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PARTITIONING BETWEEN SEDIMENT AND POREWATER OF RADIOCESIUM
FROM CHERNOBYL FALLOUT

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ABSTRACT

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 sediment from the northwest Black Sea to adsorb radiocesium. Two methods were used to determine partitioning between liquid and solid phases; batch tests and porewater separation. In the batch test, Cesium Cs-137 tracer was added to mixtures of sediment and bottom water, with contact solutions ranging from 85 Bq/ml to 1,760 Bq/ml. This work was done in an argon atmosphere, at 9.5°C, which is the average temperature of the sediment. The distribution ratios (R_D) for individual batch tests ranged from 390 to 1,770 ml/g. Isotherms were linear for all cores and R_D values calculated from the slopes of the isotherms ranged from 660 to 1,660 ml/g. A second approach was used to determine the partitioning of Cs-137 between the porewater and the sediment under more natural conditions. The top 2 cm of two cores were sectioned and the sediment and the porewater were separated. In both samples the sediment contained significant amounts of Cs-137 (0.361 and 0.072 Bq/g, respectively) and Cs-134 (0.045 and 0.010 Bq/g, respectively) indicating the presence of fallout from the Chernobyl reactor accident. There was no cesium activity observed in the porewater at a minimum detectable level of 0.008 Bq/ml.

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 accident are dominated by it.

Approximately half of the volatile elements (cesium, 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 form was associated with particles of the reactor fuel and the other was initially contained in the vapor phase. The vapor 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 cesium 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 particles averaged 1.3 Bq/g (Buesseler et al., 1987, Buesseler and Livingston, 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 I. 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 Cesium.

| | Station Position | | Water | Sediment |
Station #	Latitude (N)	Longitude (E)	Depth (m)	Temperature (°C)
TABLE I BLACK SEA CORE STATIONS AND DATA - JUNE 1990				
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

Cesium in solution is a monovalent cation that seldom forms complexes (Onishi, et al. 1981) under environmental conditions. Adsorption of cesium from freshwater is not significantly influenced by the presence of organics in sediment (Schell et al., 1981). At low water to sediment

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ratios (< 200 mg/l), the partitioning of cesium between solid and liquid is controlled by colloids. At higher ratios, ion exchange dominates and follows a Freundlich isotherm (Aston & Duursma, 1973). Laboratory adsorption experiments show that illite has a strong affinity for adsorbing cesium which is due in part to structural properties of illite, specifically the 10 Å c-axis spacing and the effect of potassium on inducing and maintaining collapse of the c-axis (Tamura & Jacobs, 1960). These physical characteristics are more critical to cesium 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 adsorb cesium (Francis & Brinkley, 1976).

The adsorption of cesium onto calcium-illite involves three discrete types of exchange sites, each with its own degree of selectivity for cesium. 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 interlattice 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, 1971). At low concentrations (< 1 meq/100 g) most adsorption of cesium 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, 1981; Evans et al., 1983). Adsorption is typically observed to be rapid as shown in Table II. However, in a recent study, while most

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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 cesium. Others have observed slow, continuing adsorption/desorption of cesium-137 from contaminated freshwater sediment and ascribed this process to migration from mineral lattices (Evans et al., 1983).

TABLE II
TIMES TO EQUILIBRIUM FOR ADSORPTION OF CESIUM-137

Time (hours)	Comment	Reference
24	Rhine River	Schneider & Block, 1968
$17 \pm 10 \ (t^{1/2})$	Thin layer technique, seawater	Duursma & Eisma, 1973
2 - 24	Nitelva River sediment, river water	Garder & 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 (1981). Values in marine environments range from 17 to 9,000 ml/g as shown in Table III. Typically adsorption of cesium in seawater is reduced by a factor of about five compared to freshwater.

