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QUARTERLY REPORT

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DISTRIBUTION COEFFICIENTS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS

Prepared For The

Division of Safeguards,
Fuel Cycle and Environmental Research
U. S. Nuclear Regulatory Commission
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DISTRIBUTION COEFFICIENTS FOR RADIONUCLIDES IN AQUATIC ENVIRONMENTS

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1 December 1979 - 29 February 1980

Introduction This project was initiated in August 1976 to obtain new and better information for predicting the fate of transuranic elements, particularly americium and plutonium, in aquatic environments. The program has since been expanded to include the isotopes ^{85}Sr, ^{106}Ru, ^{137}Cs and ^{244}Cm. Our general approach has been to determine distribution coefficients (K_ds) for these radionuclides in constant shaking experiments using <63µm sediments and filtered water from a variety of aquatic environments. These experiments showed that the K_d values for the above radionuclides may vary significantly among sediment-water systems from different environments. A major research objective of the present contract is to determine how changes in selected environmental parameters can affect the distribution coefficients. During the past quarter we have used sediment-water systems from four locations in the Hudson River Estuary to study the effect of salinity and increased concentrations of selected organic ligands on the distribution coefficients of ^{60}Co, ^{106}Ru, ^{137}Cs and ^{241}Am. We have also evaluated a liquid scintillation counting technique to measure ^{244}Cm in both the dissolved and particulate phases of experimental systems.

This is the second quarterly report for the fourth year of this project.

Hudson River Estuary Sediment-Water Systems Distribution coefficients were determined for ^{57}Co, ^{106}Ru, ^{137}Cs and ^{241}Am for four different sediment-water systems from the Hudson River Estuary. Water and <63μm sediment samples from mp. 0.1 (S=21°/₀₀), mp. 18.6 (S=18°/₀₀), mp. .. (S=3.3°/₀₀) and mp. 60.0 (S=0.3°/₀₀) were used for constant shaking experiments at 4°C and a sediment concentration of 200 mg/l. Sodium azide (NaN_3) was added as a biological poison at a concentration of 0.1% weight/volume NaN_3. The average distribution coefficient obtained at the four stations for each of the radionuclides is shown in Table 1.

After the spiked sediment-water systems were allowed to equilibrate for approximately 125 hours individual organic ligands were added to experimental containers from mp. 0.1, 18.6 and 43.3. The organic ligands, ethylene diamine tetraacetic acid; 1-nitroso - 2-naphthol; 1,10 phenanthroline; humic acid or glycolic acid, were added at concentrations of ~1 x 10^{-5}M. This is equivalent to 0.25 - 1.5 x 10^{-6}g C/ml (0.25 - 1.5 ppm). Samples were collected at four different times, 21 - 165 hours, after the addition of ligands for determination of distribution coefficients. The results of these calculations are presented in Tables 2 - 5.

These data will be interpreted in the progress report for the third quarter.

Measurement of ^{244}Cm Preliminary observations have been made to evaluate a liquid scintillation technique for measuring ^{244}Cm. The proposed experimental method consists of introducing ^{244}Cm as the only radionuclide in

a constant shaking sediment-water system. Five ml sub-samples are filtered through 0.45μm Millipore filters. The filtrate is acidified with 1/10 volume concentrated H_2SO_4 and then diluted tenfold with distilled H_2SO_4. Four mls of the solubilized filter samples and 4 mls of filtrate are counted in 10 ml of scintillation cocktail (3a70B, Research Products International). Samples prepared in the above manner and spiked with ^{244}Cm immediately prior to counting have been evaluated for counting efficienty. For acidified samples the counting efficienty on a Packard Tri-Carb Liquid Scintillation Spectrometer is evuivalent for both dissolved and particulate samples. In addition, the counting efficiency is nearly 100% and both sample types can be counted in the same channels.

During the next quarter this technique will be used to determine K_d values of ^{244}Cm in sediment-water systems from the Hudson River Estuary.

Table 1. Average K_d values for ^{57}Co, ^{106}Ru, ^{137}Cs and ^{241}Am in sediment-water systems from four stations in the Hudson River Estuary.

Radionuclides	^{57}Co		^{106}Ru		^{137}Cs		^{241}Am	
Station	n*	Average K_d** ml/g x 10^{-2}	n	Average K_d ml/g x 10^{-3}	n	Average K_d ml/g x 10^{-2}	n	Average K_d ml/g x 10^{-5}
mp. 0.1	7	3.63 ± 0.63	6	4.94 ± 0.22	6	6.35 ± 0.63	7	>2.3
mp. 18.6	7	2.92 ± 0.62	7	3.23 ± 0.43	7	5.82 ± 0.70	7	2.55 ± 0.38
mp. 43.3	7	9.04 ± 1.03	6	3.22 ± 0.40	6	1.74 ± 0.08	7	>2.1
mp. 59.8	2	52.1 ± 2.5	4	2.55 ± 0.16	5	1.66 ± 0.13	4	1.35 ± 0.29

* n = The total number of K_d values determined for a radionuclide in a given sediment-water system "at equilibrium".

