid phases must be at instantaneous equilibrium; 4) the flow rate must remain constant. Any deviation from the above conditions will result in an error between measurement and theory. The first condition, that the porosity must remain constant throughout the column requires the column packing to be uniform. Errors of 0.01 and 0.20 in the porosity cause errors of 3% and 31% in C/C_0 , respectively. Thus, the column packing is critical. For sand columns this error can be held relatively small, but for heavier sediments this error severely limits the usefulness of the breakthrough data. Thus, much of the quantitative work has been done with sand columns. Packing problems do not severely limit work on the Hanford sediments since they are predominantly sands.

The second condition, that the equilibrium constant (K_d) must be known and remain constant in both time and space, requires the exchange capacity be homogeneous with respect to the column length, and the accompanying ions in solution be in equilibrium with the solid phase. This condition is not met when a waste solution which contains essentially Na^+ is disposed to a sediment which is Ca^{++} saturated (based).

The condition that equilibrium must prevail at all points in the column between the solution and solid phases requires that the flow rate be less than some maximum rate. This maximum rate is probably a function of both the exchange capacity and the texture of the sediment. In addition, this maximum flow rate may not be a constant at all points on any given breakthrough curve; as is evidenced by the tendency of many measured breakthrough curves to be asymmetrical. (80,88,92)

The final condition that a steady flow rate must be maintained, is relatively well met by the use of positive displacement pumps in sediment column experiments.

When all of the conditions are met, good agreement is obtained between measured and predicted [Equation [8]^(92,72,93,91)] breakthrough. A computer program is available to simplify the mathematical solution of Equation [8].⁽⁹⁴⁾

Field experiments - Some actual field experiments have been attempted at Hanford to determine if the empirical methods used to estimate breakthrough are realistic. Only a single group of experiments were conducted to measure breakthrough data in the field.^(95,96,77) Two sizes of model cribs were used in the experiments to measure breakthrough of ^{85}Sr from a 600 ppm $\text{Ca}(\text{NO}_3)_2$ solution. The cribs were small compared to actual waste disposal cribs.

Lateral spread from the field column (actual volume of sediments directly below the crib and above the groundwater) was found to be constant and negligible when extrapolated to the size of waste disposal cribs. It was also found by calculating an effective column volume (wetter volume) that laboratory prediction and field column data were in good agreement.

Coring beneath abandoned cribs has been conducted to compare short column radionuclide distribution with field column distributions, and the agreement has been satisfactory. The corings have shown that the bulk of the sorbed ^{137}Cs and ^{90}Sr is contained in the first few meters of the crib columns which are high above the saturated zone.

Raymond⁽¹⁸⁾ found that the maximum sorptions of ^{137}Cs and ^{90}Sr were 3 and 7 feet below the crib, and essentially all of these nuclides were sorbed at least 100 feet above the groundwater. Brown⁽⁹⁷⁾ found that 99.9% of the long-lived radioisotopes were sorbed in the first 10 meters of a 60-meter sediment column. These data are in good qualitative agreement with laboratory column data.

Sorbed Radionuclide Removal Studies

Once radionuclides are sorbed in the sediment profile, they are in the zone where organism contact is minimal. If the long-lived radionuclides are not removed from this zone prior to many half-lives, radioactive decay will reduce the amount of these radionuclides to innocuous levels. However, the possibility of removal exists which could limit the amounts of long-lived isotopes that can be disposed.

Possible mechanisms of desorption (removal) of radionuclides include: leaching, diffusion, and particulate transport.

Leaching - Leaching is the dissolution of soluble components by percolation of solution through a sediment. Natural leaching at Hanford would be due to the percolation of precipitation through the vadose zone to the regional groundwater. The maximum total precipitation that could leach the Hanford sediments for long periods of time would be the average precipitation per year times the number of years in the period. The actual leaching volume would be the above figure minus that water lost through evapotranspiration and specific retention. This figure is low and may in fact approach zero.

Brown⁽⁹⁷⁾ reported that it required 50 column volumes to leach 11% of the ^{137}Cs and 4% of the ^{90}Sr from a Hanford sediment. Miller and Reitemeier⁽⁹⁸⁾ found it took 260 and 250 inches of water and 1N CaCl_2 solution, respectively, to move ^{89}Sr 2 inches and 5 inches, respectively, in a slightly calcareous soil. Hajek⁽⁴⁶⁾ found that only 0.1% and 7% of the Pu and Am, respectively, are leached by 15 column volumes of groundwater.

Thus, it would take many column volumes of water to leach even a small amount of any of the above radioisotopes to the groundwater. A column volume would be 100-200 feet of water which is equal to the total precipitation on the Hanford project for from 200-400 years. Thus, even the maximum possible natural leaching would not be an important removal mechanism for sorbed radionuclides.

Diffusion - Diffusion is the movement within and between phases to obtain intermingling of molecular and/or ionic particles due to a concentration gradient. The maximization of entropy is the driving force of the reaction. Diffusion as compared to mass transport is generally a slow method of transport.

Hajek⁽⁴⁶⁾ found that the diffusion velocity of Pu and Am was 10^{-4} times the transporting solution velocity, and thus, would be a negligible removal mechanism from a sediment profile in 10 half-lives (essentially complete activity depletion). Hajek⁽¹¹⁾ determined the diffusion coefficient for ^{90}Sr and ^{137}Cs in Hanford sediments and found that ^{90}Sr would diffuse about 0.5 feet in the first 10^3 years (~ 30 half-lives). Diffusion of ^{137}Cs was even slower. Thus, diffusional removal from the vadose zone can be discounted as a significant mechanism.

