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THE IN-SITU ADSORPTION OF ^{90}Sr
IN A SAND AQUIFER
by
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EXECUTIVE SUMMARY

Although there is a plethora of journal articles concerning how radionuclides might migrate in the subsurface from waste disposal sites (i.e. computer simulations and laboratory experiments), the scientific literature contains extremely few papers dealing with the fate of radioactive wastes in-situ. Since the lab experiments and computer simulations must be based on realistic assumptions about the in-situ hydrological and geochemical processes controlling the fate of such radionuclides, it is imperative that in-situ field studies be undertaken.

This paper reviews the in-situ migration and fate of the extremely toxic radionuclide, ^{90}Sr, which was disposed to unlined pits overlying a sand aquifer at the Chalk River Nuclear Laboratories (CRNL), 200 km northwest of Ottawa. After some 30 years of monitoring and study of the plume of radiostrontium in the aquifer, it is apparent that its retardation to only 3% of the ground water velocity is due to the specific adsorption of the radionuclide by oxide particles attached to the surface of the sand grains. The results have significant implications for how radionculide transport is modeled and how field and lab experiments are conducted.

RÉSUMÉ EXPLICATIF

Bien qu'une foule d'articles traitent de l'étude en laboratoire et par simulation de la migration des radionucléides depuis les sites d'élimination, les documents scientifiques sur le devenir in situ de ces déchets radioactifs sont extrêmement rares. Puisque les expériences réalisées en laboratoire et les simulations par ordinateur doivent reposer sur des hypothèses réalistes concernant les processus hydrologiques et géochimiques qui influent sur le sort des radionucléides, des études in situ s'imposent.

Ce rapport examine la migration et le devenir in situ du radionucléide extrêmement toxique ^{90}Sr, qui a été enfoui dans des fosses sans revêtement interne reposant sur un aquifère de sable aux Laboratoires nucléaires de Chalk River (LNCR), situés à 200 km au nord-ouest d'Ottawa. Au bout de trente ans de surveillance et d'étude, on a constaté que le ralentissement du panache de radio-strontium dans la couche aquifère, où il se déplace à une vitesse de 3 % de celle des eaux souterraines, semble dû à l'adsorption spécifique du radionucléide sur les particules d'oxyde à la surface des grains de sable. Cette découverte aura d'importantes répercussions sur la modélisation et l'étude sur le terrain et en laboratoire du transport des radionucléides.

RÉSUMÉ

Il y a plus de 30 ans, des déchets radioactifs liquides renfermant le radionucléide ^{90}Sr ont été enfouis dans une couche aquifère de sable peu profonde aux Laboratoires nucléaires de Chalk River, situés au Canada, au nord-ouest d'Ottawa. Depuis cette époque, des chercheurs surveillent la migration et le sort de ces substances. Des carottes de sable radioactifs ont été prélevées afin d'analyser la nature du processus d'adsorption qui a ralenti le déplacement du ^{90}Sr à sa vitesse actuelle de 3 % de celle des eaux souterraines.

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ABSTRACT

Over 30 years ago liquid radioactive wastes containing ^{90}Sr were disposed into a shallow sand aquifer at the Chalk River Nuclear Laboratories northwest of Ottawa, Canada and have since been monitored to observe their migration and fate. Cores of the radioactive sands have been recovered from the aquifer and analysed to determine the nature of the adsorption process by which the ^{90}Sr was retarded to its present rate of 3% of the ground water velocity.

Results indicate that the distribution ratio (R_D) and distribution coefficient (K_D) of ^{90}Sr are most strongly affected by spatial variations in the adsorption capacity of the aquifer, estimated by the specific surface area, rather than the variations in the concentration of competing cations. The adsorption of ^{90}Sr appears dominated by specific adsorption to Fe/Al/Mn oxide coatings on the sand grains rather than by electrostatic adsorption, i.e. classical cation exchange.

The consequences of this are that (1) the transport of the ^{90}Sr is probably kineticly controlled over the long term, therefore detailed solute transport modeling should account for such reaction kinetics; (2) linear adsorption models, such as the K_D approach, although presenting an informative account of solute partitioning, give only a first approximation of the adsorption process and (3) field and laboratory tests to estimate the adsorption parameters must be conducted at representative ground water velocities using aquifer materials that have been defined in great detail.

INTRODUCTION

The purpose of this paper is to describe the in-situ adsorption of ^{90}Sr from the analysis of cores taken from a contaminated fluvial sand aquifer at the Chalk River Nuclear Laboratories (CRNL) of Atomic Energy of Canada Ltd., 200 kilometres northwest of Ottawa, Ontario. Some of the results of an earlier survey reported in Jackson et al. (1984) were not reproducible. This article presents and analyses data

from a second and more detailed field survey of the ^{90}Sr plume at CRNL, shown in Figure 1, and reviews the results of previous work conducted at CRNL over the past ten years (Jackson and Inch, 1980 and 1983; Pickens et al., 1981; Patterson and Spoel, 1981).

ADSORPTION OF SOLUTES FROM GROUND WATER

Electrostatic and Specific Adsorption

It has long been known that the surfaces of oxides (e.g. $Fe(OH)_3$, MnO_2, clay minerals, feldspar, quartz, etc.) hydroxylate upon equilibration with water and that a surface charge develops from the dissociation of the hydroxide groups, i.e. $SOH = SO^- + H^+$, where S indicates a surface site and SO^- is the ionized site. Ionic solutes in ground water associate with such surfaces on aquifer grains by electrostatic and specific adsorption (Parks, 1975 and 1982).

Major-ion electrolytes, such as Ca^{2+} and Na^+, are sorbed by both mechanisms. In electrostatic adsorption, the cations accumulate in the outer part of the electrical double layer (edl) of charge created around the adsorbents; such reactions are considered rapid and reversible. Specifically-adsorbed ions, however, are frequently neither rapidly nor exchangeably sorbed (Parks, 1975), due to formation of non-electrostatic (e.g. covalent) rather than electrostatic (e.g. ionic) bonds. Furthermore, their sorption within the inner edl can even cause the surface charge of the adsorbent to be reversed. For example, Fuerstenau et al. (1981) studied the

adsorption of the alkaline-earth cations (Ba, Sr, Ca, Mg) on titanium oxide and showed that at concentrations $> 10^{-5}$ M these cations undergo strong specific adsorption resulting in charge reversal. In the formation of such surface complexes, the adsorption sites act as ligands and permit the treatment of the adsorption reactions as equivalent complexation reactions (Parks, 1982).

If the adsorption/desorption reactions are fast relative to the ground water velocity, the assumption of local chemical equilibrium is likely to be attained, provided that the reactions are fully reversible. However, several observers have noted that certain trace metal contaminants are very strongly sorbed by hydrous metal oxides which form coatings on sand grains in aquifers. Stollenwerk and Grove (1985) have noted, for example, that some Cr(VI) is retained by alluvium due to specific adsorption by ferric oxides; Ku et al. (1975) also noted sorption of chromium (and cadmium) by ferric oxides on aquifer materials on Long Island, N.Y. Similar irreversible adsorption of 20-40% of the ^{90}Sr in the outwash aquifer at CRNL has been documented by Jackson and Inch (1980 and 1983) and attributed to specific adsorption by Fe/Mn/Al oxide coatings on the aquifer grains. The detailed radiochemical analysis of mineral separates from the same aquifer conducted by Lyon and Patterson (1985) showed that the only quartz and feldspar grains having measurable nonexchangeable radioactivity were those with iron oxide coatings.

Adsorption Isotherms

Disregarding the mechanism of adsorption and assuming that local chemical equilibrium is attained during solute transport, the sorption of contaminant species "i" relative to its solute concentration may be described by one of several isotherms (Parks, 1982):

Linear:
$$S_i = K_{D,i} c_i \qquad (1)$$

Langmuir:
$$S_i = \frac{S_{max} K_{L,i} c_i}{1 + K_{L,i} c_i} \qquad (2)$$

Freundlich:
$$S_i = k c_i^{\,n} \qquad (3)$$

Ion exchange:
$$S_i = K_{i,j} S_j \, c_i / c_j \qquad (4)$$

where
S_i = mass of sorbed solute per unit mass of aquifer material or "adsorption density";

$K_{D,i}$ = solute distribution coefficient in mass sorbed per unit of mass of aquifer material divided solute concentration in solution;

c_i = solute concentration in solution in mass per unit volume of water;

S_{max} = maximum adsorption capacity;

$K_{L,i}$ = conditional equilibrium constant;

k =   Freundlich sorption coefficient;

n =   exponent indicating isotherm linearity;

$K_{i,j}$ =   selectivity coefficient for ion exchange reaction involving ionic species i and j.

(All parameters in consistent units).

