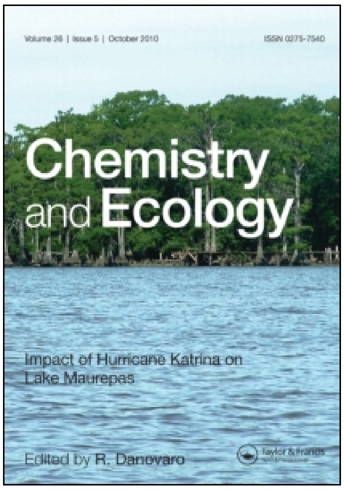

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# PARTITIONING OF Cs-137 BETWEEN SEDIMENT AND WATER FROM THE BLACK SEA

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As part of a joint USA/USSR Environmental Agreement to determine the distribution and concentration of Chernobyl radioactivity in the northwest Black Sea area, the sediment from eight stations was collected and analyzed to assess the ability of this material to adsorb radiocaesium. Batch tests were conducted in which Cs-137 tracer was added to mixtures of sediment and bottom water, with contact solutions ranging from 85 Bq ml$^{-1}$ to 1,760 Bq ml$^{-1}$. This work was done in an argon atmosphere at 9.5°C, which is the average temperature of the sediment. Isotherms were linear for all cores and distribution ratios ($R_D$) calculated from the slopes of the isotherms ranged from 660 to 1,660 ml g$^{-1}$. These isotherms fit a simplified Freundlich isotherm. Correlations of $R_D$ to a number of sediment parameters describing texture and mineralogy were determined. A close relationship was observed between $R_D$ for caesium and the percentage of illite contained in the samples.

KEY WORDS: Cs-137, Black Sea sediments, Chernobyl

## INTRODUCTION

The reactor accident at Chernobyl, which occurred on April 26, 1986, released approximately $10^{17}$ Bq of Cs-137 (Anspaugh *et al.*, 1988) as calculated from measurements of Cs-137 deposition throughout the world. It is important to understand the fate of this radionuclide because external doses incurred more than a few weeks after the incident are dominated by it.

Approximately half of the volatile elements (caesium, iodine) contained in the Chernobyl reactor core were released as compared to a small percentage of the more refractory elements (Goldman *et al.*, 1987). Releases of volatile elements from the reactor apparently took place in two physical forms. One was associated with particles of the reactor fuel and the other was initially contained in the vapour phase. The vapour phase then either condensed to form particles that were dominated by a single radionuclide as was observed for ruthenium and barium (Devell, 1988), or became associated with other airborne particles. There is evidence that most of the caesium was released in the volatilized form. The ratio of Cs-134/Cs-137 varied in the fuel from 0 to 1.2 due to differing residence times of various fuel rods in the reactor. However, in the environment this ratio was consistently 0.55 (± 0.09), implying that this element was mixed in the gas phase (Torvonen *et al.*, 1988) prior to deposition associated with settling of solid particles or rain.

The immediate result of the Chernobyl accident on radionuclide distributions in the Black Sea has been discussed by Buesseler *et al.*, (1987) in the context of fallout radionuclides observed in sediment traps. The specific activity of Cs-137 on trapped

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*This work was sponsored by the US Environmental Protection Agency

particles averaged 1.3 Bq $g^{-1}$ (Buesseler *et al.*, 1987, Livingston and Buesseler, 1986) and the mean activity in the surface water was 170 Bq $m^{-3}$ (Buesseler and Livingston, 1986).

During June of 1990, box core samples were obtained from stations in the northern part of the Black Sea, as part of a joint USA/USSR Environmental Agreement to determine the distribution and concentration of Chernobyl radionuclides in the Black Sea area. The positions of these stations as well as the water depth and sediment temperatures are given in Table 1. A chart of these stations is given by Neiheisel *et al.* (this volume). Bottom water samples were also obtained from all stations except 10 and 12. Frozen subcores were sent to Brookhaven National Laboratory for analysis of the sediments' ability to adsorb caesium.

**Table 1** Black Sea core stations and data – June 1990

| *Station #* | *Station Position*<br>*Latitude (N)* | *Longitude (E)* | *Water Depth (m)* | *Sediment Temperature* °C) |
|---|---|---|---|---|
| 4 | 45° 14.0′ | 29° 48.7′ | 27 | 7.2 |
| 5 | 44° 42.5′ | 31° 20.8′ | 105 | 8.4 |
| 6 | 44° 38.9′ | 31° 29.2′ | 544 | 9.5 |
| 7 | 44° 10.8′ | 30° 28.2′ | 114 | 9.2 |
| 8 | 43° 57.9′ | 30° 49.0′ | 555 | 9.5 |
| 10 | 43′ 58.9′ | 31° 22.8′ | 1288 | 9.9 |
| 11 | 44° 32.4′ | 31° 56.0′ | 1109 | 11.0 |
| 12 | 44° 54.1′ | 32° 13.3′ | 335 | 9.8 |

## BACKGROUND

Caesium in solution is a monovalent cation that seldom forms complexes (Onishi, *et al.*, 1981) under environmental conditions. Adsorption of caesium from fresh water is not significantly influenced by the presence of organics in sediment (Schell *et al.*, 1981). At low water to sediment ratios ($< 200$ mg $l^{-1}$), the partitioning of caesium between solid and liquid is controlled by colloids. At higher ratios, ion exchange dominates and follows a Freundlich isotherm (Aston and Duursma, 1973). Laboratory adsorption experiments show that illite has a strong affinity for adsorbing caesium which is due in part to structural properties of illite, specifically the 10° c-axis spacing and the effect of potassium on inducing and maintaining collapse of the c-axis (Tamura and Jacobs, 1960). These physical characteristics are more critical to caesium adsorption than are parameters such as cation exchange capacity and surface area. Field studies in freshwater systems also show that the mica-illite suite of minerals preferentially adsorbs caesium (Francis and Brinkley, 1976).

