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SPECIFIC ACTIVITY OF RADIONUCLIDES SORBED BY MARINE SEDIMENTS

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ABSTRACT

The problem investigated is whether the binding of radionuclides to sediments in a waste disposal area can be predicted from studies of the trace metal contents of the sediments. To evaluate this, the experimentally determined distribution coefficients of various radionuclides and the existing corresponding stable isotope distribution were compared for different marine sediments. The ratio of the distribution coefficient of the radionuclide to that of its stable counterpart was considered to be a measure of the amount of the stable isotopes available for exchange with radionuclides in seawater. The ratios were normally low but ranging from about 0.1% to 100% for the different elements and sediments tested. The practical application for waste disposal evaluations is that the ratios estimated for a particular area, might be used for a first approximation of the maximum amount of radionuclides that might be held by the sediment at a given specific

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activity in the sea water.

INTRODUCTION

An insight into the radionuclide accumulation by marine sediments cannot easily be given on the basis of the stable metal composition of the sediments. Stable metals are bound partly in the crystal lattices of the sediment particles, and this part will not easily exchange with the metals in solution in the sea water. Therefore the specific activity of radionuclides in the sea water, given in terms of amount of radionuclide per quantity of total stable counterpart, can only be applied to the calculation of the maximum amount of radionuclide that can be held by the sediment when the exchangeable part of the stable metal of the sediment is known.

The purpose of this paper is to investigate the extent of the exchangeable part for different elements that have radioisotopes in fallout and radioactive wastes and for various marine sediments. The number of sediments considered is thirty-five. These are from different origin in the oceans and of different composition, and sampled for the Monaco Laboratory by a number of National Institutions (Table I ; Duursma and Eisma, in press [2]). The radionuclides concerned are ^{90}Sr , ^{137}Cs , ^{106}Ru , ^{59}Fe , ^{65}Zn , ^{60}Co , ^{147}Pm , ^{54}Mn , $^{95}\text{Zr/Nb}$ and ^{144}Ce . Not all their stable counterparts could be determined by neutron activation, and the literature data are used to complete the picture on the stable element composition of sediments and seawater.

The percentage of stable metal ^{in a sediment} /exchangeable with the radioactive counterpart in seawater will be calculated from the ratio of the experimentally determined distribution coefficients of the radionuclide to the distribution coefficients of the stable metals between seawater and sediment. Knowing such ratios, or the range of variation

in these ratios, it will be possible to have a better approach for evaluation of the radionuclide-sediment attachment in a waste disposal area, based on the stable element composition of the sediments and the specific activity of the radionuclide in the seawater.

MATERIALS , ^HMETHODS AND RESULTS

The sediments used for the radionuclide experiments and trace metal analysis were obtained from the three oceans and from several seas (Table I). All the information on the various parameters of the composition of the sediments ^{such as} ~~size~~/grainsize, base-exchange capacity, clay/mineral composition and also on the radionuclide sorption-distribution coefficients, as well as information on the methods used, can be found in the articles of Duursma and Bosch [1] and Duursma and Eisma [2]. The code numbers of the sediments used here are the same. The trace metal analyses of the ^{and some seawater samples} sediments/were carried out by ^I at the Battelle Laboratories neutron activation/with the method described by Robertson, *et al.* . . . . . [3].

The results on the stable metal concentrations in sediment and ^{average} seawater are given in Tables II and IV. The/distribution coefficients for the radionuclides and the stable metals, and their ratios are given in Table III. These ratios , if expressed in percentages supply the percentages of stable metal in the sediment that is exchangeable with the radioactive counterpart and stable metal in seawater.