Little is known about the shape of solute adsorption isotherms under field conditions. By laboratory experiment, both Couchat et al. (1980) and Reynolds et al. (1982) found radiostrontium adsorption on sands to be linearly dependent on solute concentration and therefore used a linear isotherm to describe radiostrontium transport in sand columns. Stollenwerk and Grove (1985) have shown that Cr(VI) sorption to alluvial materials taken from an aquifer at Telluride, Colorado may be described by a Langmuir isotherm, at least under laboratory batch conditions. However, Parks (1982) has pointed out that geochemical systems exist for which no single, simple isotherm can reproduce the experimental behaviour of the system, e.g. Hg(II) sorption on hydrous ferric oxide.

Cation Exchange

Reviews of radiostrontium sorption and transport have traditionally stressed the controlling roles of pH and Ca^{2+} concentration (e.g. Tamura, 1972; Francis, 1978). The detailed ion exchange studies of Sr on clay minerals by Wahlberg and colleagues (1965) served to underline the importance of the role of Ca^{2+} as the principal competing cation among hydrogeologists interested in

radionuclide transport and sorption. More recently, Johnston and Gillham (1984) pointed out that the natural, stable Sr content of soils and aquifer sediments and their associated ground waters can profoundly affect the measured value of the distribution ratio of ^{90}Sr, R_D, and hence K_D.

A simple procedure for evauating the potential influence of a competing ion on ^{90}Sr adsorption is via the mass action equation, written in this case for Ca^{2+}/Sr^{2+} ion exchange at ionized sites, SO^-:

$$(SO)_2Ca + {}^{90}Sr^{2+} = (SO)_2{}^{90}Sr + Ca^{2+} \qquad (5)$$
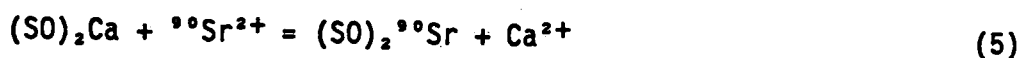
for which the selectivity coefficient is $K_{Sr,Ca}$ as in equation 4. A graph of log (aqueous) Ca^{2+} versus log R_D, obeying such mass action equilibria, should yield a straight line of slope -1 because

$$R_D = K_{Sr,Ca} \, S_{Ca}/c_{Ca} \qquad (6)$$

where $S_{Ca^{2+}}$ and c_{Ca} represent the concentrations of sorbed and aqueous Ca, respectively, as in equation 4. Should Na^+ be the principal competing ion, then the slope will equal -2 (see Tamura, 1972; Rafferty et al., 1981). Such an analysis assumes that only the two ions affect the value of R_D and that all data points represent sorbents with similar adsorption capacity.

METHODS OF SAMPLING AND ANALYSIS

For the past 30 years the migration of ^{90}Sr from the disposal of liquid wastes in "A" Disposal Area at CRNL has been carefully monitored. On the basis of this information, cores of the ^{90}Sr plume were collected during 1978, 1981 and 1985 along the central axis of the plumes and analysed by the methods shown in Figure 2.

Undisturbed samples of contaminated aquifer materials were recovered during the 1985 field survey from four locations within the aquifer materials (see Figure 1) using the cohesionless sediment sampler described by Munch and Killey (1985). The core tubes from each of the four cores (i.e. AA45, 38, 27 and 34) were cut into 6 cm sections, which were labelled as AA45/1, AA45/2 etc., and centrifuged within 24 hours of recovery in order to separate the interstitial waters from the aquifer materials. Subsequent total beta counting for ^{90}Sr on both phases yield the distribution ratio:

$$R_D = \frac{Bq/g}{Bq/mL} \qquad (7)$$

where Bq is the number of becquerels or radioactive distintegrations per second measured on a low beta counter corrected for detector efficiency using standard sources with geometries similar to that of the samples.

Interstitial or ground waters were analysed by atomic absorption spectrophotometry for cations that might compete with ^{90}Sr for adsorption sites. pH measurements were made with a combination electrode. Grain size analyses were conducted by dry sieving. Optical and x-ray diffraction methods were employed to identify potential mineral adsorbents. Cation-exchange capacities (CEC) were measured by a modification of the USGS method (Beetem et al., 1962) in which approximately one gram of aquifer material is placed in a test tube, saturated with 0.1 M strontium acetate, spiked with ^{85}Sr and gently rotated for 24 hours, after which time the phases are separated and counted in a well-type gamma counter.

Aquifer sediments contaminated with radiostrontium were subjected to a sequential extraction procedure to estimate the nature of the adsorption process. The procedure, based on that of Suarez and Langmuir (1976), sequentially treated a 2 gram sample of contaminated aquifer material with (1) 0.1 M $SrCl_2$ to estimate the exchangeable fraction of ^{90}Sr and (2) 0.1 M hydroxylamine hydrochloride (pH 1.5 buffer) to estimate that fraction associated with hydrous metal oxides of Al, Fe and Mn. The residual radioactivity is presumed fixed to the aquifer materials. The hydroxylamine hydrochloride solution was analysed by atomic adsorption spectrophotometry to determine the amount of metal oxides associated with the aquifer materials.

Specific surface areas of the aquifer materials were measured at CRNL with a Micromeritics Accusorb 2100E analyser by the BET method (Brunnauer, Emmett and Teller, 1938). Dried samples were placed in a flask and outgassed at 100-125°C and 10^{-4} mm Hg overnight. This flask

was then placed in a Dewar flask containing liquid nitrogen and equilibrated with increasing pressures of nitrogen, or, for very low surface areas, krypton gas. Specific surface areas were computed by measuring the volume of gas adsorbed as a function of applied pressure.

PREVIOUS WORK

The Hydrogeology of the Flow System

Detailed accounts of the physical and chemical hydrogeology of the ground water flow system at CRNL have been given by Pickens et al. (1981) and Jackson and Patterson (1982), respectively. Figure 3 shows the water table map of the flow system in June 1985.

The flow system is composed of a sequence of fluvial and aeolian sands (Catto et al., 1982) which are typically fine-grained (0.25 - 0.125 mm) with less than 2% silt and clay. These sands have a bulk density of about 1.7 g/cm^3, calculated from the average porosity (n=0.38) measured by Parsons (1960). Contained within the fine sand units are interstratified sand and silt units, which form aquitards within the local flow system as shown in Figure 4. The sands abut against a till ridge on top of which is situated "A" Disposal Area. This ridge is flanked by fine to coarse sands (perhaps reworked channel materials) and less permeable units. The hydrostratigraphic boundaries shown in Figure 4 were drawn on the basis of both core logging and information from a ground-probing radar study.

In general, flow is downward into and then horizontally through the sand aquifer south of "A" Disposal Area (see Jackson and Patterson, 1982, Figure 3). The hydraulic conductivity of the sands is in the range 2×10^{-4} to 2×10^{-5} m/s as determined by short and long-term aquifer response tests to a pumping well and by slug tests on piezometers (Pickens et al., 1981). Ground water velocities within the aquifer are of the order of 5-25 cm/day (Pickens et al., 1978), with a residence time in the flow system of between 10 and 20 years (Jackson and Inch, 1980).

Most (80-90%) sand-sized grains are either quartz or feldspar with minor amounts of biotite, vermiculite, sericite (weathered from feldspar), amphibole, muscovite and garnet present. Trace amounts of calcite (<0.01% by weight), organic carbon (~0.04%) and the hydrous oxides of aluminum (~0.04% by weight extractable metal), iron (~0.06%) and manganese (~0.001%) have also been measured in the aquifer sediments. Cation exchange capacities are of the order of 1 meq/100 g.

Approximately 10^8 L/year of acid precipitation is recharged to the flow system with approximately the following composition: pH~4, E_H~0.6V, sulfate ~10 mg/L, nitrate ~3 mg/L and chloride ~1 mg/L (Jackson and Inch, 1980). Within the flow system, ground waters undergo chemical evolution resulting in a pH increase from pH4 to pH7 due to silicate mineral weathering and carbonate mineral dissolution. This is accompanied by an E_H drop, presumably due to the oxidation of organic matter and ferrous iron, which produces anoxic conditions within the sand aquifer. In the oxic part of the

aquifer, biotite is weathered to vermiculite, simultaneously resulting in the formation of associated oxide coatings which become strong adsorbents of ^{90}Sr (Jackson and Patterson, 1982; Jackson and Inch, 1983; Inch and Killey, 1987).

^{90}Sr Studies

In 1954 about 7 m^3 of medium-level liquid radioactive waste containing about 2x10^{12} Bq of ^{90}Sr was released into a pit in the "A" Disposal Area lined with lime and dolomite. A further disposal in 1955 contained approximately 10^{13} Bq of ^{90}Sr, however no attempt was made to neutralize as before (Merritt and Mawson, 1967). In recent time ^{90}Sr has migrated through the flow system at a rate of approximately 3% of the ground water velocity.

The spatial distribution of ^{90}Sr within the aquifer during the summer of 1985, shown in plan view of Figure 1, is presented in cross-sectional view in Figure 5. It appears that the interstratified sand and silt units have not become significantly contaminated and that the majority of the radiostrontium is migrating in the fine sand unit. The mass of ^{90}Sr in the centre of Figure 5 is from both 1954 and 1955 disposals, the outlines of which are now indistinguishable.