Particulate transport - Particulate transport in normal operating conditions is due to the Tyndall effect on colloidal-sized, dispersed (peptized) particulates. Peptization takes place in dilute salt solutions and/or at high pH in water saturated sediments. The dispersed particles would be transported by mass flow in the percolating solution.

Rhodes et al.⁽³⁹⁾ found that when large volumes of low-salt wastes were disposed to Hanford swamps the sedimentary columns plugged due to the filtering out of dispersed particles. Carlile and Hajek⁽⁹⁹⁾ showed that the partial breakthrough of ^{90}Sr was almost immediate when river water (low salt content) was percolated through N-area sediments. This breakthrough, due to the movement of dispersed particulates, could be prevented by pre-leaching the columns with river water to remove easily dispersed particulates.

Several research workers have reported decreased uptake of ^{90}Sr , Pu, ^{95}Zr , ^{144}Ce , ^{90}Y , and ^{106}Ru at high pH; probably due to dispersion and particulate movement. (5,100,39,77,61)

In all of the above cases losses would be relatively low if the pH of the system were not above 9.0, and if salt were added to the dilute systems. In certain special cases preleaching of the sediment column may be practicable. In all cases the remedial measures are known to keep sorbed radionuclide movement to insignificant levels. Thus, there seems to be little likelihood of radionuclide movement from the vadose zone below 20 feet prior to complete decay depletion, except possibly by some catastrophic event in which the vadose zone itself were physically moved.

SATURATED ZONE

The saturated zone on the Hanford project is generally 100-300 feet below the land surface in the disposal areas.⁽¹⁰¹⁾ The saturated zone is the second most desirable zone to which radioactive wastewater can be disposed. This zone is a less desirable disposal zone than the vadose zone below 20 feet since wastes entering the zone will over a period move to the Columbia River where organism contact is presumable. However, the saturated zone is a more desirable zone of disposal than the vadose zone above 20 feet,

since decay occurs with no organism contact during the travel time of radiocontaminants to the Columbia River. Thus, the concentrations of short half-lived radionuclides will be greatly reduced prior to possible organism contact at the Columbia River.

The Effects of Ground Disposal at Hanford on the Saturated Zone

Ground disposal of wastes has been responsible for: 1) an increase in the Hanford water table elevation with attendant changes in the flow system; 2) introduction of radioactive contamination into the groundwater; 3) introduction of nonradioactive contaminants into the groundwater.

Groundwater Elevations

The disposal of large volumes of slightly contaminated coolant water to the Hanford sediments has produced two large groundwater mounds near the processing areas. The mounds slowly developed as coolant water was initially disposed to the ground, and have shown some fluctuations when the coolant water disposal was altered. The Eastern and Western mounds rose to 25 and 90 feet respectively above the pre-Hanford project groundwater level by 1957.⁽⁸⁾ Even 20 miles to the southeast of the mounds, the water table increase was noted to be 0.5 feet.⁽¹⁰²⁾ Since 1957 the mound height has generally stabilized with only minor changes being noted.⁽¹⁰³⁾ The mounds have locally reversed the flow of the groundwater, and have increased the hydraulic gradient and flow rate of water to the Columbia River.⁽¹⁰²⁾

Radioactive Contamination

Radioactive contamination of the groundwater to date has been maintained in small localized areas with the exception of Ru and ³H contamination. Ruthenium-106, which has a half-life of one year will decay substantially prior to entry into the Columbia River. Tritium contamination is present in much of the groundwater. Currently ³H is present in the groundwater at 10-50% of the MPC approximately 5 miles from the Columbia River at high water stage.^(104,17) Low concentrations of ⁹⁰Sr, ⁶⁰Co, and ¹³⁷Cs have been detected near disposal cribs, but these radionuclides have not been more than a few hundred feet from any crib.

Nonradioactive Contamination

Of the nonradioactive contamination in the groundwater, nitrate (NO_3^-) appears to be the most predominant. The MPC for nitrate is 45 ppm. Concentrations of nitrate as high as 1700 ppm have been detected in the groundwater from wells near disposal sites. Concentrations of nitrate as high as 57 ppm have been reported from wells approximately 5 miles from the Columbia River (Figure 1); however, these concentrations appear to have declined somewhat in 1967. A thorough sampling and well evaluation program should be utilized to monitor groundwater nitrate contamination.

Associated Research Related to the Saturated Zone

Research related to the saturated zone can be divided into three categories of investigations; these can be broadly termed: 1) geological definition, 2) monitoring, and 3) flow model development.

Geological Definition

To develop an effective waste disposal plan at Hanford it was recognized early that: 1) the stratigraphy of the project must be defined in considerable detail, and 2) the aquifer characteristics of the sediments of the saturated zone must be evaluated by field measurements. (105,106,107)

Stratigraphy - The stratigraphic relationships of the formations of the Hanford project have been well defined by Hanford geologists. The general stratigraphy of the Hanford sediments was reviewed in the introductory section so further discussion will not be given. A more detailed review of the Hanford stratigraphy can be obtained from references 6, 108, 109, 110, 111, 112, 9, and 113.