DISCUSSION

Before going into discussion about the results obtained with the marine sediments, it is worthwhile to realize what exactly the term distribution coefficient means. For the radionuclides, the distribution coefficient gives the amount of radionuclide bound

to the sediment per unit volume (dry weight divided by specific weight of dry sediment) divided by the amount of radionuclide in seawater, also per unit volume. The distribution coefficient thus obtained is a dimensionless factor, characteristic for the conditions at which it was determined experimentally. These conditions must ensure that an apparent equilibrium is obtained, which is for most of the radionuclides the case after a period of some days to some weeks [1,2]. On the other hand the value of the distribution coefficient must not vary much with variation in the quantity of radioisotope and small amounts of carrier and with variation of the sediment/seawater ratio, comparable to what can occur in the sea⁴⁻⁵⁻⁷. Roughly the variation in the distribution coefficients is small enough to ensure that this will not result in unreliable figures which can be used for a first evaluation programme.

For the distribution coefficients of the stable metals, which are also given as the same dimensionless factors, the problem is whether the sediment of the sea bottom is really in equilibrium with the seawater as far as the exchange of trace metals is concerned. This problem needs to be settled first before discussing the general results, given in Table III. Hitherto an experiment was set up where four marine sediments, and one fresh-water sediment were equilibrated with seawater for a period of three weeks. The conditions were kept strictly under control during the experiment so that no contamination by trace metals could occur, also not during sampling and the ensuing trace metal analysis. Six grams (dry weight) of wet preserved sediment in three litres of seawater were shaken regularly at a temperature of 4-8°C. After centrifugation, the seawater and sediment from each flask was analysed together^{with} the original seawater and sediment. The results are shown in Table IV. In general the original seawater and sediment concentrations of the determined trace metals are of the same order of magnitude as those of the shaken samples.

(sediment No. 6)

Some large variations/are probably exceptions, since for these values a budget calculation of the elements, which are determined ^{both} in the water and in the sediment, does not show a close balance. Taking the experiment as a whole, it would seem sensible that a discussion of the distribution coefficients of stable metals in the sediment/seawater systems should be made.

The way in which both the distribution coefficients of the radionuclides and stable metals are used is by taking the ratio of the two. This means the ratio of two ratios :

$$\frac{\text{amount of radionuclide in sediment}}{\text{amount of radionuclide in seawater}} ; \frac{\text{amount of stable metal in sediment}}{\text{amount of stable metal in seawater}} \quad (1).$$

Converting this relation it might read :

$$\frac{\text{am. radionuclide in sediment}}{\text{am. stable metal in sediment}} ; \frac{\text{am. radionuclide in seawater}}{\text{am. stable metal in seawater}} \quad (2),$$

which is simply the ratio of the specific activity in sediment to that in seawater.

If exchange of radionuclide with stable metal in the sediment is completely possible, and if the radionuclide in the seawater is in the same form(s) as the stable metal, then relations (1) and (2) should be equal to 1 or 100%. Any lower value will indicate that the exchangeability of the stable metal of the sediment with radionuclides of the seawater (equally with stable metals of the seawater) is only partly possible. An exception needs to be made for those elements where the stable metal and radionuclide in the seawater are not in the same physical-chemical form. This is primarily the case when part of the stable metal in seawater is bound to organic complexes which do not exchange with ionic inorganic forms, and therefore also not with the metal of the sediment. In such a case the distribution coefficients of the stable metal has to be calculated from only the concentration of the exchangeable part of the metal in

As the data of Table III show, a relatively large fraction of the sediments of the stable metal is exchangeable in the case of strontium and cobalt. For the others only a minor part of the stable metal is exchangeable with its radioactive counterpart or stable metal in seawater. For all elements the range of variation in the exchangeability with various sediments is relatively large. This range is determined by taking the variations of both distribution coefficients into consideration. The highest value of the range is the ratio of the highest distribution coefficient of the radionuclide divided by the lowest value of the distribution coefficient of the stable metal; the lowest value of the range is determined by the reverse procedure. Another method of approach would have been to calculate the ratio of the distribution coefficients of each sediment and radionuclide and stable metal, and take from these values the mean value for each element. This approach, however, did not result in better figures, so the results of the former approach (Table III) are taken for further discussion.