** Average K_d is the mean for all determinations. Error terms are one standard deviation from the mean for replicate determinations.

Table 2. Distribution Coefficients for ^{57}Co ($K_d \times 10^{-2}$ ml/g) in sediment-water systems from the Hudson River Estuary at different times following the addition of organic ligands. Error terms are two standard deviations of propagated counting errors.

Station	Time (Hours)	Organic Compound				
		EDTA	NN	PM	Humic	GA
mp 0.1	21	3.92 ± 1.21	3671 ± 1297	707 ± 117	6.36 ± 1.20	4.21 ± 1.24
	44	3.44 ± 1.17	2849 ± 2255	603.5± 64.2	4.31 ± 1.18	4.48 ± 1.19
	92	3.03 ± 1.00	2600 ± 1338	695 ± 75.9	3.19 ± 1.05	3.15 ± 1.05
	165	2.45 ± 0.93	893 ± 391	428 ± 29.93	3.36 ± 1.19	2.35 ± 1.03
mp 18.6	21	1.75 ± 0.91	9159 ± 6026	1124 ± 209	4.15 ± 0.85	2.82 ± 1.06
	44	1.95 ± 1.02	3732 ± 2278	1215 ± 148	5.13 ± 1.23	3.34 ± 1.10
	92	1.80 ± 0.87	2973 ± 1478	1193 ± 277	3.85 ± 1.14	2.64 ± 0.97
	165	1.28 ± 0.84	5239.7± 3544	976 ± 91.08	2.86 ± 1.06	3.60 ± 0.95
mp 43.3	21	4.22 ± 1.04	>3424	1548 ± 418	11.73 ± 1.58	8.73 ± 1.18
	44	4.41 ± 0.99	2509 ± 1273	1352 ± 243	11.58 ± 1.53	10.46 ± 1.55
	92	3.46 ± 0.92	5573 ± 4421	1178 ± 242	12.50 ± 1.44	8.47 ± 1.35
	165	2.65 ± 0.89	2447 ± 959	668 ± 56	10.29 ± 1.40	8.43 ± 1.42

EDTA: ethylene diamine tetraacetic
NN: 1-nitroso - 2-naphthol
PM: 1,10 - phenanthroline monohydrate
Humic: humic acid
GA: glycolic acid

Table 3. Distribution Coefficients for ^{106}Ru($K_d \times 10^{-3}$ ml/g) in sediment-water systems from the Hudson River Estuary at different times following the addition of organic ligands. Error terms are two standard deviations of propagated counting erros.

		Organic Compound				
Station	Time	EDTA	NN	PN	Humic	GA
mp 0.1	21	5.91 ± 0.36	5.72 ± 0.35	6.46 ± 0.42	6.12 ± 0.39	5.93 ± 0.33
	44	5.14 ± 0.33	5.31 ± 0.38	6.80 ± 0.43	6.03 ± 0.36	6.59 ± 0.39
	92	5.09 ± 0.36	4.65 ± 0.30	6.04 ± 0.39	5.06 ± 0.35	4.98 ± 0.32
	165	4.14 ± 0.31	4.27 ± 0.31	5.63 ± 0.38	5.11 ± 0.35	4.75 ± 0.34
mp 18.6	21	3.54 ± 0.26	3.47 ± 0.23	4.12 ± 0.28	3.33 ± 0.21	3.05 ± 0.24
	44	3.48 ± 0.27	3.07 ± 0.28	4.36 ± 0.27	3.42 ± 0.25	3.51 ± 0.24
	92	2.89 ± 0.26	2.65 ± 0.21	3.67 ± 0.28	2.97 ± 0.23	3.42 ± 0.23
	165	2.74 ± 0.23	2.38 ± 0.18	3.25 ± 0.36	2.80 ± 0.21	2.96 ± 0.21
mp 43.3	21	3.39 ± 0.29	3.40 ± 0.25	4.45 ± 0.32	3.85 ± 0.26	3.52 ± 0.22
	44	3.18 ± 0.27	3.35 ± 0.25	4.73 ± 0.32	3.79 ± 0.32	3.75 ± 0.29
	92	3.00 ± 0.25	3.07 ± 0.24	4.25 ± 0.30	3.41 ± 0.25	3.30 ± 0.25
	165	3.03 ± 0.23	2.83 ± 0.24	3.47 ± 0.26	3.01 ± 0.23	2.77 ± 0.24

EDTA: ethylene diamine tetraacetic acid
NN: 1-nitroso - 2-naphthol
PM: 1,10 - phenanthroline monohydrate
Humic: humic acid
GA: glycolic acid

Table 4. Distribution Coefficients for ^{137}Cs ($K_d x 10^{-2}$ ml/g) in sediment-water systems from the Hudson River Estuary at different times following the addition of organic ligands. Error terms are two standard deviations of propagated counting errors.