The adsorption of caesium on to calcium-illite involves three discrete types of exchange sites, each with its own degree of selectivity for caesium. The most selective sites account for 0.5% and 3% of the total exchange capacity of the mineral and are located at the frayed edges of the crystallites or in inter-lattice positions. The least selective type of site is associated with the planar surfaces of the mineral and accounts for 96.5% of the total exchange capacity (Brouwer *et al.*, 1983; Sawhney, 1972). At low concentrations (< 10 meq $g^{-1}$) most adsorption of caesium takes place on the highly selective sites and is reversible (Brouwer *et al.*, 1983). Adsorption and desorption are nevertheless related to the concentration of competing ions, particularly potassium, ammonium, magnesium and sodium, (e.g. Patel *et al.*, 1975; Schell *et al.*, 1981; Evans *et al.*, 1983). Adsorption is typically observed to be rapid as shown in Table 2. However, in a recent study, while most adsorption (~95%) took place rapidly, some continued over much longer times (Comans *et al.*, 1991). This was attributed to isotopic exchange with previously sorbed stable caesium. Others have observed slow, continuing adsorption/desorption of caesium-137 from contaminated freshwater sediment and ascribed this process to migration from mineral lattices (Evans *et al.*, 1983).

**Table 2** Times to equilibrium for adsorption of caesium-137

| *Time (hours)* | *Comment* | *Reference* |
|---|---|---|
| 24 | Rhine River | Schneider and Block, 1968 |
| $17 \pm 10 (t^{1/2})$ | Thin layer technique, sea water | Duursma and Eisma, 1973 |
| 2–24 | Nitelva River sediment, river water | Garder and Skulberg, 1964 |
| < 48 | Anoxic, oxic marine sediment | Seymour, *et al.* 1979 |
| $17 (t^{1/2})$ | Bombay sediment, sea water | Patel, *et al.*, 1975 |
| 50 to > 75 | Varied with sediment type | Schell, *et al.*, 1981 |
| > 384 | Slow adsorption continues | Comans, *et al.*, 1991 |

Distribution coefficients ($K_D$) from a variety of marine and freshwater sediments were reviewed by Onishi *et al.* (1981). Values in marine environments range from 17 to 9,000 ml $g^{-1}$ as shown in Table 3. Typically adsorption of caesium in sea water is reduced by a factor of about five compared to fresh water.

**Table 3** Distribution coefficients for caesium in marine environments

| *Distribution coefficients* | *Sediment* | *Reference* |
|---|---|---|
| 40 | Carbonate | Aston and Duursma, 1973 |
| 450–900 | Illitic | Aston and Duursma, 1973 |
| 2,000–9,000 | Marine Red Clay | Cheng and Hamaguchi, 1968 |
| 42–559 | Four types of sediment from the Pacific | Schebetkoviskii and Kuzketsov, 1971 |
| 17–640 | Near shore bays and estuaries | Schell, *et al.*, 1981 |
| 52 | Anoxic Marine | Seymour, 1977 |
| 17 | Oxic Marine | Seymour, 1977 |
| 18–180 | NW Atlantic 3,800 m Disposal Site | Fuhrmann and Colombo, 1983 |
| 10–1,000 | NE Atlantic | Kershaw *et al.*, 1986 |

## MATERIALS

The approach taken to determine the distribution of Cs-137 between the aqueous and solid phases of Black Sea sediment uses a batch adsorption method. All work was conducted in an argon atmosphere to eliminate effects of oxidation such as alteration of reduced minerals and changes in pH.

### *Radioactive Tracers*

To determine distribution ratios it is important to conduct experiments using several concentrations of the species of interest in the contact solution. Therefore, four tracers were made that contained varying concentrations of carrier-free caesium-137 with concentrations of caesium calculated to be $2.8 \times 10^{-17}$ moles per Bq. The tracers were all made with filtered sea water from the Atlantic Ocean. Table 4 shows the activity in each. Molarity of caesium in these solutions is very low but it is appropriate for the levels of contamination of concern.

**Table 4** Activity of Cs-137 used in tracers

| *Tracer ID* | *Activity (Bg ml$^{-1}$)* | *Cs Concentration (moles ml$^{-1}$)* |
|---|---|---|
| B1 | $3.70 \times 10^4$ | $1.04 \times 10^{-12}$ |
| B2 | $1.85 \times 10^4$ | $5.18 \times 10^{-13}$ |
| B3 | $6.29 \times 10^3$ | $1.76 \times 10^{-13}$ |
| B4 | $1.78 \times 10^3$ | $4.98 \times 10^{-14}$ |

### *Sediment Samples*

Sediment samples were taken by cutting through the frozen cores to obtain the top 2 cm of each core. These sections were immediately placed in plastic containers and transferred to a glove box containing an argon atmosphere. After the sediment thawed, its $E_h$ was taken with a platinum redox probe attached to an Orion Research model 399A meter. Samples of the sediment were taken for detemination of water content by drying at 105°C. The sediment was a black, anoxic mud containing high percentages (mean of 63%) of the clay size fraction. Exceptions to this were cores 5 and 7 which contained about 20% sand, mostly in the form of shells. The sediment is described in detail elsewhere (Neiheisel *et al.*, 1992 this volume).

### *Bottom Water*

The water used as the contact solution was taken from just above the bottom at each station and was filtered through Whatman number 41 filter paper and then through 0.45 micrometer Type HA Millipore filter membrane under a slight vacuum. Water was obtained for stations 4, 5, 6, 7, 8, and 11. Water from stations 11 and 8 was used for sediment from stations 10 and 12 respectively. Argon was bubbled through the bottom water before the water was added to the sediment to remove oxygen from the water.

# METHODS

## *Kinetics Experiment*

Before starting the adsorption experiments it was necessary to determine the amount of time that was needed for the contact solution to attain a constant concentration after the start of the experiment. This is a kinetics experiment and precedes the actual determination of distribution ratios. Approximately 1.5 g of wet sediment from the bottom of core 12 were weighed into polyethylene containers, followed by 22.7 ml of filtered sea water from the Atlantic Ocean and 0.5 ml of tracer. The tracer was made by adding 1 ml of solution containing $1.85 \times 10^6$ Bq of caesium-137 to 49 ml of filtered Atlantic sea water. Each container for the kinetics experiment received $1.85 \times 10^4$ Bq. Correcting for the water content of the sediment (49.3%), the ratio of solid to liquid was 0.032. The experiment was run at 19°C. At intervals ranging from 4 hours to 4 days, 1.5 ml of liquid were removed and filtered with a 0.45 micrometer syringe filter. One ml of the filtered liquid was counted for 4,000 seconds on a sodium iodide gamma-ray detector. Results were calculated as the ratio of sample counts to original counts in the tracer and plotted as a function of time.