It is very difficult to give general rules for different trace-metals in sediments with regard to their exchangeability with dissolved radionuclides and trace metals, but some indications can be given. As extensively discussed by Duursma and Eisma [2], the sorption of radionuclides by marine sediments can be grouped into three categories: (i) the radionuclides sorbed by isotopic-exchange processes with kinetics comparable to ion-exchange processes (^{90}Sr , ^{137}Cs , and in some way also ^{65}Zn); (ii) the radionuclides that are isotopically exchanged by precipitation and dissolution reactions, often with the formation of compounds with the sedimentary particles (^{59}Fe , ^{95}Zr); and (iii) the radionuclides that show a mixture of both former kind of reactions (^{60}Co , ^{54}Mn , ^{106}Ru , ^{147}Pm , ^{144}Ce , where ^{147}Pm has no natural stable counterpart). As these radionuclide reactions are

- 7 -

equally representative of the reactions possible with the stable metals, it is understandable that the exchangeable part of the stable metals in the sediments can be expected to be greater for those elements whose sorption and desorption is by a kind of exchange reaction with kinetics comparable to ion-exchange reactions. Although this is probably the case with strontium and partly with cobalt with their elevated exchange percentages, it is not the case so much for cesium. However, the ^{137}Cs sorption kinetics are very much of the ion-exchange type [2], and ^{137}Cs sorption has out of all the radionuclides tested the best relationship with both the claymineral composition of the sediment and its base-exchange capacity. It must therefore be suspected that a large part of the stable cesium is bound inside the crystal lattices of the sedimentary particles. This is not necessarily only for the clayminerals, because ~~there~~ could not be found a proportionality between the stable cesium concentration of the sediments and the illite or other claymineral concentration in the sediment. Other kinds of relationships have been sought, for example between the base-exchange capacity of the sediments [2] and the concentration of the various stable metals in the sediment, but again without finding a proportionality. Only the concentration of stable cobalt in sediments was in some rough way proportionally related to the base-exchange capacity of the sediment, which, however, was not the case for the ^{60}Co distribution coefficients [2].

It can be concluded therefore that ^{for the moment,} ~~in general~~ for the evaluation of the maximum amount of radionuclides that can be held by marine sediments, the stable element concentration and the factor of exchangeability might give the best approximate results.

It should be realized that the exchangeability factors as given here are not applicable for pollution studies where stable metals arrive in the sea. The specific activity approach for radionuclides requires that the radionuclides plus carrier do not disturb the natural equilibrium between the metals in seawater and sediment. Any

introduction of large amounts of stable metal exceeding the concentrations of the natural stable metals in seawater will cause chemical reactions other than isotopic exchange, ^{as} such ^{ion-exchange} reactions with other metals on the sediment or precipitation. This fact also holds for radioactive effluents which are enriched ^{with} $\frac{1}{2}$ large quantities of ^{for example} carrier, ~~like for~~ chromium added ^{an} as ^{anti-corrosive} substances in the cooling water of reactors. In such a case the specific activity in the water is lower, but the reactions can not be predicted on the basis of the specific activity as is done here.

The specific activity approach for sediments also does not hold for fresh-water sediment introduced into the sea, since this sediment is not in equilibrium with seawater in terms of its trace metal balance. This is shown by the relatively high Sr strontium, cesium and zinc values for the "exchangeability" of sediment No. 56 in Table IV.

For waste-disposal evaluations, where this is not the case, the calculated percentages of the exchangeability of the elements in sediments mentioned here, give some background material for judging the maximum quantities of radionuclides that might be bound by the sediments in a future disposal area. It must be remembered that these data only concern the maximum quantities, and that the real quantities depend on the pattern in which the radionuclides are transported by horizontal and vertical diffusion in the water, and on the degree of scavenging of the radionuclides by the sediments brought into suspension by currents and tides. This degree of scavenging depends on the hydrographical and sedimentological conditions in the particular area and on the distribution coefficients of the radionuclides themselves (see therefore [2]).

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Table I. Provenance and characteristics of marine sediment samples.

The donors are acknowledged for the supplying of the samples [2].