The stratigraphy has been determined; 1) directly, from Hanford well logs, 2) indirectly, from geophysical methods which include airborne magnetometer surveys, ground magnetometer surveys, and seismic tests. (114) The topography of the buried basalt bedrock is easily measured by magnetometric methods which define the magnetic permeability of the earth, while the magnetite content of basalt allows the definition of its surface. Several buried basalt ridges that have previously escaped detection were

defined by aerial magnetometric survey.⁽¹¹⁴⁾ Seismic methods require the introduction of energy into the earth (usually by detonations) and measurement of the response. At Hanford seismic methods are probably most useful in detecting bedrock and tracing interbasalt beds.⁽¹¹⁴⁾

Field measurements for aquifer characteristic definition - Extensive field measurements of sediment and aquifer characteristics have been made at Hanford. These include: 1) pumping tests, 2) tracer tests, and 3) water level tests conducted to obtain needed data on: 1) permeability of the Hanford sediments, 2) groundwater flow rates, and 3) well performance.

Pumping tests - Multiple and single-well pumping tests were conducted to obtain drawdown data to be used as input to nonequilibrium and modified equilibrium formulas to calculate sediment permeability. Permeabilities calculated by the above methods ranged from 1.3×10^4 - 6.7×10^4 , 9.0×10^2 - 5.0×10^3 , 5.0×10^1 - 5.8×10^2 , 1.5×10^2 - 4.5×10^2 , and 1.0×10^0 gallons per foot²/day for the glaciofluvial, mixed glaciofluvial Ringold, Ringold sand and gravel, Ringold conglomerate, and Ringold clay sediments, respectively.⁽¹¹⁵⁾ Thus, the glaciofluvial sediments are about 100 times more permeable than the associated Ringold sediments.

Pumping tests were also used to obtain specific capacity data, which was in turn used as a check of the Hanford sediment permeability. The specific capacity is the yield of water per foot of drawdown in a well. The permeabilities estimated with this method were in good agreement with drawdown permeabilities.^(116,115,3)

In step-drawdown pumping tests used to measure well efficiency, it was found that wells with low efficiencies were drilled 8-10 years prior to the tests, and might be expected to be less efficient.⁽¹¹⁵⁾ The step-drawdown tests give a well criterion that allows the selection of wells which give the most valid data for aquifer evaluation and groundwater modeling.

Tracer tests - Tracer tests were conducted to determine the rate and direction of groundwater movement. From the groundwater movement rate the

average sediment permeability can be calculated with Darcy's law by assuming sediment homogeneity. The permeabilities calculated from tracer tests ranged from 9.5×10^4 - 16.5×10^4 gallons per foot²/day for the glaciofluvial sediments. The higher value was calculated from the initial tracer arrival times and was higher by a factor of two than permeabilities measured from pumping tests. The lower value, which was calculated from model tracer arrival, was in approximate agreement with pumping permeabilities.^(8,115)

A second tracer technique utilized the increased conductivity of solutions containing electrolytes. The technique (direction-velocity) is used as an in-well measurement of the direction and velocity of water movement. In the technique an electrolyte is added to the well and the conductivity is measured continuously between two electrodes. As water flows through the well the concentration of electrolyte decreases, resulting in a decreased conductivity. The rate that the concentration decreases is proportional to the flow rate, and an empirical relationship between flow rate and rate of change of concentration is known, allowing the flow rate to be calculated.

A modification of the direction-velocity technique is used to determine water flow direction in a well. In this modification an electrolyte is released at the well's center and is detected at the well perimeter by moveable conductivity electrodes. The electrodes which first detect the electrolyte are in the direction that the electrolyte (and water) is moving.⁽¹⁰²⁾

Water level tests - Water levels of wells were monitored by recording hydrographs. The causes for fluctuations noted include: 1) annual rise and fall due to Columbia River stage, 2) diurnal changes in response to atmospheric pressure, and 3) random changes in response to far-distant earthquakes.⁽¹⁰²⁾

The annual changes, especially those due to groundwater recharge from the Columbia River during spring high water stage, are the most useful hydrologically. Rowe⁽¹¹⁷⁾ developed an equation for estimating the permeability of sediments into which water moved as a result of increased river stage. Permeabilities determined with Rowe's method were found to be in good agreement with pumping tests.^(118,104) It was found that wells

affected by recharge due to high river stage showed a temperature decrease, allowing unaffected wells to be easily distinguished from affected wells. Wells affected were found as far inland as 2.5 miles, and a map of the affected area on the Hanford project was published.⁽¹⁰⁴⁾

Monitoring

An extensive program of groundwater monitoring has been conducted since the start-up of the Hanford project. More than 500 wells have been drilled primarily for the surveillance of radionuclide and nonradioactive contamination.⁽¹¹⁹⁾

Secondary purposes for the wells at Hanford are to obtain well logs for the determination of the Hanford stratigraphy and to collect sediment samples for laboratory studies. Two different well drilling techniques have been used to drill the Hanford wells. These are termed "bailer" and "drive barrel" methods. The effect of "bailer" versus "drive barrel" drilling on sample modification was evaluated and no measurable differences were found.⁽¹²⁰⁾

The groundwater monitoring program has three functions: 1) evaluate the effect of the disposal of radioactive and nonradioactive wastes on groundwater quality, 2) determine when 10% of the MPC of the limiting radionuclide is attained so that ground disposal can be shifted to a new disposal facility, and 3) provide input data for the flow model program.