| TABLE III |||
| DISTRIBUTION COEFFICIENTS FOR CESIUM IN MARINE ENVIRONMENTS |||
Distribution Ratio (ml/g)	Sediment Type	Reference
40	Carbonate	Aston & Duursma, 1973
450 - 900	Illitic	Aston & Duursma, 1973
2,000 - 9,000	Marine Red Clay	Cheng & Hamaguchi, 1968
42 - 559	Four types of sediment from the Pacific	Schebetkoviskii & Kuzketsov, 1971
17 - 640	Near shore bays and estuaries	Schell, 1981
52	Anoxic Marine	Seymour, 1977
17	Oxic Marine	Seymour, 1977
18 - 180	NW Atlantic 3,800 m Disposal Site	Fuhrmann & Colombo, 1982
10 - 1,000	NE Atlantic	Kershaw et al., 1986

MATERIALS

Two approaches were taken to determine the distribution of Cs-137 between the aqueous and solid phases of Black Sea sediment. One technique is a batch adsorption method and the other method is an experimental attempt to observe the distribution of Cs-137 between the porewater and the sediment in the top 2 cm of several cores. In both cases all work was conducted in an argon atmosphere to eliminate effects of oxidation.

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 cesium-137 with concentrations of cesium calculated to be 2.8×10^{-17} moles per Bq. The tracers were all made with filtered seawater from the Atlantic Ocean. Table IV shows the activity in each.

TABLE IV ACTIVITY OF Cs-137 USED IN TRACERS		
Tracer ID	Activity per ml Becquerels	Cs Concentration (moles)
B1	3.70×10^4	1.04×10^{-12}
B2	1.85×10^4	5.18×10^{-13}
B3	6.29×10^3	1.76×10^{-13}
B4	1.78×10^3	4.98×10^{-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 determination of water content by drying at 105°C. The sediment was a black, anoxic mud containing high percentages 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., 1991).

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.

METHODS

Kinetics Experiment - Before starting the actual 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 seawater from the Atlantic Ocean and 0.5 ml of tracer. The tracer was made by adding 1 ml of solution containing 1.85×10^6 Bq of cesium-137 to 49 ml of filtered Atlantic seawater. Each container for the kinetics experiment received 1.85×10^4 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 fil-

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tered 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 plotted 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 generally 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. Blanks were also made which contained 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

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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 μm syringe filters, into plastic test-tubes for radio-nuclide 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

$$R_D = \frac{A_s \times V_1}{W_s \times A_1}$$

where A_s is the activity of the sediment phase (found by subtracting A_1 from the activity of the blank), W_s is the weight of the sediment phase corrected for moisture, A_1 is the activity of the solution equilibrated with the sediment, and V_1 is the volume of the solution equilibrated with the sediment. .

Pore Water Methods - Three sections of cores were subjected to another approach for determining the partitioning of radionuclides between the sediment and the pore water. Selected sections were cut from cores while

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still frozen and placed in a glove box containing an argon atmosphere. After they thawed, they were placed into stainless steel pore water squeezers. The pore water was extracted through a Whatman number 41 filter into a polyethylene bottle. This extract was then filtered through a 0.45 μm pore size syringe filter. The sediment was removed from the squeezer, dried and weighed into plastic petri dishes. The sediment and the pore water were analyzed using a planar intrinsic germanium gamma-ray detector with counting times ranging from 1,000 minutes to 4,000 minutes depending on activity.

Because no cesium activity was observed in the porewater with the planar gamma-ray detector, samples were also counted on a high efficiency well-typed intrinsic germanium gamma-ray detector which was equipped with anti-coincidence circuits. This system is described by Cumming and Alburger (1985). Four ml of the liquid samples were placed in glass tubes and evaporated to dryness. Counting times were 1,000 minutes.

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 actual 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 about half that at 19°C. Consequently the R_D experiments were run for 28 days.