Sample No.	Ocean, Sea Location	Depth m	Ship, Code, Donor	Type
ATLANTIC OCEAN				
1	N. Atlantic 75°51' N 14°30' W	375	ANTON DOHRN, 287/10 Dr. K. Vollbrecht Hamburg	Grab, grey, dry.
3	C. Atlantic 27°53' N 45°55' W	4020	CHAIN, Ch-61-174 9/XII/66 Dr. J. Philips Woods Hole	Core, 43 cm /5 cm; light brown, moist.
4	C. Atlantic 26°42' N 39°23' W	4150	CHAIN, Ch-61-171 6/XII/66 Dr. J. Philips Woods Hole	Core, 45 cm /5 cm; light brown, moist.
5	C. Atlantic 26°13' N 52°28' W	5384	PILLSBURY P-6303-21A Dr. F.F. Koczy Miami	Core, 129 cm /5 cm; brown, moist.
6	Tongue of Bahamas 24°48' N 77°32' W	1828	GERDA, G-6017-23 Dr. F.F. Koczy Miami	Core, 134 cm /5 cm; light grey, moist.
8	Caribbean 14°57' N 68°55' W	4126	PILLSBURY P-6304-9 Dr. F.F. Koczy Miami	Core, 125 cm /5 cm; light brown, moist.
9	Off Ivory Coast 04°35' N 06°45' W	60	REINE-POKOU 17/X/69 Dr. L. Martin Abidjan	Grab, grey, moist.
11	North of Amazone 06°13' N 52°03' W	170	CALYPSO April 1968 Comm. J.Y. Cousteau Monaco	Grab, sandy, moist
12	Amazone Delta 02°27' N 48°00' W	110	CALYPSO April 1968 Comm. J.Y. Cousteau Monaco	Grab, sandy, moist.
13	Romanch Trench 00°09' N 18°19' W	7200	KURCHATOV, cruise 1 Jan/Feb. 1967 Dr. J.A. Bogdanov Moscow	Grab, brown, moist.
18	Cape Good Hope 43°02' S 18°00' E	200	CALYPSO 16/III/1968 Comm. J.Y. Cousteau Monaco	Grab, sandy, moist.

Sample No.	Ocean, Sea, Location	Depth m	Ship, Code, Donor	Type
ATLANTIC OCEAN				
20	Antarctic Sea 57°34' S 23°58' W	7648	VEMA, V14-55 Dr. R. Ku Lamont	Core, 100 cm /5 cm; grey, dry.
PACIFIC OCEAN				
21	S.E. Alaska 57°31' N 133°00' W	80	MAYBESO, station no. 5 Dr. T. Loder College, Alaska	Grab, grey, moist.
24	C. Pacific 24°58' N 176°16' E	5879	VEMA, V14-67 Dr. R. Ku Lamont	Core, 90 cm /5 cm; red clay, moist.
25	E.Tr. Pacific 18°16' N 131°46' W	5210	SPENCER F. BAIRD Jyn-V-50P, 10/IX/61 Dr. W.R. Riedel La Jolla	Core, 300 cm /5 cm; brown, moist.
26	E.Tr. Pacific 01°20' N 114°54' W	3800	ROBERT CONRAD RC-10-70 Dr. R.Ku Lamont	Core, 90 cm /5 cm; light brown, moist.
27	C.Tr. Pacific 07°30' S 157°41' W	5455	AMPHITRITE AMPH-90-P Dr. T. Walsh La Jolla	Core, 300 cm /5 cm; brown, moist.
28	Off New Zealand 41°42' S 175°28' E	965	TARANUI D-596 Dr. W.v.d.Linden Wellington	Core, 235 cm /5 cm; grey, moist
29	S. Pacific 50°02' S 127°31' W	3970	USNS ELTANIN PH-25-4B Dr. H.G. Goodell Tallahassee	Core, 53 cm /5 cm; white, moist.
INDIAN OCEAN				
30	Arabian Sea 20°07' N 67°55' E	3047	ATLANTIS II PC-16 Dr. D. Wall Woods Hole	Core, 1088 cm /unreg.cut grey, dry.
31	Bombay Harbour 18°60' N 72°57' E	11	MYSIS 15/X/68, No. 63 Dr. B. Patel Bombay	Grab, grey, moist.