## *Batch Method*

The batch method is a standard approach to determining the capacity of a soil or sediment to adsorb a species of interest. Variations are described by Booth (1988). One method is oriented to marine sediments. Another method, ASTM standard practice D4319 (ASTM, 1984), is more general for any soil-like material. The batch method consists of exposing a quantity of sediment to a liquid (the contact solution) that contains a known amount of a species of interest. After a period of time, during which adsorption of the species of interest should approach equilibrium, the liquid is separated from the solid and the concentration of the species of interest is measured. The quantity of the species of interest is compared to that in the original contact solution and normalized to the amount of solid present. The value obtained is a distribution ratio ($R_D$) and is an indicator of the adsorption capacity of the sediment.

Approximately 2 g of wet sediment were weighed into each plastic bottle followed by 19.5 ml of filtered Black Sea bottom water. Reference samples were also made which contained sea water and tracer but no sediment. After equilibrating for several hours, 1.0 ml of tracer was added to each bottle. This work was done in a glove box containing argon. The sample bottles were placed into two large plastic jars with gas fittings on them. They were removed from the glove box and were put in an environmental chamber. Tubing was connected to the jars and a constant flow of argon was provided. The environmental chamber was held at 9.5°C (the average temperature of the sediment when it was retrieved) for the 28 days of the experiment. Periodically, the samples were shaken to suspend the sediment. After 28 days the sample containers were removed from the environmental chamber in lots of 4. Liquid was withdrawn by plastic syringe and filtered, through disposable 0.45 $\mu$m syringe filters, into plastic test-tubes for radionuclide analysis. These aliquots were counted using an automatic sodium iodide gamma-ray detector for 1,000 or 4,000 seconds depending on their activity. The activity in the samples was compared to the activity in reference tracers prepared from dilutions of the original tracers. The formula for calculating distribution ratios ($R_D$) is

$$R_D = \frac{A_s \times V_l}{W_s \times A_l}$$

where $A_s$ is the activity of the sediment phase (found by subtracting $A_l$ from the activity of the reference tracer), $W_s$ is the weight of the sediment phase corrected for moisture, $A_l$ is the activity of the solution equilibrated with the sediment, and $V_l$ is the volume of the solution equilibrated with the sediment.

### *Conductance*

Conductance of the bottom water was measured according to ASTM method D1125, "Electrical Conductivity and Resistivity of Water." A Model 32 YSI conductance meter was used with a 1.0 cm cell. Standards were prepared from KCl salt. Temperature was maintained at 25°C. The sea water samples were diluted, 1 ml sea water in 49 ml distilled water, to save bottom water samples.

### *Organics*

The organic content of the sediment was determined by $H_2O_2$ digestion according to the method by Gross (1971). Dried samples, approximately 1 g in weight, were digested in 10% $H_2O_2$, with later additions of 30% $H_2O_2$.

## RESULTS

### *Kinetics Experiment*

The kinetics experiment results are shown in Figure 1. Most, if not all, adsorption of Cs-137 was complete after 10 days. This experiment was conducted at 19°C while the $R_D$ determinations were conducted at 9.5°C. Since adsorption is an activated process the rate of adsorption at 9.5°C should be equal to or slower than the rate at 19°C. Consequently the $R_D$ experiments were run for 28 days to assure that they went to completion.

### *Batch Method*

Distribution ratios for individual batch experiments range from 390 ml $g^{-1}$ in core 5 to 1,770 ml $g^{-1}$ in core 4. Six replicates of the 880 Bq experiment for core 4 were prepared. The mean of these $R_D$ values is 1,695 ml $g^{-1}$ with a standard deviation of 4.3%. The low scatter of the results provided enough confidence in these determinations that some experiments were conducted with single samples to preserve core material and bottom water.

The data from the batch experiments were used to calculate $R_D$ values through developing isotherms. An isotherm is a graph of concentration of the species of interest on the solid phase plotted against its concentration remaining in the liquid. When data are plotted for a number of concentrations of the species of interest, the slope of the plotted line is the $R_D$. Figures 2a-c are examples of isotherms for cores 4, 7 and 12, respectively. Table 5 gives the distribution ratios that were calculated as the slope of the best-fit line for each core. The table also gives the correlation

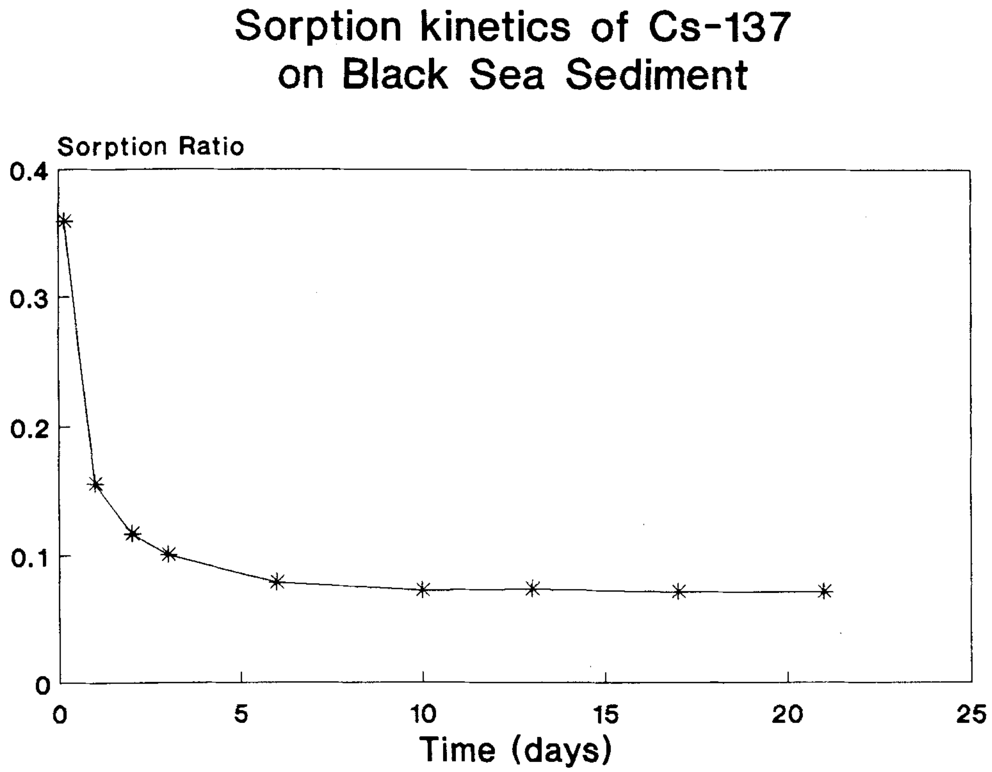

**Figure 1** Data from the kinetics experiment show that at 19°C essentially all adsorption of Cs-137 was complete within 10 days.

coefficient ($R^2$) which describes the goodness-of-fit of the line to the data points. In all cases the fit is excellent. These plots are useful because they show that the data all fall within the linear (and therefore predictable) range of the relationship of $R_D$ to activity of Cs-137 in the contact solution.