Since the CRNL ground waters are undersaturated with respect to $SrCO_3$, $SrSO_4$ and $CaCO_3$ mineral phases, Jackson and Inch (1983) concluded that the retardation of ^{90}Sr must be due to adsorption and not precipitation. In their subsequent analysis of the 1981 data, Jackson et al. (1984) concluded that the "adsorption of ^{90}Sr is

dependent on the concentration of the principal competing cations (Ca^{2+} and stable Sr^{2+}) and the abundance of mineral adsorbents (e.g vermiculite) with high specific surface area". Inch and Killey (1987) found a strong correlation between radiostrontium adsorption and specific surface area in their examination of cores from "A" Disposal Area taken during the 1985 survey. They noted that small variations in the abundance of vermiculite in the aquifer sediments would have a major effect on radiostrontium adsorption because of the high specific surface area of this alteration product. However, they also perceived effects due to variations in the concentrations of ions that compete with ^{90}Sr for adsorption sites. The geochemical environment of the contaminated aquifer is shown schematically in Figure 6.

Figure 7 shows the relationship between the specific surface area and adsorbed radiostrontium in seven magnetically-separated samples of aquifer sediment collected during the 1981 survey. The samples labelled "Ferromag" consist of various grain size separates of biotite, vermiculite, hornblende and other dark minerals, whereas the samples labelled "Qtz + Fs" consist of quartz and feldspar, comprising 80-90% of all grains in the aquifer. The strong correlation between the two variables, $r = 0.98$, indicates that small increments in the abundance of high specific surface area minerals, such as vermiculite, can greatly affect the amount of adsorbed ^{90}Sr.

The evidence from which Jackson and Inch (1983) concluded that specific adsorption of ^{90}Sr by Fe/Mn/Al oxides was occurring was partly based on the sequential extraction of grain-size fractions of cores near AA38. When the extracting solutions were analysed for Fe,

Mn and Al as well as for ^{90}Sr, a strong correlation was demonstrated and is shown in Figure 8. The two maxima in the extracted metals are due to the similar maxima in specific surface area caused by the abundance of vermiculite in the coarse grain-size fraction and the usual effect of finer grain sizes on specific surface area.

RESULTS AND DISCUSSION

Ionic Competition and Specific Surface Area

In this section, the roles of ionic competition and variations in the specific surface area are assessed for their ability to affect the measured value of the distribution ratio (R_D). The geochemical data derived from the coring, centrifugation and analysis of the contaminated aquifer sediments during the 1985 survey are shown in Table 1. The locations of the boreholes (AA45, 38, 27 and 34) are shown in Figures 1 and 5.

Figure 9 shows the relationship between Ca^{2+} and R_D for the 1985 CRNL data based on equations (5) and (6), while the correlation coefficient matrix for the data of Table 1 is given in Table 2. Given the past preoccupation with the effects of competing ions on R_D values, it is somewhat surprising that neither Ca^{2+} nor Sr^{2+} (see Table 2) show a negative correlation with the R_D value for ^{90}Sr as would be expected if their competition with ^{90}Sr for adsorption sites controlled the distribution ratio. A previous study of this same plume (Jackson et al., 1984) had suggested that such was the case,

however, this earlier conclusion is not sustainable in the light of the evidence of the present study. Similarly, the pH correlation coefficient is negative (see Table 2) rather than positive, indicating that an increase in hydrogen ion activity would raise the R_D value, a result contrary to all experience. A final point of interest is that the correlation coefficient for Na^+ vs R_D is zero, indicating that sodium is an indifferent electrolyte vis-a-vis Sr^{2+}. Since road salt has entered this ground water flow system in substantial quantities, this result suggests that ^{90}Sr will not be significantly displaced by it.

Thus it appears that the traditional paradigm of radiostrontium adsorption being controlled by competing cations, whether Ca^{2+}, stable Sr^{2+} or H^+, does not satisfactorily explain the in-situ distribution ratios from the CRNL sand aquifer. To make this point explicit, two groups of data of internally similar competing cation concentrations are abstracted from Table 1 and presented as linear adsorption isotherms in Figure 10. The first group, Type A data in Figure 10, has Ca^{2+} concentrations in the range 1.0-1.4 meq/L, Sr^{2+} concentrations in the range 2.3-3.3 ueq/L, pH values in the range 5.6-6.1 and a strontium isotope ratio (SIR), as defined by the ratio stable: radioactive, of about 100,000 to 1. The second group, Type B data, has much lower concentrations of Ca^{2+} (0.6-1.0 meq/L) and Sr^{2+} (1.5-2.0 ueq/L), a higher pH range (6.2-6.4) and a lower SIR, about 10,000 to 1. All other factors being equal, one would expect that the data from the group indicating the lesser amount of ionic competition for adsorption sites, i.e. Type B data, would have a higher

distribution coefficient, K_D. As the figure shows, the converse is true.

The factor that is not equal in these two data groups and the factor that appears to play the dominant role in controlling the values of R_D is the adsorption capacity of the aquifer material for ^{90}Sr, here represented by the specific surface area. Figure 11 illustrates the dependency of the ^{90}Sr distribution ratio, R_D, on the specific surface area. It should be noted that those cores associated with Type B data that have been analysed for SSA show much lower SSA values than those cores associated with Type A data, with one exception. Unfortunately, CEC measurements have not proven sensitive enough to reveal this association.

Specific Adsorption of [90]Sr

The relative unimportance of ionic competititon in affecting the distribution ratio of ^{90}Sr merits comment. The results of the 1981 survey indicated that the R_D values were indeed negatively correlated with the concentrations of the competing cations; the squared correlation coefficients for Ca^{2+} and stable Sr^{2+} were approximately 0.6. However, the 1981 survey employed only 14 cores rather than the present 28 and therefore is likely less representative of the whole plume than is the 1985 survey. No account was taken in the 1981 survey of the variation in specific surface area, although the effect of minerals of high surface area, e.g. vermiculite, was apparent in the results (Jackson et al., 1984).

The absence of observable effects of ionic competition on the value of R_D during the 1985 survey may also derive from the preference of oxide minerals for Sr over Ca and Mg during specific adsorption (Fuerstenau et al., 1981). This preference arises from the fact that alkaline earth ions dehydrate upon specific adsorption and that the relative ease of dehydration determines the affinity sequence of adsorption, i.e. Ba>Sr>Ca>Mg. The ease by which hydrated alkaline earth cations are dehydrated is measured by the Born equation and is a function of ionic charge and radius (Moore, 1972). This absence of competitive effect then implies that the specific adsorption of ^{90}Sr by Fe/Al/Mn oxides may well account for more than the 20-40% heretofore estimated.

This point has also been made by Killey and Munch (1987) who found that only 35% of ^{90}Sr was exchangeable with 0.1 M MgCl$_2$ but over 50% released by a treatment that partially dissolves the oxide coatings. This estimate of 35% and the previous estimate of 80% exchangeable ^{90}Sr made by Jackson and Inch (1983) are probably overestimates because the SrCl$_2$ treatment used by Jackson and Inch has recently been shown to cause a decrease in surface area indicating the dissolution of the oxide coatings and the subsequent release of ^{90}Sr (Inch and Killey, 1987). Thus it appears that specific adsorption of radiostrontium by oxide coatings on sand grains is controlling the long-term transport of the radionuclide through the aquifer system. This same conclusion was recently reached by Melnyk et al. (1984) in their simulation of ^{90}Sr transport from glass blocks of radioactive wastes experimentally placed in the same aquifer at CRNL.

Measurement of the Adsorption Parameters

During the past ten years a series of estimates of the parameters describing the partitioning of ^{90}Sr between aquifer materials and ground water in the Lower Perch Lake Basin have been made. These estimates are summarized in Table 3. The methods used include (1) coring of the contaminated aquifer, as in the case of this present paper, (2) laboratory batch methods, (3) field tracer test methods and (4) in-situ adsorption isotherms based on cores of contaminated aquifer materials. The values of R_D and K_D are compared on the assumption of linear adsorbate partitioning between the aquifer grains and the associated ground waters.

The first set of estimates listed in Table 3 (i.e. A, B, C) are distribution ratios from cores of the aquifer sediments taken by procedures used in this present paper. The comment statements are based upon specific surface area measurements reported in this paper and in Inch and Killey (1987) and estimates based upon these two sources (e.g. "SSA probably >0.5"). The values seem to be largely a function of the specific surface area of the aquifer materials. The batch R_D estimates (D, E) in all probability also reflect this effect. The aquifer materials of Patterson and Spoel (1981) (see their Table 3) show the distribution ratios and the sum of the exchangeable cations at a maximum in the smallest grain size fraction, indicating the importance of the specific surface area on cation adsorption.