Components measured in the groundwater include: gross β , ^3H , gross α , ^{90}Sr , ^{137}Cs , ^{60}Co , groundwater temperature and groundwater quality parameters. Groundwater quality parameters which are measured include: pH, NO_3^- , $\text{SO}_4^{=}$, Na^+ , Ca^{++} and organic fraction. The general status of the groundwater is evaluated by integration of present gross β and ^3H contamination data, temperature measurements, radionuclide data, water quality data, and previous data on all of the above. A data summary and interpretation report is published semiannually as an "AEC Research and Development Report." The latest such report published (BNWL-1047) references earlier reports.⁽⁵⁰⁾

Flow Model Development

An accurate prediction of radionuclides travel times in the groundwater to the Columbia River is necessary to the functioning of a safe disposal program. The present approach of ceasing ground disposal when 10% of the MPC is detected in groundwater is a conservative approach, regardless of what the travel time to the river may be. Even if the unsaturated-sediment effluent were pumped directly into the Columbia River, no significant health hazard would occur and, in addition, further dilution will occur in the river water. Any waste will enter the river at less than the unsaturated-sediment effluent concentration due to: 1) decay during travel to the river, 2) sorption reactions, 3) hydrodynamic dispersion, 4) diffusional dispersion, and 5) groundwater dilution. At one end of the spectrum, the above mechanisms will greatly lengthen the travel time for sorbed radionuclides such as ^{90}Sr , but only slightly lengthen the travel time for nonsorbed ^3H at the opposite end of the spectrum.

The development of flow and sorption models have been the object of intensive study at Hanford. Significant progress has been made toward development of such models. (121,122,123) The present approach is to: 1) measure groundwater potentials of the area in the field for use as input data in the "GENORO" computer program to obtain an equation for the potential; 2) substitute the potential equation into the "STREAM" computer program to calculate the permeability distribution of the area, 3) substitute the permeabilities from the "STREAM" program into the "STEADY" computer program with appropriate boundary conditions for complete solution of the boundary value problem. (124,125)

The "GENORO" program requires the input of functional forms which include power, trigonometric, and logarithmic series. The program is a data fitting procedure in which 450 to 580 observations are usually used to obtain 45 to 36 fitting terms.

The "STREAM" program carries on three operations in addition to the permeability function. These are generation of streamlines, water travel times, and flux distributions.

The boundary value problem which is solved by "STEADY" interrelates all pertinent variables to solve for new potentials for any flow system changes. The boundary value problem combines the equation of state, Darcy's law, and the equation of continuity to derive the basic partial differential equation describing fluid flow.^(124,126) Nonlinearity of the basic partial differential equation somewhat complicates obtaining the solution. Partially saturated flow in porous-heterogeneous media (saturated flow and homogeneous media are special cases of the general equation) is completely described by the basic equation.

Both digital and analog simulation methods of solution of the general equation have been attempted.⁽¹²⁷⁾ Analog methods of solution have proved successful, but are probably limited to the special case of saturated flow and are estimated to be more costly than digital methods. The digital computer solution is based on finite difference methods. To obtain the solution, the partial differential equation is reduced to a system of simultaneous equations which are solved iteratively. The "STEADY" computer program can solve one-, two-, and three-dimensional and axisymmetrical problems with up to 800 grid points. Unfortunately, large, three-dimensional flow systems may require the solution of 18,000-25,000 simultaneous equations, which limits the present computer program to two-dimensional flow problems. Two-dimensional approximations of three-dimensional flow have been successfully solved for the proposed Ben Franklin Dam and 1301-N crib flow problems.^(128,123)

VADOSE ZONE ABOVE 20 FEET - UPPER VADOSE ZONE

The upper vadose zone is the zone in which possible organism contact with radioactive wastes is most probable; thus, this zone is the least desirable zone for radioactive waste disposal. At Hanford only low-level wastes ($<5 \times 10^{-5}$ $\mu\text{C}/\text{ml}$ of gross β) are deliberately and routinely disposed to this zone. Other sources of radioactivity to the vadose zone above 20 feet include: 1) fallout from offsite nuclear tests,⁽¹²⁹⁾ 2) off gas and particulate escape from the chemical processing facilities,⁽¹³⁰⁾ and 3) natural radioactivity.⁽¹³¹⁾ Of the three additional sources of radioactivity only the second of the above sources is due to the presence of the Hanford nuclear facility.

Once radionuclides are present in an environment there are three methods by which they can react with the human population inhabiting the environment: 1) direct exposure, 2) inhalation of radionuclides with air during normal breathing, and 3) ingestion of radionuclides with food and water. Of these three methods the third is of primary interest due to the possible concentration of radionuclides by food chains. Concentration might include: 1) deposition on plants or soil surface of an initial concentration of radionuclides, 2) ingestion of contaminated plants by herbivorous animals, 3) ingestion of herbivorous animals by carnivorous animals and scavengers, and 4) ingestion of animal and animal products by man. At each step of this food chain there is the possibility of the concentration of radionuclides by a metabolic process. As an example of this concentration, fish and ducks have been shown to concentrate ^{32}P manyfold in the Columbia River due to the successive concentration of P by each member of the natural food chain. (132,133,134)

Effects of the Hanford Nuclear Facility on the Upper Vadose Zone

Sufficient data for a comprehensive appraisal of the effect of the Hanford project on the upper vadose zone is not readily available, but some trends can be seen from routine monitoring data and Hanford environmental studies. Possible contamination of the environment by the Hanford works is discussed under the categories of: 1) swamp disposal, 2) fallout from local escapes of radionuclides.