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Batch Method - Results of the batch method are presented in Table V. The
distribution ratios are shown for each core and for the four activities
of Cs-137 that were used as tracers. Distribution ratios range from 390
ml/g in core 5 to 1,770 ml/g in core 4. Six replicates of the 880 Bq
experiment for core 4 were prepared. Results are also shown on Table V.
The mean of these R_D values is 1,695 ml/g 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. These values are
typical for a marine sediment. Parameters such as grain size, mineralogy
and organic carbon content will effect R_D values. These parameters were
investigated for sediment samples taken at the same stations in the Black
Sea (Neiheisel et al., 1991).

The data from the batch experiments were used to calculate R_D
values in a second way, 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 is
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 VI gives the
distribution ratios that were calculated as the slope of the best-fit
line. The table also gives the correlation 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)

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range of the relationship of R_D to activity of Cs-137 in the contact solution. Distribution ratios for cores 5 and 7 are anomalously low. The surficial sediment from these cores contain large quantities of shell fragments which effectively reduce its overall sorption capacity.

	TABLE V ADSORPTION OF Cs-137 ON BLACK SEA SEDIMENT DISTRIBUTION RATIOS FOR INDIVIDUAL BATCH DETERMINATIONS				
	Initial Activity of the Cs-137 Tracer (Bq)				
	1,760	880	300	85	
Core Number	Distribution Ratios (ml/g)				Average
4	1,650 1,630	1,590 1,730 1,680 1,770 1,770 1,630	1,490 1,670	1,300 1,450	1,613
5	580	630	600	390	550
6	940	1,050	960	780	933
7	1,160 1,140	1,120 880	1,050 1,110	880	1,049
8	1,240 1,120	1,180 1,070	1,100	970	1,113
10	970	1,280	1,020	870	1,035
11	1,820	1,540	1,480	1,120	1,490
12	1,210 1,360	1,340 1,330	1,210 1,080	1,160 970	1,208

TABLE VI
ADSORPTION OF Cs-137 ON BLACK SEA SEDIMENT
DISTRIBUTION RATIOS BASED ON ISOTHERMS

Core Number	Distribution Ratio R_D (ml/g)	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.

Langmuir Model - The Langmuir model was also used to analyze the adsorption data. This model is also empirical but it has been discussed in terms of the thermodynamics of adsorption (Burchill et al., 1981). For such a discussion it is assumed that certain conditions are met, namely:

- the surface phase is homogeneous
- the solutions are very dilute

An advantage to this model is that it includes the concept of a finite surface capacity. The basic Langmuir equation for adsorption from solution is:

$$A_{ads} - \frac{n_s C}{K_L + C}$$

The equation used to analyze experimental data is shown below and is taken from Rubin and Mercer (1981).

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$$A_{ads} = -K_L \left(\frac{A_{ads}}{C} \right) + n_s$$

where C is the concentration of the species of interest in solution, n_s is the adsorption capacity of the surface phase, and K_L is the Langmuir isotherm constant. From this a plot of C/n_s versus C can be used to estimate K_L and n_s.

Figure 3 is a plot of a Langmuir isotherm for all samples. The average K_L is estimated to be 1,030 ml/g. The slope is essentially zero implying that, in the range of the data, the adsorption capacity is very large and independent of concentration. The four highest points on the plot are all from core 5. This anomalous data indicates low adsorption ratios (R_d) which are the result of a large quantity of shells in the sediment at this station. The ratio of Cs-137 adsorbed on the solid to Cs-137 in solution (at the end of the experiment) remains constant at approximately 0.001 which means that 99.9% of the Cs-137, that was originally in solution, was adsorbed. That almost all of the Cs-137 was adsorbed is not surprising considering that the concentration of cesium added to each container varied between approximately 5 x 10^{-14} and 2.5 x 10^{-15} moles per ml.