The value (E) for the ^{85}Sr breakthrough curve of Pickens et al. (1981) was derived from comparison with a (non-reactive) iodide breakthrough curve during a single-well injection/withdrawal tracer test conducted at the leading edge of ^{90}Sr plume. This value of 3.4 mL/g is lower than the others because of the high ground water velocities (1-13 m/day) created by the test in comparison with natural velocities in the aquifer (5-25 cm/day). Under such conditions local chemical equilibrium was not likely attained and the kinetics of the adsorption/desorption process were apparent in breakthrough and recovery curves.

The final value (G) is for the least squares fit of the linear adsoprtion isotherm to all 28 data points for the 1985 survey of the ^{90}Sr plume. This value of 6.5 mL/g represents an average partition coefficient for the whole aquifer. As Figure 10 has shown, it ignores the important changes in adsorption capacity within the apparently homogeneous aquifer that govern the local partitioning of the radiostrontium.

Probably the most reliable values would be obtained by natural gradient tracer tests, conducted in the principal hydrostratigraphic units, which would be defined on the basis of their adsorption capacities. Such tests could be supplemented by long-term column tests conducted at appropriate ground water velocities, i.e. 5-25 cm/day. From such tests rate constants for the adsorption and desorption reactions would be calculated.

Two techiques used in assessing the adsorption of radionuclides by aquifer materials have been found wanting during the course of

these studies. Firstly, the data of Inch and Killey (1987) demonstrate that 0.1M SrCl$_2$ solutions used to estimate the exchangeable fraction of adsorbed ^{90}Sr cause a significant loss of surface area from the aquifer grains, implying dissolution of the oxide coatings and release of adsorbed ^{90}Sr. Consequently, such tests lead to an overestimation of the fraction of exchangeable ^{90}Sr and an underestimation of the oxide-associated fraction. Column tracer tests conducted at the appropriate ground water velocity, redox environment and ionic strength would likely give more reliable results. Secondly, the cation exchange capacity tests conducted on the aquifer materials discussed in this and previous papers have not revealed the substantial variation in adsorption capacity indicated by both the distribution ratios and the specific surface area measurements. SSA measurements appear to be of the first importance in analysing the variability in the adsorption capacity of aquifer materials.

Implications for Solute Transport Modeling

Contaminant transport codes are now widely used to describe the migration and fate of pollutants in ground water. Such codes contain models accounting for the adsorption of the solute from ground water. Frequently the model is in the form of an adsorption isotherm, in particular the linear isotherm (eq. 1), describing the equilibrium partitioning of the solute between sorbed and solution phases (e.g.

Pickens et al., 1981). Under certain circumstances it may be possible to employ ion exchange mass action equilibria to describe the solute transport as in equation 4 (e.g. Kipp et al., 1986). Some recent models have incorporated not only a linear isotherm but also kinetic expressions describing slow solute adsorption and/or desorption reactions (e.g Melnyk et al., 1984).

The attractiveness of the linear isotherm or K_D approach is its simplicity of form. While it provides useful information of contaminant transport, the results can only be considered first approximations of the transport processes of trace metal ions in typical granular aquifer systems. Such systems frequently contain a heterogeneous distribution of adsorbents and varying aqueous chemical compositions along the flow path and display slow contaminant sorption kinetics due to the occurrence of specific adsorption reactions. In the present study, the heterogeneous spatial distribution of the adsorbents leads to significant variations in the value of K_D (see Figure 10) in an aquifer system that appeared to the naked eye to be homogeneous.

Other isotherms may of course be used. The Freundlich isotherm (eq. 3) has a variable slope on a log S_i vs. log c_i plot that approaches unity only if the adsorption sites are of uniform energy (Sposito, 1981). This property can be used to test adsorption data to determine the uniformity of the aquifer materials as adsorbents. Figure 12 shows two Freundlich isotherms. The first (curve A) is for all 28 cores from the whole of the aquifer and therefore is a least

squares fit to data from (1) weathered, fine- to medium-grained sands containing 2% or 3% vermiculite (AA45), (2) unweathered, fine-grained grey sand with no ferromagnesian minerals (AA38) and (3) unweathered, fine- to medium-grained, grey sand rich in biotite (AA27 and AA34). The Freundlich exponent (i.e. isotherm slope) for the whole aquifer isotherm is 0.79. However, if only the data from the similar cores taken from the front of the plume (i.e. AA27 and AA34) are considered, the isotherm slope (curve B) increases to 0.92. This increased uniformity is partly due to the decrease in standard deviation of SSA from 0.55 m²/g (whole aquifer) to 0.09 m²/g (AA27 and 34) and partly to the exclusion of the vermiculitic sands (AA45) which have high adsorption capacities and, probably, adsorption sites with a wide spectrum of site energies.

Alternatively, it may be possible to use mass action equilibria should (1) the adsorption reactions be instantaneous and reversible, (2) the selectivity coefficients for the various adsorbates be constant throughout the aquifer system for the aqueous geochemical conditions considered and (3) ions considered explicitly in the mass action equilibria comprise the total competing ion concentration and the sum of these ions when sorbed comprises the cation exchange capacity (Reardon, 1981; Valocchi et al., 1981; Valocchi, 1984; Kipp et al., 1986). In the present case, it has been demonstrated that a substantial fraction of the adsorbed ^{90}Sr is specifically adsorbed to the aquifer sediments and not readily desorbed (Jackson and Inch,

1983; Killey and Munch, 1987) and that adsorption onto the oxide coatings on the sand grains at CRNL requires 7 to 9 years to reach 95% of equilibrium (Melnyk et al., 1984). Furthermore, it is questionable whether selectivity coefficients would be constant over the range of ionic concentrations throughout the ^{90}Sr plume (Morel, 1983).

The most promising approach to modeling the transport and retardation of ^{90}Sr in the aquifer at CRNL appears to be that of Melnyk et al. (1984), who have developed a coupled linear adsorption isotherm and kinetic model. The electrostatic adsorption of ^{90}Sr to the aquifer materials is simulated by a linear adsorption isotherm, which may be justified on the evidence of the relative unimportance of competing ion competition (see Figure 10). The specific adsorption of the radiostrontium to the Fe/Mn/Al oxides is described by a kinetic model. Their analysis leads them to the conclusion that the long-term transport of the ^{90}Sr is controlled by the kinetics of the specific adsorption reaction. An improved model might allow for a stochastic description of the adsorption constant, K_{ads}, reflecting the variability in the specific surface area and, hence, the adsorption capacity of the aquifer materials.

CONCLUSIONS

Analysis of cores of contaminated aquifer materials collected from along the centre line of a ^{90}Sr plume at the Chalk River Nuclear Labs leads to the following conclusions:

1. The principal geochemical factor controlling the variation in the distribution ratio (R_D) and distribution coefficient (K_D) of [9][0]Sr in the aquifer and, hence, its retardation during solute transport is the spatial variability in the adsorption capacity of the aquifer materials as measured by the specific surface area of the aquifer sediments. Such variability is apparent in the nonlinearity of Freudlich isotherms. Less important are the effects of ionic competition between [9][0]Sr and other cations (Ca^{2+}, stable Sr^{2+} and H^+), for adsorption sites.

2. The adsorption and long-term retention of [9][0]Sr is probably dominated by the specific adsorption of the radionuclide by Fe/Al/Mn oxide particles on the sand grain surfaces and not by electrostatic adsorption involving simple ion exchange as previously reported.

3. The best method of measuring the adsorption parameters would seem to be a natural gradient tracer test supplemented with long-term laboratory column tests conducted at in-situ ground water velocities. Cation exchange capacity tests have proved to be of very limited use in identifying the spatial variability in adsorption capacity and sequential desorption tests appear to give misleading information on the amount of exchangeable [9][0]Sr.

4. Since specific adsorption results in slow adsorption and desorption rates, the modeling of [9][0]Sr transport in this aquifer system must account for the kinetics of the specific adsorption reaction. Linear adsorption isotherms adequately define the long-term partitioning of the solute between aquifer sediments

and ground water, but can only be used to obtain a first approximation of the long-term transport of the solute.

ACKNOWLEDGEMENTS

REFERENCES

Beetem, W.A., Janzer, V.J. and Wahlberg, J.S., 1962. Use of cesium-137 in the determination of cation exchange capacity. U.S. Geological Survey Tech. Bull., 1140-B.

Brunnaer, S., Emmett, P.H. and Teller, E., 1938. The adsorption of gases in multimolecular layers, J. Am. Chem. Soc., 60: 309-319.

Catto, N.R., Patterson, R.J. and Gorman, W.A., 1982. The late Quaternary geology of the Chalk River region, Ontario and Quebec. Can. J. Earth Sci., 19:1218-1231.

Couchat, P., Brissaud, F. and Gayraud, J.P., 1980. A study of strontium-90 movement in a sandy soil. Soil Sci. Soc. Amer.J., 44:7-13.

Francis, C.W., 1978. Radiostrontium movement in soils and uptake in plants. Technical Information Center, U.S. Department of Energy, Oak Ridge, TN, U.S.A.