**Table 5** Adsorption of Cs-137 on Black Sea sediment distribution ratios based on isotherms

| *Core Number* | *Distribution Ratio $R_D$* | *Correlation Coefficient $R^2$* |
|---|---|---|
| 4 | 1,660 | 0.9965 |
| 5 | 661 | 0.9983 |
| 6 | 1,090 | 0.9976 |
| 7 | 880 | 0.9734 |
| 8 | 1,130 | 0.9891 |
| 10 | 1,280 | 0.9662 |
| 11 | 1,620 | 0.9662 |
| 12 | 1,370 | 0.9936 |
| All Cores | 1,190 | |

### *Freundlich Model*

A simple model of adsorption that can be applied to sediment interactions with adsorbing ions is the Freundlich model. The equation for this empirical model is

$$[A_{ads}] = K_{fr}\,[A_{soln}]^{1/p}$$

where the concentration of the adsorbed species, $A_{ads}$, is equal to the concentration of the species in solution, $A_{soln}$, adjusted with the constants $K_{fr}$ and 1/p. However, the plots shown in Figure 2 are linear which means the fitting parameter 1/p can be taken to be 1. With $1/p = 1$ the Freundlich equation becomes

$$[A_{ads}] = K_{fr}\,[A_{Soln}]$$

The value for $K_{fr}$ is equivalent to $R_D$ when the value of 1/p equals 1 (Serne *et al.*, 1990). The data from these experiments all fit the simplified Freundlich isotherm.