R_D *Determinations in Pore Water* - Three sections of cores were analyzed to determine the distribution of Cs-137 between the pore water and the sediment. Two of the samples were surficial sediment from shallow

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stations, and the third was from the bottom of a core taken in deep water. This was done to facilitate a search for radionuclides other than Cs-137 and Cs-134 by providing a natural spectrum which could be subtracted from those in which activity from Chernobyl was suspected. The activities of Cs-137 and Cs-134 for the solid and liquid phases are shown in Table VII.

In all cases there was no observable Cs-137 in the pore water while the solid phase activity in the surficial samples was substantial. For the sample with the highest activity (core 4), which had R_D values averaging 1,600 ml/g, the activity in the pore water should be 0.0003 Bq/g. This is lower than the detection limits for our current geometry by more than a factor of 30. No anthropogenic radionuclides, other than Cs-137 and Cs-134, were observed in the sediment. A possible exception to this is slightly elevated U-235 activity in core 4 as compared to the other sediment samples. This may indicate some reactor core material from Chernobyl that was deposited as particulate fallout.

		TABLE VII ADSORPTION OF Cs-137 ON BLACK SEA SEDIMENT Cs-137 AND Cs-134 ACTIVITIES IN SEDIMENT AND PORE WATER			
Core #	Section cm	Activity in Sediment (Bq/g) Cs-137	Cs-134	Activity in Pore Water Cs-137	Cs-134
4	0 - 2	0.361	0.045	BDL*	BDL*
5	0 - 2	0.072	0.010	BDL*	BDL*
12	(bottom)	BDL*	BDL*	BDL*	BDL*

*Below Detection Limit

Minimum detectable activity, based on a 100 count peak in a 10 g sample, is 0.0095 Bq/g.

CONCLUSIONS

The presence of radiocesium was observed in the surficial sediment of the two cores tested. That this material was derived from Chernobyl was based on the presence of Cs-134. Even though significant quantities of Cs-137 were observed on the solid phase, attempts to find it in the separated porewater from that sediment were unsuccessful due to limited quantities of porewater.

Distribution ratios, derived from laboratory batch tests, were determined at four concentrations of carrier-free Cs-137 tracer. Simple Freundlich isotherms of concentration in the liquid versus concentrations of the solid were linear and gave R_D values ranging from 661 to 1,660 ml/g. The two lowest values were associated with sediment having significantly more material in the sand size fraction (~20%) than did other samples. Analysis with the Langmuir model gives an overall K_L value of 1,030 ml/g which agrees very well with the average R_D from the Freundlich isotherms of 1,190 ml/g. The Largmuir model results imply that for this data the amount of Cs-137 adsorbed is small compared to the total capacity of the sediment.