Fuerstenau, D.W., Manmohan, D. and Raghavan, S., 1981. The adsorption of alkaline-earth metal ions at the rutile/aqueous solution interface. In: P.H. Tewari (Editor), Adsorption from Aqueous Solutions. Plenum Press, New York, U.S.A., pp. 93-117.

Inch, K.J. and Killey, R.W.D., 1987. Surface area and radionuclide sorption in contaminated aquifers. Water Poll. Res. J. Canada, 22:85-98.

Jackson, R.E. and Inch, K.J., 1980. Hydrogeochemical processes affecting the migration of radionuclides in a fluvial sand aquifer at the Chalk River Nuclear Laboratories. Scientific Series No. 104, Inland Waters Directorate, Environment Canada, Ottawa, Canada, 58 pp.

Jackson, R.E. and Patterson, R.J., 1982. Interpretation of pH and Eh trends in a fluvial-sand aquifer system. Water Resources Res., 18:1255-1268.

Jackson, R.E. and Inch, K.J., 1983. Partitioning of Strontium-90 among aqueous and mineral species in a contaminated aquifer. Environ. Sci. Technol., 17:231-237.

Jackson, R.E., Inch, K.J., Patterson, R.J. and Killey, R.W.D., 1984. Field measurement and geochemical interpretation of ^{90}Sr adsorption in a contaminated aquifer. Eos, 65:207.

Johnston, H.M. and Gillham, R.W., 1984. Natural Sr concentrations and Kd determinations. J. Geotechnical Engin., 110:1459-1472.

Killey, R.W.D. and Munch, J.H., 1987. Radiostrontium migration from a 1953-54 liquid release to a sand aquifer. Water Poll. Res. J. Canada, 22:107-128.

Kipp, K.L. Jr., Stollenwerk, K.G. and Grove, D.B., 1986. Ground water transport of strontium-90 in a glacial outwash environment. Water Resources Res., 22:519-530.

Ku, H.F.H., Katz, B.G., Sulam, D.J. and Krulikas, R.K., 1978. Scavenging of chromium and cadmium by aquifer material - South Farmingdale-Massapequa area, Long Island, New York. Ground Water, 16:112-118.

Lyon, K.E. and Patterson, R.J., 1985. Retention of ^{137}Cs and ^{90}Sr by mineral sorbents surrounding vitrified nuclear waste. Scientific Series No. 148, Inland Waters Directorate, Environment Canada, Ottawa, Canada, 19 pp.

Melnyk, T.W., Walton, F.B. and Johnson, L.H., 1984. High-level waste glass field burial test: leaching and migration of fission products. Nuclear and Chemical Waste Management, 5:49-62.

Merritt, W.F. and Mawson, C.A., 1967. Experiences with ground disposal at Chalk River. In: Disposal of Radioactive Wastes into the Ground, International Atomic Energy Agency, Vienna, Austria, pp. 79-93.

Moore, W.J., 1972. Physical Chemistry (fourth edition). Prentice-Hall Inc., Englewood Cliffs, New Jersey, U.S.A., 977 pp.

Morel, F.M.M., 1983. Principles of Aquatic Chemistry. Wiley-Interscience, New York, U.S.A., 446 pp.

Munch, J.H. and Killey, R.W.D., 1985. Equipment and methodology for sampling and testing cohesionless sediments. Ground Water Monitoring Review, 5:38-42.

Parks, G.A., 1975. Adsorption in the marine environment. In: J.P. Riley and G. Skirrow (Editors), Chemical Oceanography, Academic Press, New York, U.S.A., 1(2):241-308.

Parks, G.A., 1982. Problems and opportunities in adsorption research. Chemistry in Australia, 49:389-395.

Parsons, P.J., 1960. Movement of radioactive wastes through soil. Part 1: Soil and ground water investigations in lower Perch Lake Basin. Atomic Energy of Canada Ltd., Chalk River, Ontario, Canada, AECL Report No. 1038.

Patterson, R.J. and Spoel, T., 1981. Laboratory measurements of the strontium distribution coefficient Kd-Sr for sediments from a shallow sand aquifer. Water Resources Res., 17:513-520.

Pickens, J.F., Cherry, J.A., Gillham, R.W. and Merritt, W.F., 1978. Field studies of dispersion in a shallow sandy aquifer. In: Proceedings, Invitational Well Testing Symposium, Lawrence Berkeley Laboratory, Berkeley, California, LBL-7027, pp. 55-62.

Pickens, J.F., Jackson, R.E., Inch, K.J. and Merritt, W.F., 1981. Measurement of distribution coefficients using a radial injection dual-tracer test. Water Resources Res., 17:529-544.

Rafferty, P., Shiao, S.-Y., Binz, C.M. and Meyer, R.E., 1981. Adsorption of Sr(II) on clay minerals: effects of salt concentration, loading and pH. J. Inorg. Nucl. Chem., 43:797-805.

Reynolds, W.D., Gillham, R.W. and Cherry, J.A., 1982. Evaluation of distribution coefficients for the prediction of strontium and

cesium migration in a uniform sand. Canadian Geotech. J., 19:92-103.

Sposito, G., 1981. The Thermodynamics of Soil Solutions. Oxford, Clarendon Press, U.K., 223 pp.

Stollenwerk, K.G. and Grove, D.B., 1985. Adsorption and desorption of hexavalent chromium in an alluvial aquifer near Telluride, Colorado. J. Environ. Qual., 14:150-155.

Suarez, D.L. and Langmuir, D., 1976. Heavy metal relationships in a Pennsylvania soil. Geochim. Cosmochim. Acta, 40:589-598.

Tamura, T., 1972. Sorption phenomena significant in radioactive-waste disposal. In: T.D. Cook (Editor), Underground Waste Management and Environmental Implications, Amer. Assoc. Petrol. Geologists Memoir No. 18, Tulsa, OK, U.S.A., pp. 318-329.

Wahlberg, J.S., Baker, J.H., Vernon, K.W. and Dewar, R.S., 1965. Exchange adsorption of strontium on clay minerals. U.S. Geological Survey Bull. 1140-C.

TABLES

1. Chemical analysis of 1985 core subsections, each of 6 cm length. pH, Na^+, Ca^{2+}, Sr^{2+}, Mg^{2+} and $aq^{90}Sr$ are all aqueous concentrations of the interstitial waters. $Ads^{90}Sr$ is the adsorbed concentration on the aquifer grains. R_D is the ratio of adsorbed to aqueous ^{90}Sr in the core subsection.

2. Correlation coefficients for geochemical variables from 1985 survey of ^{90}Sr plume.

3. Estimates of K_D and mean values of R_D by various methods and their standard deviation(s) and sample size (n_s). SSA indicates specific surface area in m^2/g.

Table 1. Chemical analysis of 1985 core subsections, each of 6 cm length. pH, Na⁺, Ca²⁺, Sr²⁺, Mg²⁺ and aq⁹⁰Sr are all aqueous concentrations of the interstitial waters. Ads⁹⁰Sr is the adsorbed concentration on the aquifer grains. R_D is the ratio of adsorbed to aqueous ⁹⁰Sr in the core subsection.

Core No.	pH	Na⁺ (mg/L)	Ca²⁺ (mg/L)	Mg²⁺ (mg/L)	Sr²⁺ (µg/L)	ads⁹⁰Sr (Bq/g)	aq⁹⁰Sr (Bq/mL)	R_D (mL/g)
AA45/1	5.8	37.8	24.1	12.4	113	52.4	6.8	7.7
3	6.1	31.3	24.8	12.4	105	58.2	5.9	9.9
4	6.1	32.8	23.1	11.7	105	70.7	5.6	12.6
5	5.8	31.3	21.6	11.1	123	54.8	4.0	13.7
7	6.0	31.6	24.2	10.9	99	66.4	6.6	10.1
AA38/1	5.8	27.3	21.3	9.2	143	37.9	2.4	15.8
2	5.8	28.2	27.0	11.0	113	41.5	3.0	13.8
3	5.9	30.7	21.3	9.6	114	50.8	4.6	11.0
4	5.6	28.2	20.6	9.5	124	83.8	5.9	14.2
5	5.8	28.8	23.8	10.5	115	39.2	6.5	6.0
6	5.9	31.6	21.3	9.8	117	62.5	8.3	7.5
8	5.9	36.3	17.4	8.6	108	185.0	26.7	6.9
AA27/1	6.1	13.7	23.6	8.9	68	249.3	29.9	8.3
2	6.2	15.2	20.6	7.1	71	252.1	31.6	8.0
3	6.2	11.2	28.0	9.5	72	186.4	28.0	6.7
4	6.2	13.9	-	-	84	133.9	19.6	6.8
5	6.2	15.2	17.7	6.9	84	91.3	18.8	4.9
6	6.2	13.9	19.1	7.1	87	46.1	9.0	5.1
7	6.2	15.8	16.9	6.6	88	18.8	2.2	8.5
8	6.2	18.6	18.4	7.4	90	7.0	0.8	8.8
AA34/1	6.3	43.1	19.9	8.6	77	132.5	23.5	5.6
2	6.4	46.5	16.0	7.2	79	171.2	32.2	5.3
3	6.4	34.4	18.1	8.3	65	284.7	39.4	7.2
4	6.3	49.9	18.1	8.8	72	235.2	45.7	5.1
5	6.4	25.7	16.8	7.5	75	250.1	38.5	6.5
6	6.2	23.3	13.5	6.0	79	234.9	39.3	6.0
7	6.2	21.1	15.0	6.1	107	189.7	29.2	6.5
8	6.3	35.3	17.4	7.3	107	137.7	18.1	7.6

Table 2. Correlation coefficients for geochemical variables from 1985 survey of ^{90}Sr plume.

	pH	Na	Ca	Mg	Sr	R_D
pH	1.00	-0.01	-0.50	-0.61	-0.82	-0.67
Na		1.00	-0.10	0.33	0.17	0.00
Ca			1.00	0.81	0.26	0.44
Mg				1.00	0.48	0.52
Sr					1.00	0.68
R_D						1.00

Table 3. Estimates of K_D and mean values of R_D for ^{90}Sr by various methods and their standard deviation(s) and sample size (n_s). SSA indicates specific surface area in m^2/g.