Swamp Disposal

Swamp disposal has usually been limited to solutions that have a total β concentration of less than $5 \times 10^{-5} \mu\text{C}$ per milliliter. This standard was adopted based on experience with early Hanford swamp disposal. It was found that when the inlet solution activity of a swamp was increased to $10^{-4} \mu\text{C/ml}$ of gross β that the shoreline contamination rose to 10^4 counts per minute and the contamination was spread up to 200 feet from the swamp by wind. In a second swamp, when the activity of the input solution was $10^{-3} \mu\text{C}$ per milliliter of gross β , it was found that the shoreline contamination attained values as high as 3.5 rads per hour. (135)

Even at the standard low input level of 5×10^{-5} $\mu\text{Ci}/\text{ml}$ of gross β it may be that the solution activity of swamps can attain relatively high values due to solution concentration by evaporation. Some of the large volumes of water disposed to swamps are essentially salt free. Such waters can be expected to disperse soil colloids, which in turn can be expected to plug the sediment pores and restrict percolation.⁽¹³⁶⁾ With restricted percolation a greater portion of the water can be expected to be evaporated in the hot, low-humidity desert climate of Hanford, concentrating the radionuclides in the resulting solution. Rare earth and transition metals can be expected to precipitate in any solution which is not strongly acid due to hydrolysis, which will further tend to plug the sediment and limit percolation. Hydrolysis may also result in a buildup of radionuclides in the swamp sediment. Apparently, the above process could be occurring in some Hanford swamps, since the solution concentration of some Hanford swamps has approached and exceeded the limit of 5×10^{-5} μCi per liter for open waters for a portion of the year.⁽¹³⁷⁾

If need for the abandonment of a swamp were to arise, it would probably be necessary to cover the exposed swamp bed with soil to prevent the dissemination of the sediment by wind and indigenous animals.⁽¹³⁵⁾ It would seem that an assessment of the possible buildup of activity in the sediment should be conducted to evaluate the necessity of establishing measures that would be taken to remove a swamp from service.

Aerial Fallout of Particulate Escapes

The practice of surveying control plots for particulates above background with a G.M. counter does not assess the possible buildup of long-lived radionuclides such as ^{90}Sr . Particulates containing ^{90}Sr and ^{137}Cs were identified in 1966 and 1967 on Hanford control plots.^(138,137) The vast majority of these particles were found on 2 of the 46 monitored control plots which are adjacent to the 200 areas. Thus, it would seem logical to suspect that these particles were present in stack effluent from the chemical processing plants. An additional survey could be conducted to determine if there were any buildup of an index radionuclide such as ^{90}Sr .

In a decade-old special survey, an attempt was made to demonstrate the buildup of ^{90}Sr on the Hanford project and its environs due to possible release from the Hanford separations plant.⁽¹³⁹⁾ The sampling was conducted in 1956 and 1957, and no buildup of ^{90}Sr above that which would be accounted for by fallout was found. The survey measured the amount of ^{90}Sr that could be extracted by a neutral ammonium acetate solution.

Since a decade has elapsed since the previous ^{90}Sr study was conducted, it might prove useful to conduct a similar up-to-date study with the addition of a pH 5.0 ammonium acetate extractable fraction, which would extract SrCO_3 , and of a total ^{90}Sr fraction. The decline in the number of aerial nuclear tests which are being conducted would further aid in the assessment of any possible buildup.

There is, seemingly, relatively little hazard to humans from the Hanford plant release of ^{131}I . Even an accidental release of 60 curies of ^{131}I in 1963 resulted in the maximum exposure to a single 4-year old drinking a gallon of milk per day from a cow pastured in a local "hot spot" of 0.035 rems.⁽¹⁴⁰⁾ The FRC Radiation Protection Guide for ^{131}I exposure is 1.5 rems per year for a comparison. The quantity of ^{131}I normally released by the Hanford operations is much less than the above accidental release and, therefore, less hazardous.

Associated Research Related to the Upper Vadose Zone

Research related to the upper vadose zone can be divided into: 1) biological cycling studies and 2) environmental assessment studies. Data from biological cycling studies are used to set general standards of what is a safe and acceptable level of radioactive contamination. Data from environmental assessment studies are used in the evaluation of the hazard posed by the presence of radioactive contamination in a particular environment. Thus, both types of studies are necessary for a sound environmental evaluation of radioactive contamination.

Biological Cycling

The soil is one element in the food chain cycling of radionuclides which includes: 1) soil, 2) plants, 3) animals, 4) man. Radionuclides which enter the environment from nuclear testing and as wastes from nuclear facilities are deposited either on the soil or plant surfaces from whence they may enter the biological cycle and eventually react with animals including man.

The soil is a medium through which radionuclides move to be taken up by plants. Inputs of radionuclides to the soil can be from: 1) fallout from nuclear testing, 2) surface disposal of wastes, 3) fallout from gaseous and particulate escapes from nuclear facilities, and 4) organic residues from decaying plant and animal material which were previously contaminated.

Fallout is usually deposited on the soil as a particulate, although some gaseous forms do occur.^(129,141) Mechanisms by which fallout reaches the earth's surface include: 1) rainfall scavenging, 2) gravitational settling, 3) impaction, 4) eddy turbulence, 5) diffusion. Of the above mechanisms, rainfall scavenging was found to account for 50-90% of the ^{90}Sr fallout at five sites in 10-15 inch per year rainfall regions. Even in a two to three inch per year area (Arizona), rainfall scavenging was found to be responsible for deposition of about half of the radioactive fallout.⁽¹⁴¹⁾ Once deposited, some fallout may be redistributed by wind due to: 1) surface creep, 2) impingement of previously airborne particles (saltation), 3) resuspension in the air,⁽¹⁴²⁾ which may reinitiate the cycle.