		Organic Compound				
Station	Time	EDTA	NN	PM	Humic	GA
mp 0.1	21	6.62 ± 0.76	7.08 ± 0.85	6.93 ± 0.85	7.92 ± 0.87	7.98 ± 0.87
	44	7.12 ± 0.80	7.50 ± 0.87	7.03 ± 0.87	7.84 ± 0.87	7.69 ± 0.87
	92	6.66 ± 0.79	6.55 ± 0.80	7.88 ± 0.88	7.23 ± 0.82	7.03 ± 0.81
	165	5.89 ± 0.81	7.73 ± 0.89	8.00 ± 0.95	6.62 ± 0.87	6.99 ± 0.84
mp 18.6	21	5.96 ± 0.71	5.80 ± 0.75	6.26 ± 0.77	6.30 ± 0.72	5.40 ± 0.73
	44	6.91 ± 0.83	6.44 ± 0.80	6.33 ± 0.74	7.53 ± 0.82	6.70 ± 0.83
	92	6.13 ± 0.84	5.59 ± 0.76	6.79 ± 0.82	7.08 ± 0.82	6.42 ± 0.79
	165	5.97 ± 0.69	6.30 ± 0.73	6.55 ± 0.92	6.91 ± 0.80	6.20 ± 0.74
mp 43.3	21	16.92 ± 1.46	17.17 ± 1.34	18.91 ± 1.47	17.97 ± 1.53	16.10 ± 1.11
	44	17.77 ± 1.50	17.43 ± 1.39	18.03 ± 1.47	19.19 ± 1.60	18.21 ± 1.44
	92	17.05 ± 1.48	17.90 ± 1.40	17.31 ± 1.37	19.19 ± 1.47	16.50 ± 1.36
	165	18.86 ± 1.47	18.36 ± 1.46	18.18 ± 1.40	17.48 ± 1.42	17.36 ± 1.49

EDTA: ethylene diamine tetraacetic acid
NN: 1-nitroso - 2-naphthol
PM: 1,10 - phenanthroline monohydrate
Humic: humic acid
GA: glycolic acid

Table 5. Distribution Coefficients for ^{241}Am ($K_d \times 10^{-4}$ ml/g) in sediment-water systems from the Hudson River Estuary at different times following the addition of organic ligands. Error terms are two standard deviations of propagated counting errors.

Station	Time (Hours)	Organic Compound				
		EDTA	NN	PM	Humic	GA
mp 0.1	21	12.34 ± 3.01	>73	19.9 ± 3.4	16.7 ± 10.4	>26
	44	8.94 ± 1.70	25.8 ± 23.1	>44	>82	>49
	92	7.64 ± 3.06	>69	>38	>31	>50
	165	18.3 ±16.9	>25	>64	35.4 ± 26.1	>27
mp 18.6	21	1.72 ± 0.33	43.6 ± 29.0	>40	16.0 ± 4.2	>20
	44	1.60 ± 0.26	19.4 ± 13.7	43.9 ± 26.8	11.16 ± 5.13	45.8 ± 42.8
	92	0.70 ± 0.09	>26	23.1 ± 20.3	11.36 ± 6.32	31.9 ± 29.2
	165	0.73 ± 0.10	>40	45.5 ± 35.5	14.74 ± 4.22	48.2 ± 42.5
mp 43.3	21	1.38 ± 0.25	21.0 ± 17.6	17.5 ± 13.0	7.41 ± 1.02	34.5 ± 22.1
	44	0.91 ± 0.14	>51	>38	3.42 ± 1.26	18.2 ± 13.6
	92	0.59 ± 0.10	>51	23.0 ± 20.0	4.72 ± 1.35	>21
	165	0.57 ± 0.06	32.7 ± 23.6	40.8 ± 27.5	3.78 ± 0.88	26.8 ± 18.8

EDTA: ethylene diamine tetraacetic acid

NN: 1-nitroso - 2-naphthol

PM: 1,10 - phenanthroline monohydrate

Humic: humic acid

GA: glycolic acid