Sample No.	Ocean, Sea Location	Depth m	Ship, Code, Donor	Type
INDIAN OCEAN				
33	Off Ceylon 05°39' N 82°13' E	3994	OCEANOGRAPHER 52-Gday-OPR-476 4/VII/67, Dr. W.S. Moore, Stony Brook	Core, 100 cm /5 cm; grey, moist.
34	Off N. Sumatra 05°29' N 92°35' E	4227	OCEANOGRAPHER 177-Gday-OPR-476 1/VII/67, Dr. W.S. Moore, Stony Brook	Core, 145 cm /5 cm; grey, moist.
35	C.Tr. Indian Ocean 00°45' N 74°07' E	2679	OCEANOGRAPHER 52-Gday-OPR-476 2/VII/67, Dr. W.S. Moore, Stony Brook	Core, 225 cm /5 cm; grey, moist.
ARCTIC SEAS				
37	Laptev Sea 73°31' N 135°15' E	26	INEY 3/1966 Dr. A.F. Trshnikov Leningrad	Core, 52 cm /2 cm; grey, dry
BALTIC SEA				
41	C. Baltic Sea 57°22' N 19°58' E	220	ARANDA 1967 Dr. B. Winterhalter Otaniemi (Finland)	Core, Up 18.5 cm, down 28 cm; grey, moist. Recent + glacial.
NORTH SEA				
43	Europort 51°58' N 04°04' E	15	GEOPOTES VII 21/V/69 Ir. J. Ringers Rotterdam	Grab, black suspension.
MEDITERRANEAN				
44	W. Mediterranean 37°26' N 01°05' E	2720	METEOR M7/66/6a, X/66 Dr. E. Seibold Kiel	Core, 150 cm /10 cm; grey, moist. (Stat. 210 Swed.exp)
50	Off Toulon 42°47' N 07°29' E	2690	METEOR 24/X/66 Dr. H. Kautsky Hamburg	core, 40 cm unreg. cut grey, moist.
54	Gulf of Trieste 45°38' N 13°43' E	19	VERCELLI 32, II/67 Dr. G. Macchi Trieste	Grab, grey, moist.

Sample No.	Ocean, Sea Location	Depth m	Ship, Code, Donor	Type
MEDITERRANEAN				
55	E. Mediterranean 35°31' N 28°12' E	2360	CHAIN Ch-61-50-12, 12/IX/66 Dr. E.F.K. Zarudski Woods Hole	Core, 86 cm /5 cm; brown, grey, black, moist.
56	Var-river (Nice) 43°40' N 07°12' E	1	— VII/68 Dr. E.K. Duursma Monaco	Grab, grey, moist.
BLACK SEA				
58	N. Black Sea 43°50' N 35°31' E	2159	PILLSBURY P-6507-12 Dr. F.F. Koczy Miami	Core, 144 cm /5 cm; black, moist.
RED SEA				
59	Discovery Deep 20°17' N 38°02' E	2080	CHAIN Ch-61-80, X/66 Dr. D.A. Ross Woods Hole	Core, 164 cm /5 cm (mainly); brown, black, grey, moist.
60	EL MAR MENOR (Lagoon) 37°40' N 0°40' W	16	OLE X/71 Dr. S.W. Fowler Monaco	Grab, black, silty, wet.

ELEMENTAL CONCENTRATIONS OF OCEAN SEDIMENT
($\mu\text{g/g}$ except as noted)