Radioactive fallout particulates which are deposited on the soil surface will remain in an inert form until dissolved into the soil solution. The solubility (rate of solution) of fallout particulate may be quite variable. In a laboratory study, Lane⁽¹⁴³⁾ found that ^{137}Cs sorbed on feldspar, clay, and quartz was nearly insoluble in water and only slowly soluble in 0.1 N HCl after the above minerals were heated to temperatures sufficiently high to convert them into glass. Larson *et al.*⁽¹⁴⁴⁾ and Larson⁽¹⁴⁵⁾ found that less than 2% and 14-36% of the less than 44 μ particulates from a tower detonation were soluble in water and 0.1 N HCl, respectively. In contrast, 31% and 90% of the greater than 44 μ particulates were soluble in water and 0.1 N HCl,

respectively, in a balloon detonation; a lesser amount of the less than 44 μ material was found to be soluble. In the tower test the fireball intercepted the ground with a high intensity and a large portion of the resulting fallout may have been converted into glass. In the balloon test the fireball probably did not intercept the ground with the same intensity and a lesser amount of the fallout was converted into glass. Apparently, enough heat did intercept the ground to convert some of the smaller particulates into glass, reducing the solubility (rate of solution) of the finer fractions. Thus, the solubility of fallout apparently is a function of the fireball temperature and duration at the site of fallout pickup following the detonation of a nuclear device. The temperature at the site of fallout pickup may be a function of the bomb intensity and distance from the blast to the site of fallout pickup. Therefore, much of the fallout from nuclear tests may be sparingly and/or slowly soluble.⁽¹⁴¹⁾

The most abundant nuclides remaining in the environment from weapons tests are ^{137}Cs and ^{90}Sr .^(146,147) These two nuclides account for most of the biologically important nuclides. The general soil chemistry of ^{90}Sr and ^{137}Cs has been discussed in detail in the "vadose zone below 20 feet" section of this paper, and only the soil chemistry which affects the biotic availability of these nuclides will be discussed in this section.

Rediske and Selders⁽¹⁴⁸⁾ found that plant uptake of Sr from a nutrient solution was significant and proportional to the solution concentration of Sr up to 100 ppm Sr. Several investigators have demonstrated that plant uptake of Sr is dependent upon the solution pH and Ca concentration in base unsaturated soils.^(149,150,151,152,153,154) The above investigators found that the higher the pH and greater the percentage of Ca saturation the lower the uptake of Sr; however, in Ca-saturated soils additional Ca-amendments failed to further decrease the uptake of Sr.

A similar effect of K was found on Cs uptake by plants.^(155,156,157) An addition of K to a soil of low K fertility was found to decrease the uptake of ^{137}Cs ; while soils of high K fertility were found to be little affected by the amendment of K. The addition of stable ^{133}Cs was found to greatly increase the uptake of ^{137}Cs . The maximum uptake of ^{137}Cs by a

single cropping of Ladino Clover was found to be only 0.13% of the dose compared to a 4.4% uptake of ^{90}Sr . Nine successive croppings were found to remove a total of 24% of the ^{90}Sr , but only 0.72% of the ^{137}Cs doses.⁽¹⁵⁸⁾ Klechkovsky⁽¹⁵¹⁾ found the same uptake of ^{90}Sr and ^{137}Cs trends for peas and wheat. Thus, from an uptake point of view ^{90}Sr may be, potentially, more critical than ^{137}Cs .

One nuclide in addition to ^{90}Sr and ^{137}Cs may be biologically important due to its possible health hazard to man: radioiodine (^{131}I) is released by both nuclear detonations and by the processing of nuclear fuels.^(159,144) Iodine-131 is released to the atmosphere both as a gas and attached to particulate matter.⁽¹⁶⁰⁾ Although ^{131}I has a short half-life (8 days), it is potentially of concern due to its concentration in the thyroid gland. Iodine-131 is particularly hazardous to children due to its presence in milk and its concentration in milk by cattle eating contaminated grass.⁽¹⁶¹⁾

It has been demonstrated that ^{131}I can be sorbed directly on the leaves of plants providing an intake path to cattle feeding on contaminated forage.^(146,162,163) The presence of ^{131}I in the Hanford environs is closely monitored and only 3% of the maximum permissible dose is presently being ingested due to the drinking of local milk.⁽¹⁶⁴⁾

Radionuclides can be sorbed directly on/in the leaves of plants without being sorbed via the soil. This was demonstrated by Sommermeyer and Godt⁽¹⁶²⁾ who showed that the relative heights of the γ -spectrum of ^{95}Zr , ^{95}Nb , ^{141}Ce , ^{140}Ba , ^{149}La , and ^{103}Ru sorbed on grass were the same as fallout deposited on vaseline-coated paper; however, the above authors also found that ^{137}Cs was sorbed from a soil primarily through the roots of potatoes, showing ionic forms of radionuclides may be primarily taken up from the soil. Apparently, no attempt was made to determine if the above fallout was surface deposition and capable of being washed off. In a later study Hungate et al.⁽¹⁶³⁾ found considerable ^{131}I was potentially removable with water washing. Thus, it may be that radionuclides deposited from fallout are largely particulates which are capable of being washed from the surface and are largely incapable of being translocated.