	Method	K_D	R_D	s	n_s	Comments
		(mL/g)				
A	cores of aquifer sediments	–	11	6.8	11	SSA probably > 0.5
B	cores taken during present study	–	8.4	3.1	28	SSA: 0.2 – 3.0
C	cores taken during tracer test	–	5.7	1.6	36	SSA probably <0.5
D	batch tests for whole aquifer	–	12	7.5	60	
E	batch tests for tracer test site	–	5.7	3.5	7	SSA probably <0.5
F	breakthrough curve, tracer test	3.4	–	0.8	9	vs I⁻ breakthrough
G	in-situ isotherm	6.5	–	–	28	least squares fit, y=ax

Sources of data: A – Jackson and Inch, 1980; B and G – this paper; C, E and F – Pickens et al., 1981; D – Patterson and Spoel (1981).

FIGURE CAPTIONS

Fig. 1.   Map of the ^{90}Sr plume migrating from A Disposal Area, Chalk River Nuclear Laboratories, in plan view. Note the north-south cross section and the location of cores AA45, 38, 27 and 34.

Fig. 2.   Flow chart of methods of analysis of contaminated aquifer materials.

Fig. 3.   Water table map of the ground water flow system near A Disposal Area, CRNL. The contour interval is in metres above mean sea level.

Fig. 4.   North-south cross section of the aquifer system beneath and down gradient of A Disposal Area, CRNL. Note the tenfold vertical exaggeration of scale. The orientation of the cross section is shown in Figure 1.

Fig. 5.   North-south cross section of ^{90}Sr plume down gradient of A Disposal Area, CRNL. Note the ten-fold vertical exaggeration of scale. The location of the four cores collected in 1985 is shown by diamond symbols.

Fig. 6.   A schematic of the geochemical environment of the ^{90}Sr plume based upon the 1978 and 1981 field studies. Note the five-fold vertical exaggeration of scale. The geochemically weathered part of the aquifer is immediately beneath and down gradient of A Disposal Area. Vm indicates vermiculite abundance, CEC indicates the cation exchange capacity, pH indicates the ground water pH and SSA indicates the specific surface area of the aquifer materials.

Fig. 7. Correlation (r=0.98) between specific surface area and adsorbed ^{90}Sr associated with ferromagnesian ("Ferromag") and quartz and feldspar ("Qtz + Fs") separates further separated on the basis of grain size for an aquifer sample from approximately the 155 metre elevation near borehole AA38. The "Ferromag" separates are principally composed of biotite and vermiculite particles; sample 1 is principally vermiculite. Error bars indicate one standard deviation above and below mean value (from Inch and Killey, 1987).

Fig. 8. Amounts of ^{90}Sr, Fe, Mn and Al extracted with acidified hydroxylamine hydrochloride from various grain-size fractions of contaminated aquifer materials following extraction of the same sample with 0.1M $SrCl_2$. The core is from the 150 metre level near AA38 (from Jackson and Inch, 1983). Dpm/g may be converted to Bq/g by dividing the former by 60.

Fig. 9. Correlation between the distribution ratio for ^{90}Sr, R_D, and the concentration of dissolved Ca^{2+} in the interstitial waters associated with the cores. The positive correlation between these two variables indicates that ionic competition between Ca^{2+} and ^{90}Sr for adsorption sites is only a minor factor in determining the partitioning of ^{90}Sr in the aquifer.

Fig. 10. Linear adsorption isotherms for type A and B data. Type A data indicate a geochemical environment in which competition between hydrogen, calcium, stable strontium and radio-strontium ions is greater than with type B data. SIR

represents the strontium isotope ratio of stable strontium to ^{90}Sr ions. SSA represents the mean specific surface area of aquifer materials associated with these ground waters.

Fig. 11. Correlation of the specific surface area (SSA) of aquifer materials from the ^{90}Sr plume with the distribution ratio for ^{90}Sr, R_D, for the same materials.