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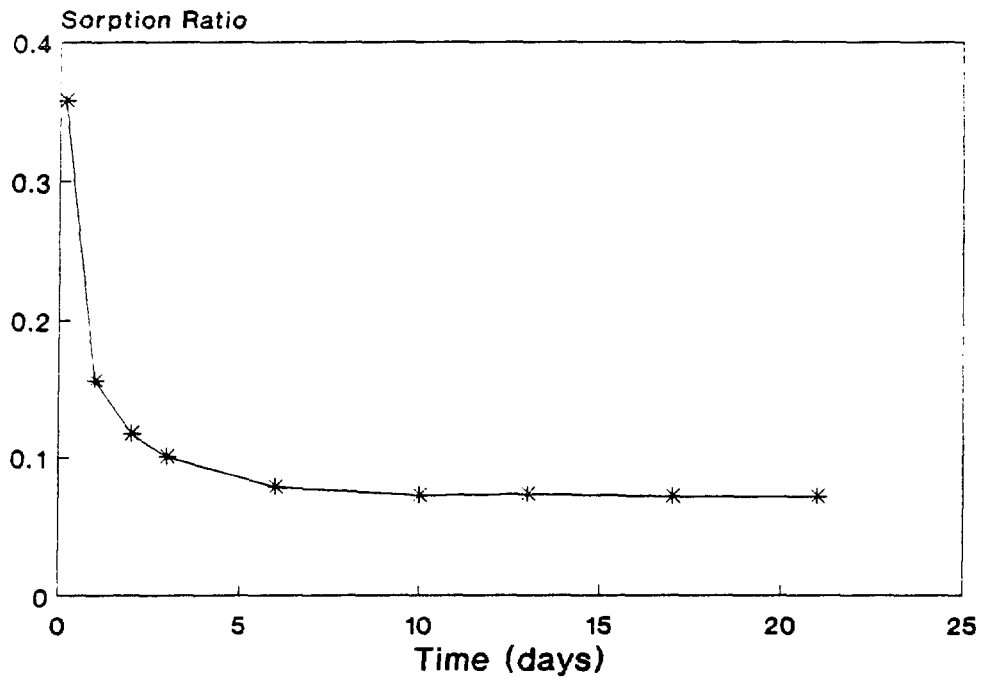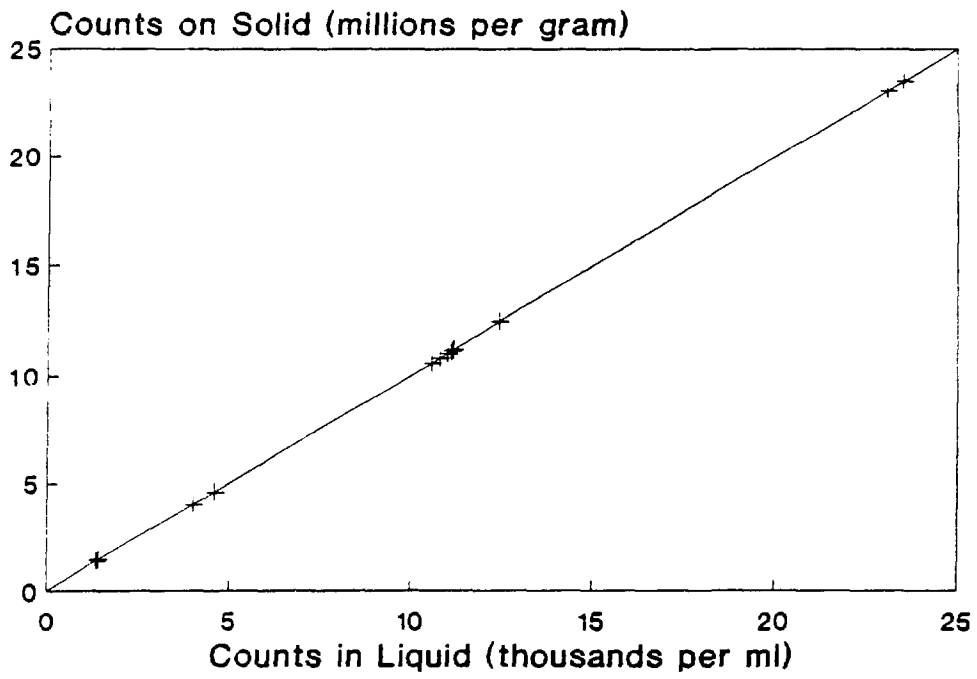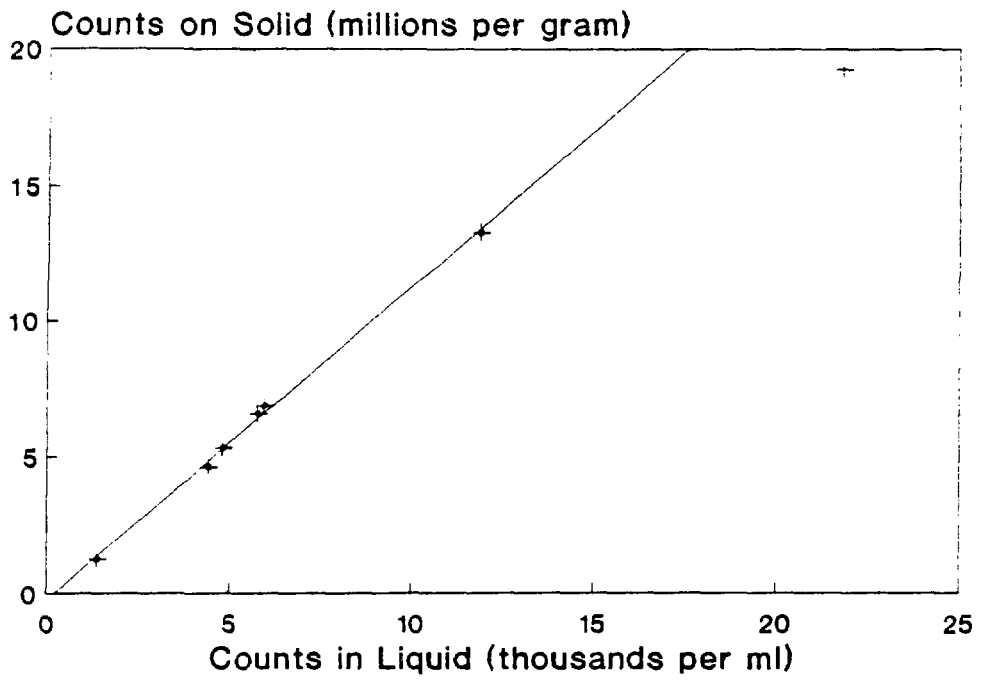