Sample #	Na(%)	Fe(%)	La	Sm	Br	Co	Sc	Cr	Eu	Hf	Th	Tb	Ta	Cs	Sr	Sb	Zn
1	1.54	4.15	31	4.8	83	11	11	92	1.0	6.8	5.2	0.65	1.9	4.8	120	0.61	
3	1.16	1.08	22	3.7	71	17	3.5	16	0.70	0.80	2.1	0.54	0.44	1.5	1450	0.61	
4	1.28	0.92	17	2.7	86	15	3.2	14	0.58	1.0	1.8	0.44	0.61	1.2	1410	0.52	
5	2.22	5.58	61	9.7	64	83	17	89	1.9	5.9	11.0	1.1	2.3	8.6	300	1.3	
6	0.86	0.068	1.9	0.52	110	0.39+0.04	0.26	8.3	0.048	0.085	0.18	<0.10	0.006+0.060	0.14+0.05	5620	0.24	
8	1.40	1.64	18	2.4	63	11	5.7	34	0.51	1.1	2.5	0.25	0.60	2.4	1470	0.51	
9	1.93	4.78	39	4.9	240	12	12	122	1.1	9.1	8.1	0.55	2.8	4.7	370	0.37	
11	1.11	6.48	26	4.1	63	9.3	7.0	54	0.72	18.7	4.8	0.51	1.9	3.0	390	1.2	
12	0.42	0.67	17	2.1	40	1.7	1.1	43	0.46	4.3	1.1	0.13	0.72	0.34	940	0.18	
13	1.98	2.68	25	3.5	180	9.4	9.1	74	0.70	5.0	3.8	0.61	1.2	3.2	750	1.2	
18	0.55	9.13	56	10.8	63	3.0	6.1	190	2.2	1.6	3.3	1.9	0.26+0.06	2.6	360	2.4	
20	2.38	5.11	8.7	2.5	67	19	26	37	0.70	2.9	1.9	0.64	0.41	1.8	1750	0.73	
21	2.67	5.27	32	4.2	49	20	16	67	1.1	3.7	5.8	0.65	1.6	3.0	700	0.17	
24	2.03	5.53	53	8.5	67	85	19	93	1.7	5.7	8.6	1.3	2.1	15	4710	2.7	5340
25	2.29	5.14	50	8.6	66	98	20	80	2.0	6.3	8.0	1.5	1.8	1.3	390	2.1	
26	1.35	0.23	6.3	1.2	100	3.8	2.0	4.0	0.24	0.26	0.12	0.25	0.03+0.02	0.81+0.14	1890	0.47	850
27	3.79	5.44	174	34	120	195	29	58	9.6	6.0	6.7	7.2	0.96+0.07	2.3	590	16	
28	2.69	3.39	30	4.3	110	8.1	11	55	0.91	5.9	6.0	0.31	1.4	6.6	340	0.37	
29	1.04	0.42	8.3	1.4	59	4.5	2.4	7.1	0.43	0.53	0.76	0.21	0.22	0.71	1210	0.16	
30	1.44	3.14	21	3.2	140	12	10	84	0.68	3.6	3.7	0.43	1.2	4.3	2280	0.92	
31	2.11	8.04	24	4.4	130	34	29	160	1.4	5.6	3.5	1.3	1.3	3.6	400	0.94	
33	2.50	5.87	35	4.7	210	26	19	130	1.0	4.1	6.5	0.75	1.4	5.2	420	1.6	
34	1.64	4.33	42	5.1	52	16	13	88	1.1	6.1	9.4	0.62	2.0	12	60+100	0.91	
35	1.77	4.85	27	3.6	130	17	14	120	0.80	3.9	4.3	0.57	1.3	5.6	730	1.1	
37	2.28	5.22	49	6.4	140	20	15	84	1.4	5.7	6.8	0.77	1.7	6.7	280	1.8	
41	0.82	3.57	43	5.6	20	12	11	68	0.94	5.4	6.2	0.70	1.5	5.6	120	0.90	
43	1.45	2.21	25	3.4	120	8.4	6.0	120	0.78	6.8	3.3	0.57	1.3	5.4	350	1.3	
44	1.14	2.55	27	3.4	61	8.8	8.6	70	0.69	3.5	3.9	0.47	1.3	7.7	520	1.2	
50	1.10	2.58	30	3.6	66	12	9.3	73	0.76	3.5	4.4	0.59	1.5	9.6	770	0.86	
54	1.48	3.47	23	3.1	87	15	11	200	0.61	3.4	3.5	0.57	1.1	6.9	540	1.1	
55	1.23	3.54	25	3.4	80	25	11	240	0.78	3.4	2.9	0.52	1.4	4.3	910	0.85	
56	0.40	2.07	20	3.0	4.0	6.4	6.1	36	0.57	4.0	3.2	0.39	0.94	4.6	570	1.0	
58	1.84	3.51	29	5.0	290	21	12	87	0.85	5.1	4.3	0.82	1.3	6.1	380	4.0	
59	15.98	3.79	2.3	0.51	230	12	1.4	13	0.17	0.53	0.30	0.06	0.12+0.03	0.55	1030	2.0	780
60	2.80	2.28	21	3.4	470	6.8	7.0	49	0.64	2.8	3.3	0.51	1.1	6.3	1870	4.0	1870