Fig. 12. Freundlich isotherms for all 28 subcores taken from the whole aquifer (isotherm A) and for only those subcores associated with the AA27 and 34 core series taken from the front of the ^{90}Sr plume (isotherm B). The greater uniformity in adsorption capacity and adsorbent geochemistry in the latter core series is evident in the greater linearity of isotherm B.
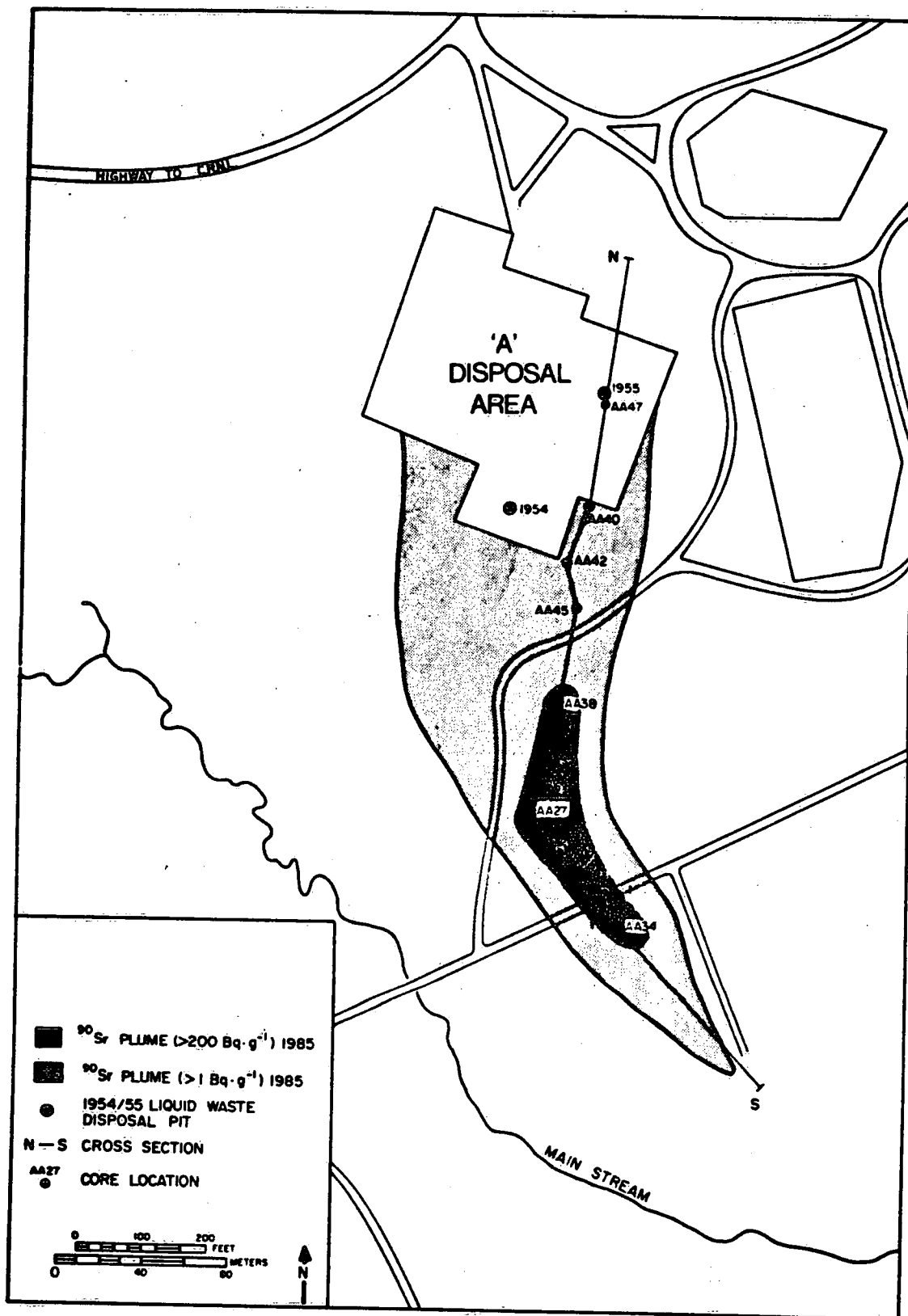
'A'
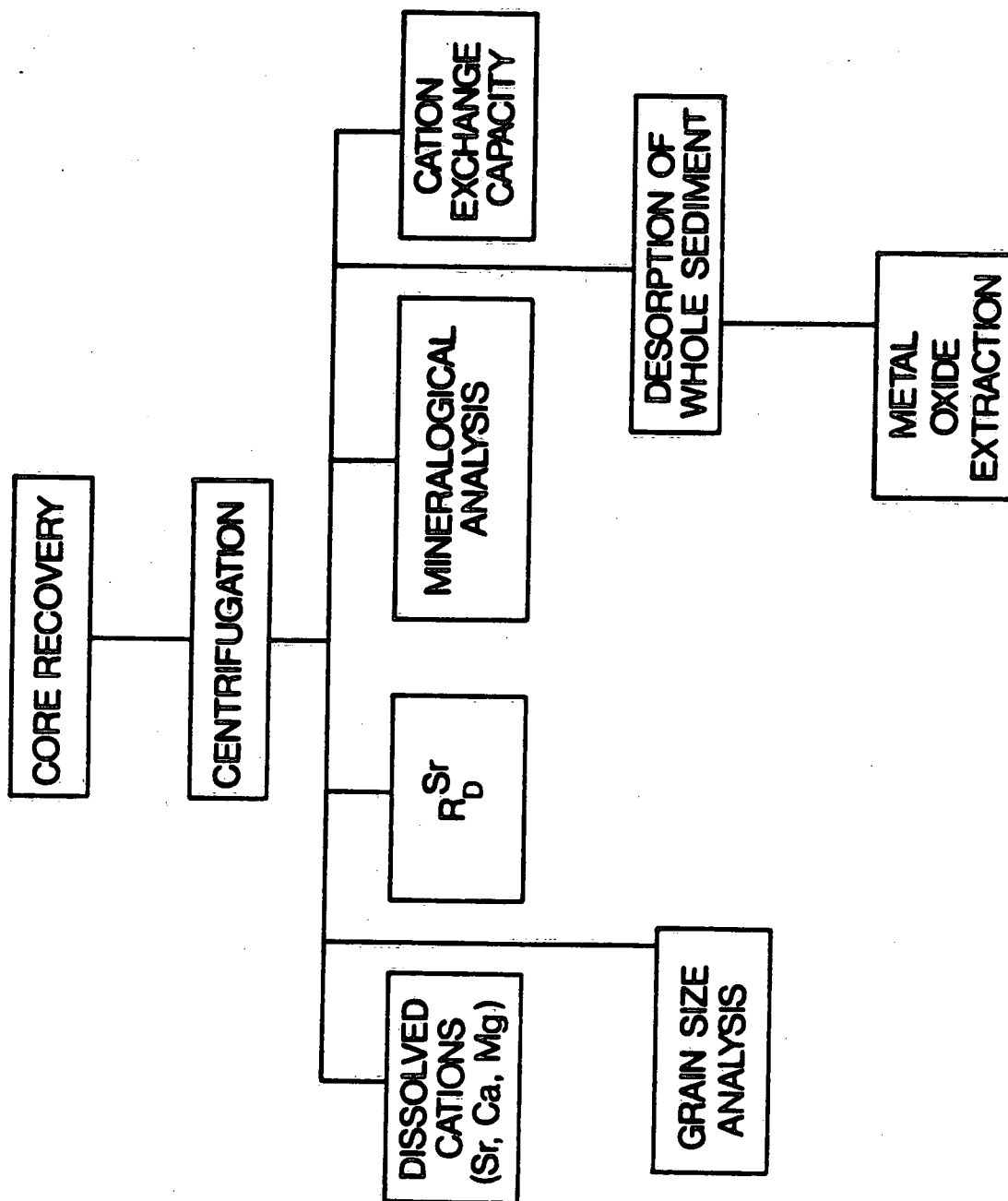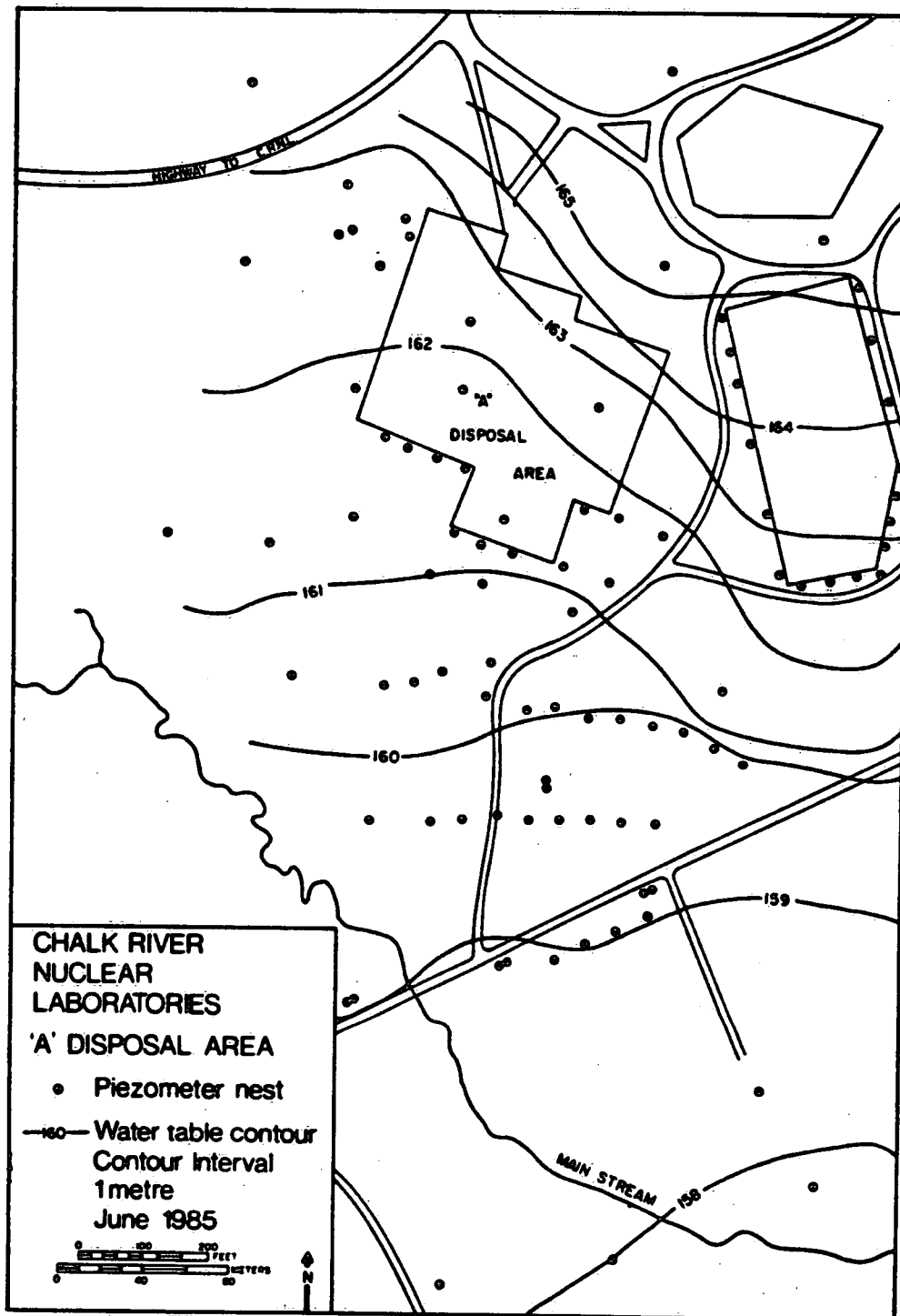DISPOSAL
AREA

HIGHWAY TO CRNL

N

S

MAIN STREAM

1955
AA47

1954

AA40

AA42

AA45

AA38

AA27

AA34

^{90}Sr PLUME (>200 Bq·g^{-1}) 1985

^{90}Sr PLUME (>1 Bq·g^{-1}) 1985

1954/55 LIQUID WASTE
DISPOSAL PIT

N—S   CROSS SECTION

AA27   CORE LOCATION

0   100   200
FEET

0   40   80
METERS

N

Figure 1

Figure 2.