## DISCUSSION

The caesium distribution ratios determined for the Black Sea sediment are within

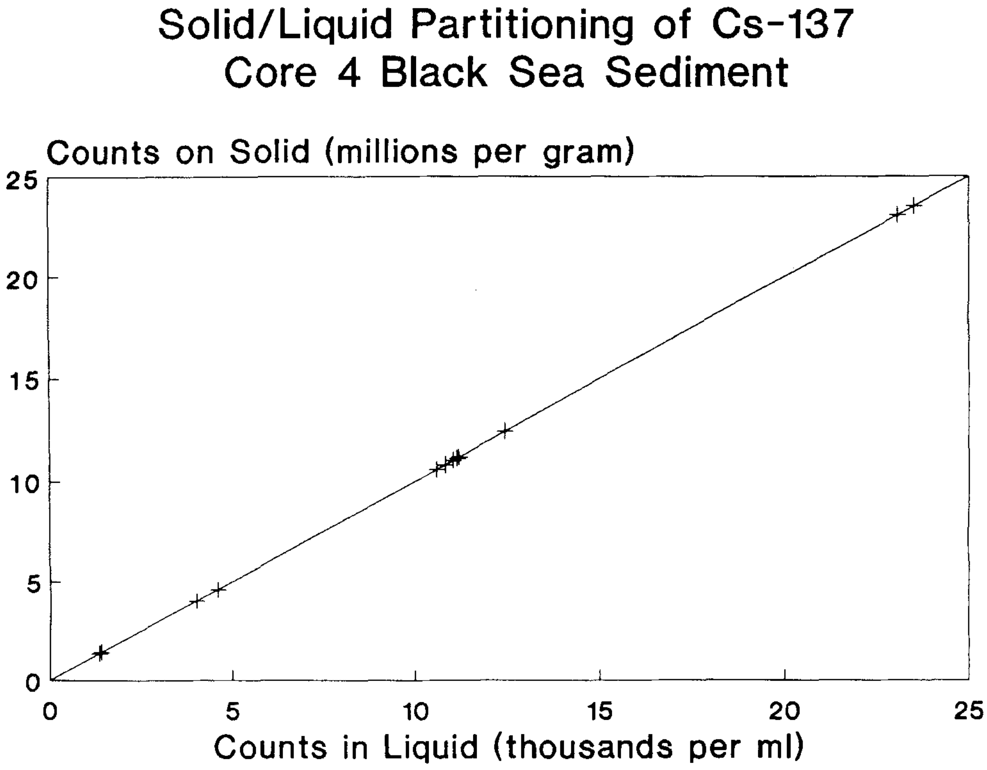

**Figure 2a** Isotherm for Cs-137 adsorption for sediment from core 4.

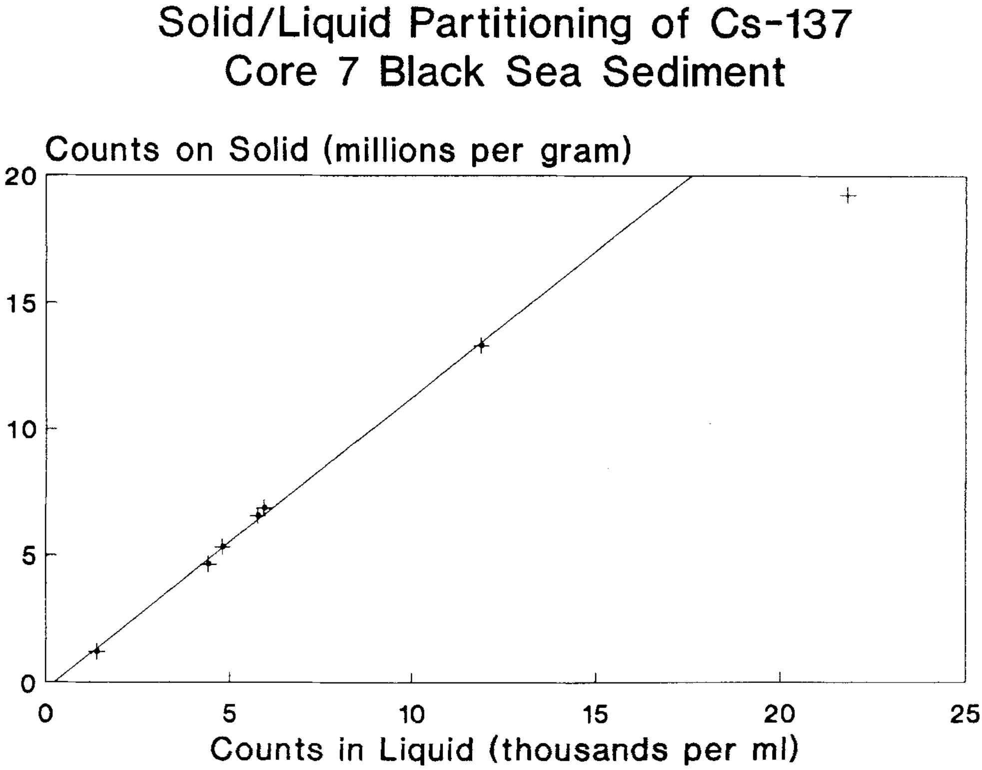

**Figure 2b** Isotherm for Cs-137 adsorption for sediment from core 7.

the very broad range that has been observed for other marine sediments (see Table 3). Most of the caesium was removed from solution very rapidly indicating that most adsorption took place on readily available sites. However, approximately 2% (based on Figure 1) of the caesium required a significantly longer amount of time to sorb, with equilibrium apparently being reached after 240 hours. This implies that a small fraction of the sorbing sites such as at interlattice sites are difficult to reach.

The concentration of caesium added to the sediment samples is very low, about $10^{-14}$ M $ml^{-1}$. This compares to the estimated concentration of caesium in the sea water of approximately $10^{-6}$ M $ml^{-1}$. The concentration of the tracer added to the batch experiments reflects the specific activity of the $^{137}Cs$ tracer (which was carrier-free). Even these values are high compared to those that could be expected as a result of radiological contamination. Nevertheless, the tracer concentration is infinitesimal compared to the background caesium in the sea water. Consequently one aspect of this investigation was to determine if the isotherm approach is still appropriate considering these conditions. From Figures 2a-c there is little question that there is a distinct and linear relationship between the quantity of $^{137}Cs$ tracer in the contact solution and the quantity of tracer adsorbed. Even under conditions of relatively high ambient caesium concentrations in sea water most of the caesium tracer was sorbed. This implies that isotopic exchange is the operative mechanism but direct proof of this is not available. That the adsorption isotherms are linear and fit a

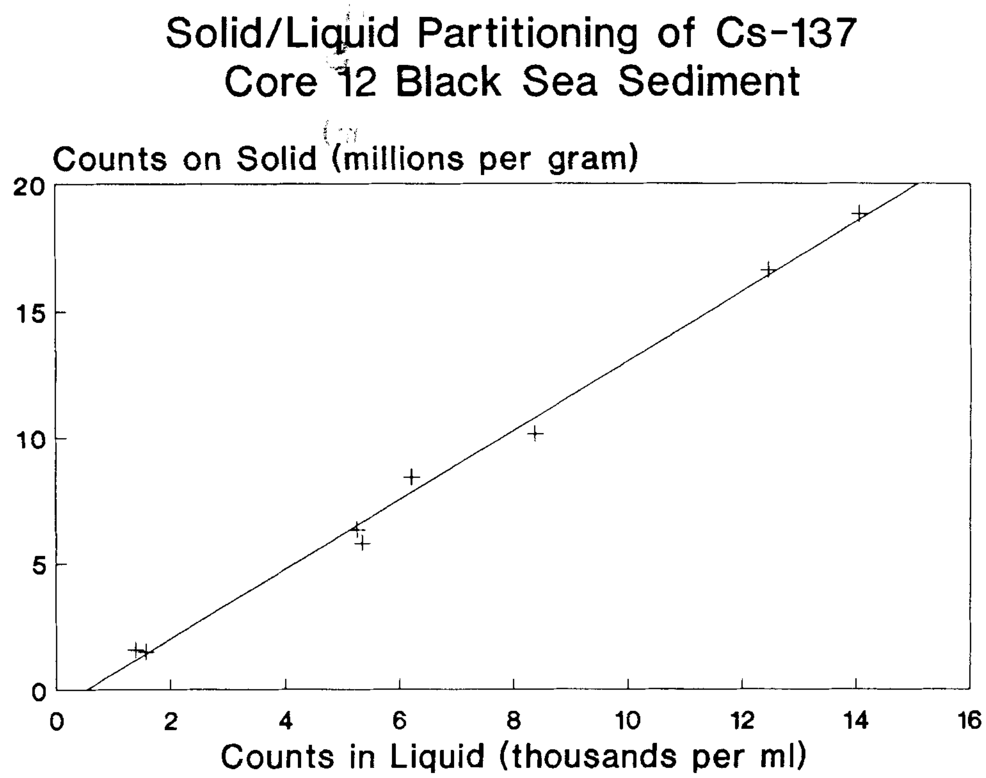

**Figure 2c** Isotherm for Cs-137 adsorption for sediment from core 12.

simplified Freundlich isotherm indicates that distribution ratios can be predicted for lower concentrations of tracers. Only at very high loadings of caesium (beyond the conceivable range for radiological contamination) would the isotherm be expected to fall off from the linear relationship.

The relationship between $R_D$ (the ability of the sediment to adsorb caesium) and other parameters that describe the sediment is of interest. Table 6 contains data for each core that have been compared to the $R_D$ determined for that core. Much of this material is drawn from Neiheisel *et al.* (this volume). Only minerals that comprise a major fraction of the sediment are considered. Correlation coefficients ($R^2$) were determined for these comparisons and are shown across the bottom of the table. All values of $R^2$ are low, indicating poor correlations for the full data sets for each core. However, inspection of the plots generated for these comparisons reveal closer relationships than indicated by the $R^2$ value. The plots of several parameters versus $R_D$ show that most of the samples fall on a trend having a correlation coefficient that indicates a linear relationship between the parameters plotted, while one or two samples are outliers. Figure 3 is an example. The correlation of quartz with $R_D$ is linear for six of the samples, with a correlation coefficient of 0.93. The slope is negative in this case; the more quartz present in a sample the lower the observed $R_D$. This is reasonable since quartz does not have a high capacity for adsorption. For most samples the quartz proportionately displaces components of