One should be cautious in interpreting plant uptake and soil data. Much of the work has been done with either nutrient solutions or pot tests. Increased uptake from nutrient solutions is to be expected relative to uptake from soils, and increased responses are often observed in pot tests relative to field tests. Cline⁽¹⁶⁵⁾ found that ^{137}Cs is only slightly available to plants grown in pots compared to nutrient solutions; however, saturated soils showed uptakes similar to nutrient solutions. Klechkovsky⁽⁵³⁾ reported that considerably less ^{90}Sr and ^{137}Cs were taken up from soils than from nutrient solutions. Even Pu has been shown to be taken up by plants from clay suspensions in amounts of 20-40% of the applied dose;⁽¹⁶⁶⁾ whereas Pu uptake from soil is negligible. Thus, applying nutrient solution data to estimate radionuclide uptake would be questionable. On the other hand Auerback and Crossley⁽¹⁶⁷⁾ demonstrated the uptake of ^{90}Sr and ^{137}Cs by corn from contaminated soil at Oak Ridge, although not to the extent that would be expected if one were to extrapolate solution and pot test data to field tests. Thus, it would seem that plant uptake data from solution and pot tests are in qualitative agreement with field tests, but not in good quantitative agreement. Since much fallout is only slowly soluble, tests made with completely soluble nuclides should be considered as the worst possible case when related to fallout. In addition, the movement of slowly soluble radionuclides from the surface where root contact is minimal to the root zone may further limit the rate of uptake by plants.

Hanford Environmental Assessment

An extensive surveillance-monitoring program is maintained on the Hanford project to assess the effects of the Hanford nuclear facility on the Hanford environment. Surface monitoring at Hanford can be divided into: 1) liquid monitoring, 2) atmospheric monitoring, 3) radiation surveys of surface contamination and external radiation exposure rates.

Liquid Monitoring

Liquid monitoring includes the weekly monitoring of the ambient levels of ^3H , ^{90}Sr , total β , and total α in Columbia River water at Priest Rapids Dam prior to the use of the water as reactor coolant. After the use of

raw Columbia River water as a coolant it is further monitored in the retention basins and at three downstream sites to determine the concentrations of radionuclides released by Hanford reactors. Drinking water is monitored on the Hanford project at four locations for gross β activity, which is used to calculate the probable, maximum gastrointestinal dose. This dose is based on an assumed intake of 1.2 liters of water per day, five days a week.

In addition to the above liquid monitoring, open waste waters including swamps, ditches, and ponds (which can be used by migratory waterfowl) are monitored for gross α and β . Special analyses including γ -scans are conducted on open waters when indicated by the routine monitoring programs. Inlet and outlet ditches in addition to swamps and ponds are also monitored.

Atmospheric Monitoring

Routine (weekly) monitoring of total β , total α and ^{131}I of the atmosphere at 23 Hanford sites is maintained. The total β and total α represents the activity of particulates entrapped by 70H filter paper, while ^{131}I is collected in a NaOH solution by the passage of air through it.

Surface Surveys

Surface surveys are routinely made at Hanford of: 1) roads, 2) control plots, 3) aerial flight paths. The Hanford roads are monitored with a bio-plastic scintillation crystal which is attached to the front of a truck. A total of 46 ten-by-ten foot land areas (control plots) are routinely monitored to detect the deposition of radioactive particulates. The particulates are located by monitoring the activity above background with a G.M. counter. Aerial surveys are routinely conducted of the Hanford environs. Eleven flight patterns are flown. The aerial surveys are conducted to insure that a capability be maintained for rapid assessment of an emergency situation.

In addition to the above routine surface surveys, exposure rates are measured on the Hanford plant and along the Columbia River shoreline. On-plant exposure rates are continuously measured with pencil-type dosimeters, while Columbia River shoreline exposure rates are measured weekly at three feet above the ground with a 40-liter ionization chamber.

The above data are summarized and published annually by the Environmental Health Department, Battelle-Northwest, for the information of the Richland Operations Office of the Atomic Energy Commission and its contractors. The documents for 1965, 1966, and 1967 are BNWL-CC-913, BNWL-CC-637-13, BNWL-CC-1197-6, respectively.

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APPENDIX A

CHARACTERISTICS OF THREE HANFORD PROJECT SOILS

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CHARACTERISTICS OF THREE HANFORD PROJECT SOILS

Selected chemical, mineralogical, and morphological properties were determined for three soil types. These soils, Ritzville silt loam, Burbank loamy sand and Ephrata sandy loam, are representative of the three general kinds of soils which occur on at least 90 percent of the project. Burbank is similar to most of the deep sands north of the Hanford highway, Ritzville is similar to the loamy soils south of the highway to the crest of Rattlesnake Hills and Ephrata is representative of small areas of sandy loams from Cold Creek Valley to the Columbia River.

SOIL DESCRIPTIONSRitzville Silt Loam

These are chiefly dark colored silt loam soils which have developed under bunch grass from silty wind-laid deposits mixed with small amounts of volcanic ash. The surface, 8 in. layer, is usually very dark grayish brown ($10YR^{3/2}$) grading with depth to a dark grayish brown ($10YR^{4/2}$) silt loam subsoil.

Burbank Loamy Sand

This is a dark-colored [surface is very dark grayish-brown ($10YR^{3/2}$); subsoil is dark grayish brown ($10YR^{4/2}$)], coarse-textured soil which is underlain by gravel. The surface soil is usually about 16 in. thick but can be 30 in. thick. The gravel content of the subsoil may range from 20 to 80 volume percent.

This is a dark colored [surface is very dark grayish brown ($10YR^{3/2}$); subsoil is dark grayish brown ($10YR^{4/2}$)], medium-textured soil which is underlain by gravelly material which may continue for many feet. The topography is generally level.