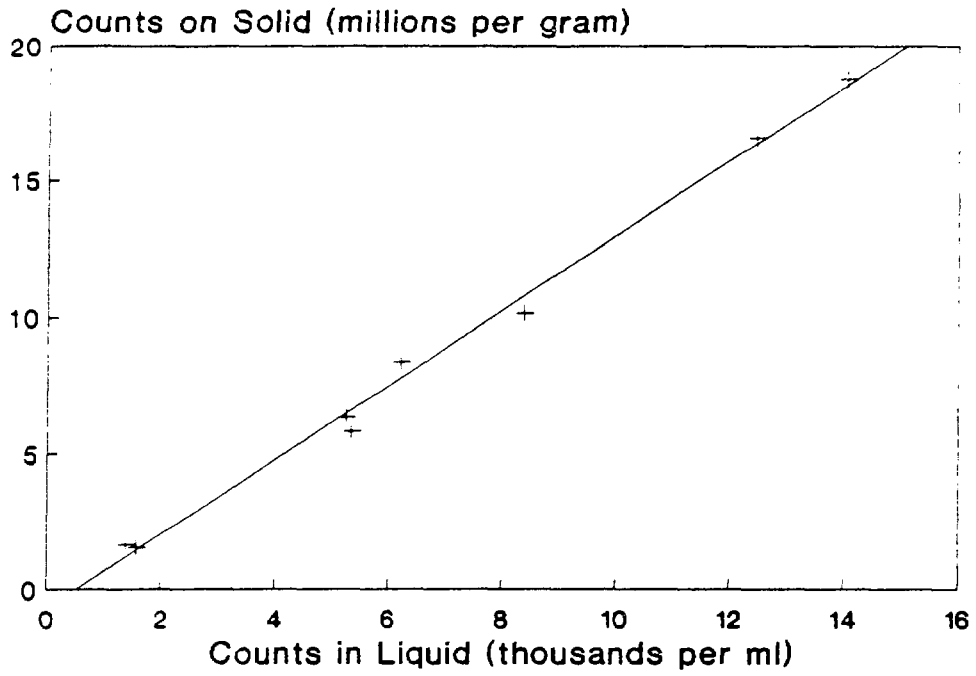
Figure 1

Figure 2A

Figure 2B

Counts on Solid (millions per gram) vs. Counts in Liquid (thousands per ml)

Figure 2C

Figure 3