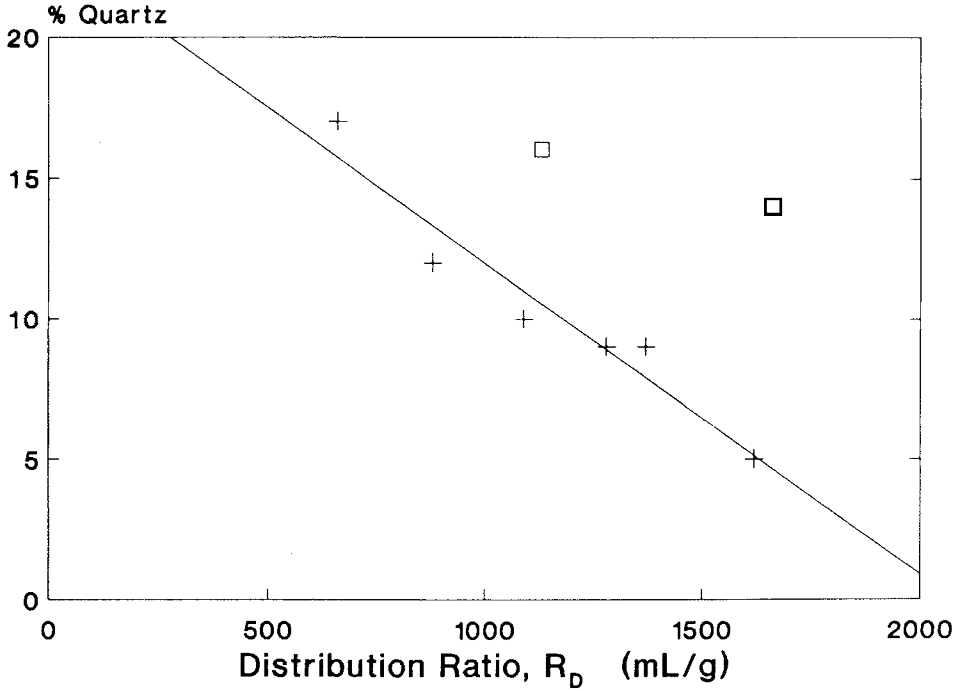

Figure 3 The correlation of quartz with the distribution coefficient of caesium is negative indicating that quartz is displacing an absorbing component of the soil.

the sediment that adsorb caesium. The two samples that are not on the trend, cores 4 and 8, have $R_D$ values that are higher than expected relative to their quartz content.

Illite, as discussed earlier, has been reported to have a high capacity to sorb caesium in fresh water systems. That capacity in marine systems is examined in Figure 4 where the percentage of illite is plotted as a function of $R_D$. The correlation coefficient for seven samples is 0.82. The one outlier is core 5 which has a much lower $R_D$ than would be expected for its illite content. This station has high percentages of sand (predominantly shell hash) and quartz which contribute to the low $R_D$ value. From Figure 5 the contribution to $R_D$ by components of the sediment, other than illite, is approximately 400 ml $g^{-1}$. For each 1% of illite in the sample, the $R_D$ for caesium increases, on the average, by about 50 ml $g^{-1}$.

Comparison of the organic fraction to $R_D$ shows a linear positive correlation that is poor. However, the two samples with the lowest $R_D$ values (cores 5 and 7) also have the lowest percentage of organics. The two samples with the highest $R_D$ values (cores 4 and 11) have high organic contents. This relationship does not always hold true since core 6 has a high organic content but a relatively low $R_D$.

Silt content has a low correlation to $R_D$ when the values are taken to have only one trend. However, the plot of percent silt versus $R_D$ can be interpreted as shown in Figure 5 where two populations of silt size particles contribute different adsorption capacities for caesium. One population having a high $R_D$ per percent of silt is shown as diamonds on the plot while the second population, with a low $R_D$ per percent of silt, is shown as squares. The correlation coefficient for the low $R_D$ population is 0.75. The two highest points are stations 4 and 6 which are characterized by $R_D$ to silt ratios

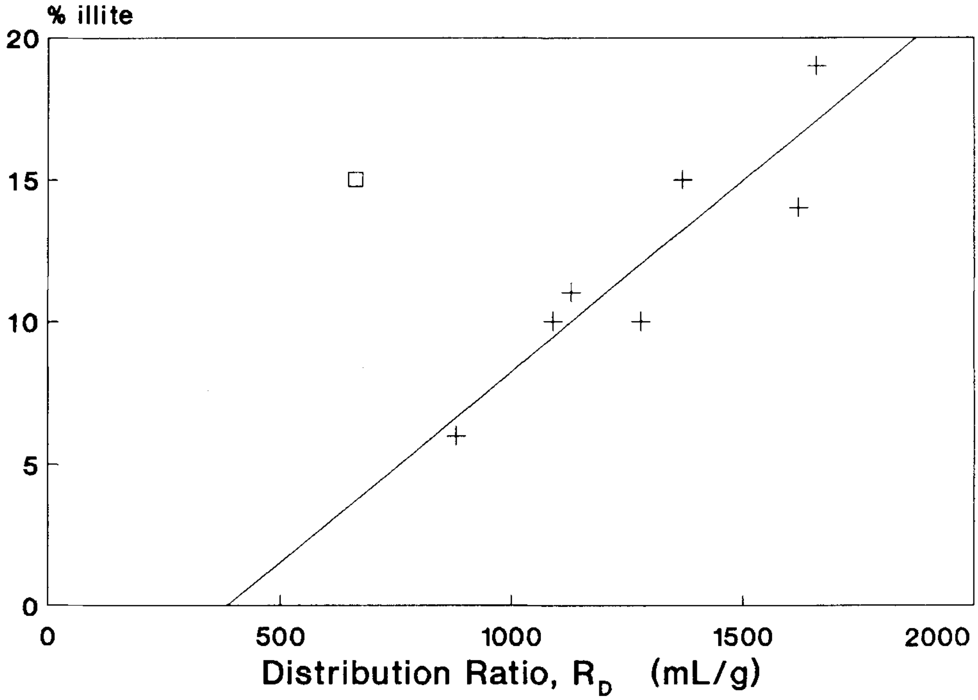

**Figure 4** The correlation of percent illite to the distribution ratio of caesium has an $R^2 = 0.82$ when the sample indicated as a square is dropped from the data.

equal to 28 and 24 respectively. Stations 11, 10 and 8 have ratios of 50, 51 and 49 respectively. There are few data points on which this interpretation can be based but it is strengthened by examining Figure 2 in Neiheisel *et al.* (1992) in which the dispersion of kaolinite from the Danube is shown to include stations 4, 5 and 6. Station 4 is located near the mouth of the Danube River and represents the population with high percentages of organics and silt. Station 6 also represents river deposits. The other population of silt size material is characteristic of samples taken at stations 8, 10 and 11. Material from station 5 and 7 represent mixtures of the two.