Mean	1.67	3.62	32.0	5.0	112	27.5	11.0	79	1.14	4.4	4.3	0.81	1.29	453	1022	1.61	2210
minus parameter	0.85	"	"	"	72	"	"	"	"	"	"	"	"	"	1023	"	2211
$\pm$	0.6	1.71	16.1	2.9	60	21.4	5.6	41	0.70	2.2	2.1	0.54	0.52	2.54	764	1.26	1565

$$K_{\text{stable metal}} = \frac{(\mu\text{g/g}) \times 2.7}{(\mu\text{g/l}) \times 10^{-3}} = \frac{\mu\text{g/g}}{\mu\text{g/l}} \times 2700$$

Correction pore water: 1 gr dry sed. $\hookrightarrow$ 0.6 ml seawater: Substit seawater conc. $\times$ 6
 $(\mu\text{g/l})$

Chert. data: MnO_2 0.4-1.97% oxide $\rightarrow (0.25-1.25) \times 10^4 \mu\text{g Mn/g}$

~~126-283 $\mu\text{g/g}$~~

126-283 $\mu\text{g/g}$

TABLE II - B

ESTIMATED ANALYTICAL UNCERTAINTY IN THE STABLE ELEMENT
MEASUREMENTS OF MARINE SEDIMENTS BY NEUTRON ACTIVATION ANALYSIS

<u>Element,</u> <u>Concentration Range</u>	<u>Uncertainty</u> <u>(% Error)</u>
Na, 0.5 - 1.0%	10
>1.0%	5
Fe, 0.05 - 1.0%	10
>1.0%	5
La, 1 - 10 ppm	10
>10 ppm	5
Sm, <1 ppm	10
>1 ppm	5
Br, >10 ppm	10
>10 ppm	5
Co, <1 ppm	10
>10 ppm	5
Sc, <2 ppm	10
>2 ppm	5
Cr, <20 ppm,	10
>20 ppm	5
Eu, <0.2	10
>0.2	5
Hf, <0.5	10
>0.5	5
Th, <1	20
>1	10
Tb, <0.5 ppm	20
>0.5	10
Ta, <1 ppm	20
>1 ppm	10
Cs, <1 ppm	20
>1 ppm	10
Sr, <500 ppm	20
500-1000 ppm	10
>1000	5
Sb, <0.5 ppm	10
>0.5 ppm	5
Zn, <800 ppm	20
>800 ppm	10

TABLE III

Exchangeability of some trace metals of marine sediments, calculated from the ratio of the distribution coefficients of radionuclides and stable metals. The mean stable metal concentrations in the sediments is calculated from the data of Table II for Sr, Cs, Fe, Zn and Co. The Mn and Zr values are derived from Chester [8]. The Ce values are taken as 26 times the Eu values of Table II, where the value 26 is taken from the mean ratio between these elements of Table IV. The seawater concentrations are from Goldberg et al [6], except for Ru [9] and Zn [7], The average distribution coefficients of the radionuclides were determined by the thin-layer technique [1,2] for only 28 of the sediments. Sediment No 56 is not taken into regard, because this is a fresh-water sediment.