PHYSICAL, CHEMICAL, AND MINERALOGICAL PROPERTIES

The physical and chemical data determined for these soils are given in the following tables. The trace adsorption data show that all soils are highly selective for cesium and strontium. The data indicate the significant effects of organic matter and calcium carbonate on trace strontium adsorption. No significant correlation was obtained between trace adsorption, clay content and cation exchange capacity.

Mineralogical analyses by X-ray diffraction were conducted on clay fractions (0.2-2 and <0.2 microns) obtained from all horizons of Ritzville, Burbank and Ephrata soils. The clay mineral composition of all clay fractions is similar. Montmorillonite is the dominant mineral in the 0.2-2 micron fraction with lesser amounts of vermiculite and mica. Montmorillonite was the only clay mineral detected in all <0.2 micron fractions.

These data show that no detectable clay mineral weathering has occurred since the parent material was deposited. The only indication of soil weathering is the accumulation of calcium carbonate in the upper C horizons of all three soils.

Particle size analysis data of soil material from A_{12} horizons show that the soil type designations should be Ritzville loam, Burbank sands and Ephrata sandy loam. Previously the Ritzville was classed a silt loam and the Burbank a loamy sand.

Cation Exchange Capacity and Extractable Cations

		Exchangeable Cations me/100g				C.E.C.
		<u>Ca</u>	<u>Mg</u>	<u>K</u>	<u>Na</u>	Me/100g
Ritzville						
A ₁₁	0-2"	6.4	2.6	0.92	0.05	
A ₁₂	2-18"	5.5	2.4	0.67	0.10	12.8
B ₁	18-27"	6.3	9.2	0.31	0.13	11.9
C ₁	27-41"	16.9	7.1	0.43	0.91	11.2
C ^{ca}	41-72"	16.4	5.2	0.59	4.00	7.9
Burbank						
A ₁₁	0-3"	5.0	<0.1	0.67	0.05	
A ₁₂	3-0.6"	5.0	0.5	0.71	0.03	4.8
AC	16-22"	12.3	<0.1	0.51	0.05	4.9
AC ₂	22-30"	14.0	1.0	0.23	0.07	4.9
IC ₂	30"+	13.5	2.3	0.32	0.21	5.7
Ephrata						
A ₁₁	0-3"	4.5	1.0	1.41	0.03	
A ₁₂	3-12"	4.3	1.9	0.71	0.06	6.0
B ₁	12-15"	6.0	1.2	0.22	0.14	6.5
IB ₂	15-19"	8.6	2.4	0.14	0.34	7.6
IIC	19-24"	15.0	2.9	0.24	0.65	7.2

Particle Size, CaCO_3 , Fe_2O_3 and Organic Carbon

		Particle Size % of Mineral Fraction			CaCO_3 % of total	Fe_2O_3 % of total	Organic Carbon % of total
		Sand	Silt	Clay			
Ritzville							
A ₁₁	0-2"	23.3	64.1	12.6		1.50	
A ₁₂	2-18"	47.0	46.0	7.0	0.8	1.44	0.34
B ₁	18-27"	54.7	42.5	2.5	1.3	0.89	0.27
C ₁	27-41"	39.8	57.7	3.3	8.9	1.09	0.13
C _{ca}	41-72"	55.0	42.0	3.0	4.2	1.11	0.17
Burbank							
A ₁₁	0-3"	76.8	16.8	6.4		0.80	
A ₁₂	3-16"	91.2	7.6	1.2	0.7	0.84	0.16
AC	16-22"	88.7	10.6	0.7	1.6	0.62	0.14
AC ₂	22-30"	82.5	16.4	1.1	5.5	0.52	0.13
IC	30"+	79.8	16.2	4.0	3.5	0.39	0.20
Ephrata							
A ₁₁	0-3"	44.2	47.1	8.7		1.22	
A ₁₂	3-12"	59.0	36.9	4.1	0.57	1.25	0.18
B ₁	12-15"	56.4	39.3	4.3	0.70	1.20	0.13
IB ₂	15-19"	62.9	31.4	5.7	0.87	0.98	0.29
IIC	19-24"	92.1	5.4	2.5	3.28	0.46	0.26

Trace Strontium and Cesium Adsorption Characteristics from 0.2N NaCl

Equilibrium Distribution Coefficients (ml/g)

	No Pretreatment		Na-Based		Organic Carbon removed		Calcite removed		Organic C and Calcite removed	
	<u>Sr</u>	<u>Cs</u>	<u>Sr</u>	<u>Cs</u>	<u>Sr</u>	<u>Cs</u>	<u>Sr</u>	<u>Cs</u>	<u>Sr</u>	<u>Cs</u>
Ritzville										
A ₁₂	21	4869	109	4869	37	2750	189	5980	96	4069
B ₁	27	4332	131	4930	46	2154	257	6486	101	3152
C ₁	27	3111	84	5455	40	2608	71	4060	61	2592
C ^{ca}	24	3529	58	6169	43	3510	82	5174	66	2578
Burbank										
A ₁₂	12	2775	57	1964	20	2467	106	2540	45	3024
AC	15	2680	41	2571	28	2682	91	2448	61	2338
AC ₂	21	2573	53	3533	35	2513	67	2070	65	2036
IC	17	1588	48	1633	38	1155	81	1478	52	1010
Ephrata										
A ₁₂	12	3941	73	2385	21	3027	115	5230	44	2671
B ₁	12	4696	68	2410	22	2835	126	4071	57	2901
IB ₂	17	3491	82	1950	33	2402	166	2316	96	1850
IIC	23	1919	55	1553	55	2012	145	2423	70	836

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