## CONCLUSIONS

Distribution ratios, derived from laboratory batch tests, were determined in sea water at four concentrations of carrier-free Cs-137 tracer. Simple Freundlich isotherms (that describe the concentrations remaining in the liquid after equilibration versus concentrations on the solid) were linear and gave $R_D$ values ranging from 661 to 1,660 ml $g^{-1}$. Correlation of $R_D$ with various major components of the sediment indicate a distinct relationship of $R_D$ with the percentage of illite contained in the sediment. The illite fraction contributed to caesium adsorption at the rate of about 50 ml $g^{-1}$ per 1% of illite. The sediment has a non-illitic distribution ratio of approximately 400 ml $g^{-1}$.

**Table 6** Correlation of environmental parameters to $R_D$ for Cs

| *Core #* | $R_D$ (ml g$^{-1}$) | *Depth* (m) | $E_h$ (mv) | pH* | *Sea water Conductivity* (m mhos) | *Sand* (%)* | *Silt* (%)* | *Clay* (%)* | *Calcite* (%)* | *Illite/ Mica* (%)* | *Feldspar* (%)* | *Quartz* (%)* | *Organic* (%)* |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 4 | 1660 | 27 | −280 | 7.63 | 0.504 | 3 | 60 | 37 | 31 | 19 | 18 | 14 | 9.4 |
| 5 | 661 | 105 | −240 | 6.91 | 0.459 | 23 | 20 | 57 | 38 | 15 | 11 | 17 | 3.1 |
| 6 | 1090 | 510 | −270 | 7.24 | 0.651 | 1 | 46 | 53 | 53 | 10 | 12 | 10 | 6.7 |
| 7 | 880 | 114 | −180 | 6.89 | 0.516 | 16 | 20 | 64 | 59 | 6 | 11 | 12 | 2.3 |
| 8 | 1130 | 564 | −290 | 6.98 | 0.462 | 0 | 23 | 77 | 50 | 11 | 9 | 16 | 4.8 |
| 10 | 1280 | 1715 | −250 | 6.96 | – | 1 | 25 | 74 | 52 | 10 | 14 | 9 | 4.8 |
| 11 | 1620 | 1109 | −300 | 6.84 | 0.413 | 3 | 32 | 65 | 55 | 14 | 12 | 5 | 5.2 |
| 12 | 1370 | 335 | −330 | 6.88 | – | 2 | 34 | 64 | 54 | 15 | 7 | 9 | 4.1 |
| Correlation Coefficient $R^2$ | – | 0.10 | 0.41 | 0.15 | 0.04 | 0.54 | 0.42 | 0.06 | 0.01 | 0.26 | 0.16 | 0.32 | 0.47 |

data from Neiheisel *et al.* (this volume).

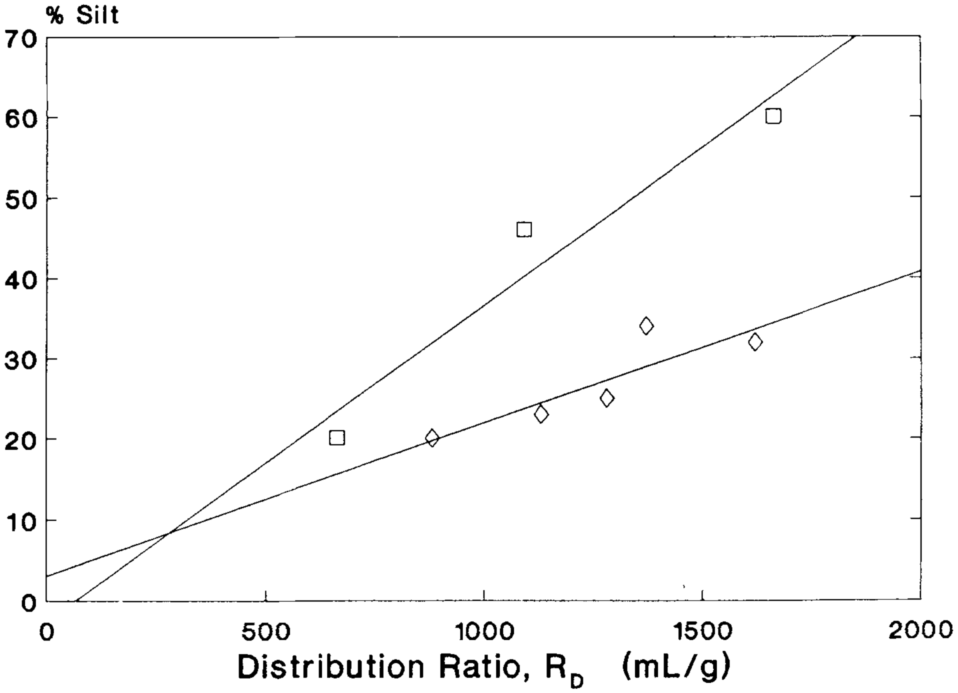

**Figure 5** The correlation of percent silt to the distribution of caesium may indicate two populations of silt.

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## *References*

American Society for Testing and Materials (ASTM) (1984), Distribution Ratios by the Short-Term Batch Method, D4319-83, January, 1984.

American Society for Testing and Materials (ASTM) (1982), Electrical Conductivity and Resistivity of Water, D1125-77, 1982.

Anspaugh, L., Catlin, R., and Goldman, M. (1988), The global impact of the Chernobyl Reactor accident, *Science*, **242,** 1513–1519.

Aston, S.R. and Duursma, E.K. (1973). Concentration effects on $^{137}$Cs, $^{65}$Zn, $^{60}$Co, and $^{106}$Ru sorption by marine sediments with geochemical implications. *Netherlands Journal of Sea Research* **6,** 225–240.

Booth, J. (editor) (1988), *A Methods Manual for Sediment Monitoring at Deep-Ocean Low-Level Radioactive Waste Disposal Sites,* EPA 520/1-88-02, US Environmental Protection Agency. 187 pp.

Brouwer, E., Baeyens, B., Maes, A., and Cremers, A. (1983), Cesium and rubidium ion equilibria in illite clay, *Journal of Physical Chemistry,* **57,** (7) 1213–1219.

Buesseler, K.O., Livingston, H.D., Honjo, S., Hay, B.J., Manganini, S.J., Degens, E., Ittekkot, V., Izdar, E., and Konuk, T. (1987) Chernobyl radionuclides in a Black Sea sediment trap, *Nature,* **329,** 825–828.