CHALK RIVER
NUCLEAR
LABORATORIES

'A' DISPOSAL AREA

• Piezometer nest

—160— Water table contour
Contour interval
1 metre
June 1985

Figure 3

Figure 4

S
SOUTH

PLSi5

1965

AA34

AA27

AA38

AA45

AA42

AA40

AA47

1955
DISPOSAL

N
NORTH

170

165

160

155

150

145

WATER TABLE

⁹⁰Sr PLUME (>100 Bq·g⁻¹)

⁹⁰Sr PLUME (>10 Bq·g⁻¹)

⁹⁰Sr PLUME (>1 Bq·g⁻¹)

Interstratified sands & silts

1985 cores

0 10 20 30 40 50 METERS

Figure 5

Weathered Zone:
Vm ~ 2%   pH < 6.0
CEC ~1.2 meq/100g SSA: 0.5 – 2.5 m²/g

WATER TABLE

⁹⁰Sr FRONT 1986

ROAD

A DISPOSAL AREA

Unweathered Zone :
Vm ~ 0.5%   pH > 6.0
CEC ~ 1.1 meq/100g   SSA < 0.5 m²/g

0   25   50 metres

10

5

0

Figure 6
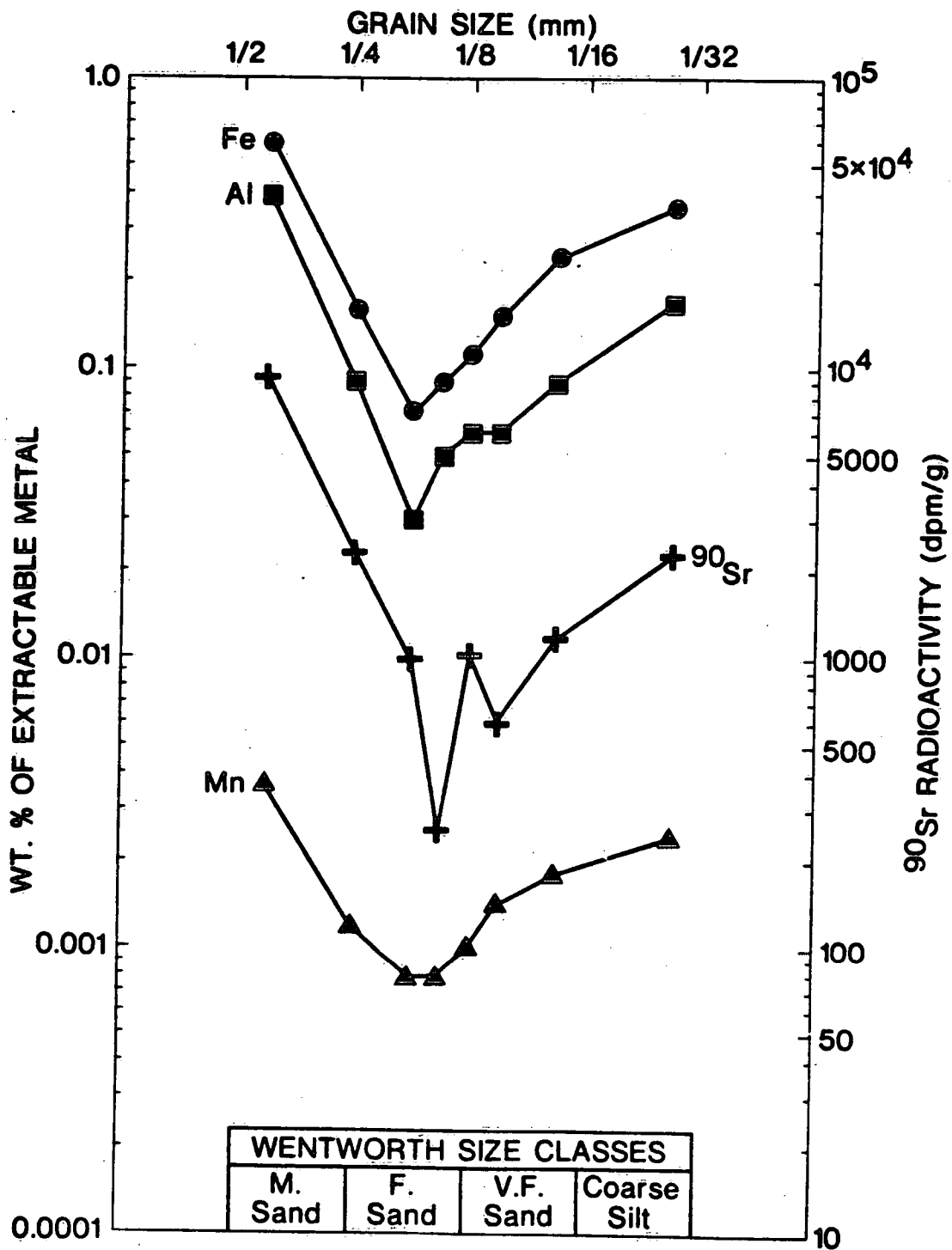
Figure 7

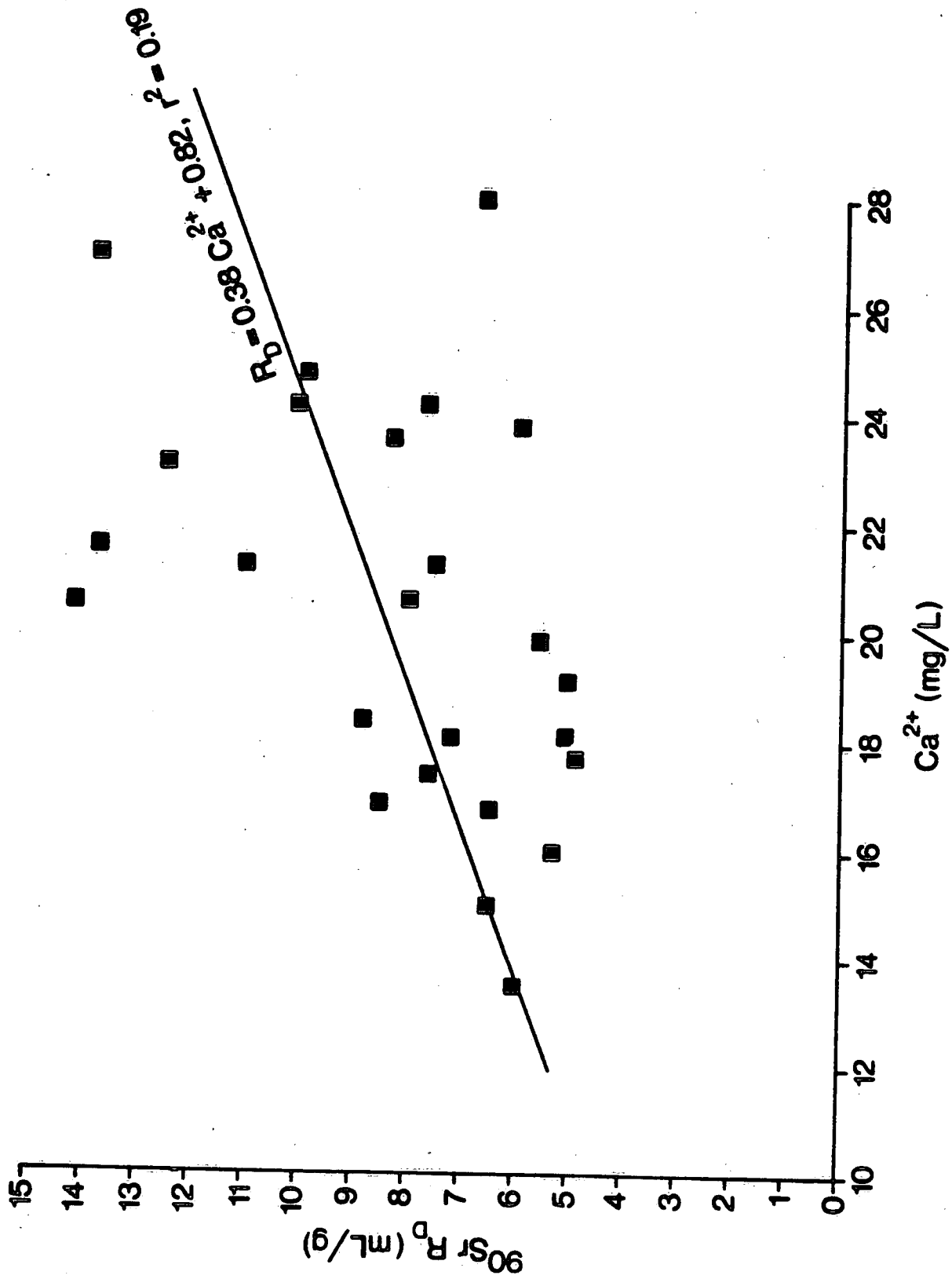Figure 8

$R_D = 0.38 Ca^{2+} + 0.82$, $r^2 = 0.19$

Ca^{2+} (mg/L)

^{90}Sr R_D (mL/g)

Figure 9

Figure 10

Figure 11

A) Freundlich isotherm for whole aquifer: $S = 13.2c^{0.79}$

B) Freundlich isotherm for front of ^{90}Sr plume $S = 8.3c^{0.92}$

● cores AA 27 and 34

LOG AQUEOUS ^{90}Sr (Bq/g)

LOG ADSORBED ^{90}Sr (Bq/g)

Figure 12