Cheng, H. and Hamaguchi, H. (1968), Studies on the adsorption of radioisotopes on marine sediments I. Role of exchangeable cations on the adsorption of caesium and exchange equilibria, *Health Physics,* **14,** 1353–1363.

Comans, R.N.J., Haller, M., and DePreter, P. (1991), Sorption of cesium on illite: non-equilibrium behavior and reversibility, *Geochimica et Cosmochimica Acta,* **55,** 433–440.

Devell, L., Nuclide composition of Chernobyl hot particles (1988), p 23–34, in: *Hot Particles from the Chernobyl Fallout,* Proceedings of an International Workshop held in Theuern, October 1987, H. von Philipsborn and F. Steinhäusler (editors), Schriftenreihe des Bergbau-und Industrie-museums, Ostbayern, Theuern, Germany. 147 p.

Duursma, E.K. and Eisma, D. (1973). Theoretical, experimental and field studies concerning reactions of radioisotopes with sediments and suspended particles of the sea, Part C: Applications to field studies, *Netherlands Journal of Sea Research,* **6,** 265–307.

Evans, D.W., Alberts, J., and Clark, R. (1983), Reversible ion-exchange fixation of cesium-137 leading to mobilization from reservoir sediments, *Geochimica et Cosmochimica Acta,* **47,** 1041–1049.

Francis, C.W., and Brinkley, F.S. (1976), Preferential adsorption of $^{137}$Cs to micaceous minerals in contaminated freshwater sediment, *Nature,* **260,** 511–513.

Fuhrmann, M. and Colombo, P. (1983). Sedimentology and geochemistry of cores from the 2800 meter Atlantic Radioactive Waste Disposal Site, BNL-37838, Brookhaven National Laboratory, unpublished report prepared for US Environmental Protection Agency, Washington, DC, USA. 45 pp.

Garder, K., and Skulberg, O. (1964), Sorption phenomena of radionuclides to clay particles in river water, *International Journal of Air and Water Pollution,* **8,** 229–241.

Goldman, M., *et al.* (1987), Health and Environmental Consequences of the Chernobyl Nuclear Power Plant Accident. DOE/ER-0332, Dept. of Energy, Washington, DC.

Gross, M.G., (1971) Carbon determination, pp. 573–596, in: Carver, R. (editor), *Procedures in Sedimentary Petrology,* Wiley Interscience, NY. 653 pp.

Kershaw, P.J., R. Van DerLeoff, M.M., Whitehead, N.E. (1986), Sediment sorption properties, pp. 89–96, in: *Interim Oceanographic Description of the North-East Atlantic Site for the Disposal of Low-level Radioactive Waste,* **2,** pp. 89–96, Dickson, R.R., Gurbutt, P.A., and Kershaw, P.J. (editors), Nuclear Energy Agency, Paris, France. 191 pp.

Livingston, H.D. and Buesseler, K.O., (1986), New Radiotracers in the Black Sea from Chernobyl Fallout, EOS; Transactions of the American Geophysical Union, 67, 1070 (Abstract only).

Neiheisel, J., McDaniel, W., and Panteleyev, G. (1992) Sediment parameters of northwest Black Sea Shelf Slope: Implications for transport of heavy metals and radionuclides, Chemistry and Ecology (this volume.)

Onishi, Y., Serne, R.J., Arnold, E.M., Cowan, C.E. and Thompson, F.L. (1981), Critical Review: Radionuclide Transport, Sediment Transport, and Water Quality Mathematical Modeling; and Radionuclide Adsorption/Desorption Mechanisms, US Nuclear Regulatory Commission Report NUREG/CR-1322, Washington, D.C. 490 pp.

Patel, B., Mulay, C.D. and Ganguly, A.K. (1975). Radioecology of Bombay Harbour – A tidal estuary. *Estuarine and Coastal Marine Science* **3,** 13–42.

Sawhney, B.L. (1972), Selective sorption and fixation of cations by clay minerals: A review, *Clays and Clay Minerals,* **20,** 93–100.

Schebetkoviskii, V.N. and Kuzketsov, Yu.V. (1971). Behavior of Cs-137 and Ce-144 in the sorption system sea water – sediment, *Radiokhimiya,* **13,** 911–913. (Quoted in Onishi *et al.* 1981).

Schell, W.R., Sanchez, A., and Sibley, T.H. (1981), Distribution Coefficients for Radionuclides in Aquatic Environments, Adsorption and Desorption Studies of $^{137}$Cs, US Nuclear Regulatory Commission Report, NUREG/CR-1852, 2, Washington, DC. 49 pp.

Schneider, H. and Block, W. (1968). On the question of the capacity of the Rhine for radioactive nuclides. Sorption of radionuclides by sediments of the Rhine. *Gas-Wasserfach.* **109,** 1410–1415.

Serne, R.J., Arthur, R.C., and Krupka, K.M. (1990), Review of Geochemical Processes and Codes for Assessment of Radionuclide Migration Potential at Commercial LLW Sites, NUREG/CR-5548, PNL-7285, US Nuclear Regulatory Commission, Washington, DC, 20555. 115 pp.

Seymour, A.H. (1977). 1976–1977 Annual Progress Report, Distribution coefficient for transuranic elements in aquatic environments. Laboratory of Radiation Ecology, College of Marine Fisheries, University of Washington. (Quoted in Onishi *et al.,* 1981).

Seymour, A.H., Neuissi, A., Schell, W.R. and Sanchez, A. (1979). Distribution Coefficients for Radionuclides in Aquatic Environments. I. Development of Methods and Results from Plutonium and Americium in Fresh and Marine Water-Sediment Systems, NUREG/CR-0801, US Nuclear Regulatory Commission, Washington, DC. 70 pp.

Tamura, T., and Jacobs, D.G. (1960), Structural implications in cesium sorption, *Health Physics,* **2,** 391–398.

Toivonen, H., Servomaa, K., and Rytömaa, T., (1988). Aerosols from Chernobyl: Particle characteristics and health implications pp. 97–105, In: *Hot Particles from the Chernobyl Fallout,* Proceedings of an International Workshop held in Theuern, October 1987, H. Von Philipsborn and F. Steinhäusler (editors), Schriftenreihe des Bergbau-und Industriemuseums, Ostbayern, Theuern, Germany. 147 pp.