	Sr	Cs	Ru	Fe	Zn	Co	Pm	Mn	Zr	Ce
Sediment					*					
mean $\mu\text{g/g}$	1023	4.53	?	3.7×10^4	2210	24.5	-	0.75×10^4	205	29.6
variation $\mu\text{g/g}$ ($\pm$)	764	2.54		1.7×10^4	1565	21.5		0.5×10^4	78	18.2
Seawater $\mu\text{g/l}$	8000	0.3	10^4	3	5	0.4 ^{0.04}	-	2	3×10^{-2}	10^{-3}
K_{stable}	350	40000	?	3.3×10^7	1.2×10^6	1.7×10^5 ⁵⁶	-	1.0×10^7	1.8×10^7	8×10^7
variation ($\pm$)	260	22000		0.75×10^7	0.8×10^6	1.5×10^5 ⁵⁶	-	0.7×10^7	0.7×10^7	5×10^7
$K_{\text{radionuclide}}$	354	1153	4.5×10^4	2.7×10^4	5.1×10^3	6.0×10^4	6.1×10^4	3.8×10^4	1.1×10^4	9.0×10^4
variation ($\pm$)	427	802	2.3×10^4	2.5×10^4	4.3×10^3	7.5×10^4	2.8×10^4	4.4×10^4	0.7×10^4	5.0×10^4
$(K_{\text{rad}}/K_{\text{st}}) \times 100\%$	100	2.9	?	0.08	0.43	$35/3$ ^{2.53}	-	0.38	0.06	0.11
max. range	100	10.9	-	0.3	2.4	$100/675$	-	3.1	0.16	0.47
min. range	0	0.6	-	0.02	0.04	0	-	0	0.016	0.03

* only the average from 3 samples

TABLE IV -A

Trace element concentrations of four marine sediments and one fresh-water sediment before and after shaking for three weeks with seawater. The quantities used were 6 grams of (dry) weight of sediment in three litres of seawater. The flasks were kept at 4-8°C. The original seawater and sediment is marked by (O), and the treated by (T). The values of the sediment samples are somewhat erratic compared to those given in Table II, which were determined with better neutron activation elemental standards than those given here. The trace metal values for the seawater samples were very accurately done. ~~xxxxxx~~

TABLE IV -B

For Sr,Cs,Fe,Co and Ce the percentages of the exchangeability of the trace metals are calculated with help of the known $[^{-2}]$ distribution coefficients of the radionuclides, determined with the thin-layer technique. The zinc exchangeability is calculated with the concentration of 40 $\mu\text{g/g}$ of zinc in sediment No. 56, which was determined by chemical methods by Dr.C.N.Murray, Monaco.

TABLE IV - A

		in $\mu\text{g/l}$											
SEAWATER			Zn	Fe	U	Cs	Sb	Co	So	Sr-relative	Rb-relative		
Original (0)			26	6	3.8	0.33	0.26	0.023	0.0023	to (0)	to (0)		
Treated with 47			5	51	5.5	0.37	0.43	0.046	0.0073	1.00	0.99		
(T)	-	25	5	86	10.4	0.54	0.36	0.128	0.0165	1.05	0.96		
(T)	-	29	71	26	5.3	0.38	0.31	0.069	0.0063	1.14	1.05		
(T)	-	6	23	24	4.6	0.37	0.38	0.083	0.0070	1.00	0.91		
(T)	-	56	6	33	5.4	0.51	0.37	0.032	0.0038	1.16	1.04		
SEDIMENTS			in $\mu\text{g/g}$										
			Sr	Cr	Sc	Co	Ce	Eu	Sb	Cs	Hf	Th	Fe %
(O) - 47	1700	96	10	14	42	0.94	0.12	5.5	1.9	8.8	3.2		
(T) - 47	1000	130	12	19	49	1.7	0.17	6.4	2.1	10	4.0		
(O) - 25	800	87	21	130	87	2.7	0.33	8.4	4.0	15	5.8		
(T) - 25	800	65	21	104	83	4.4	0.34	8.4	5.0	18	2.8		
(O) - 29	3400	4.8	1.9	4.8	5.6	0.54	0.018	0.39	0.33	1.3	0.38		
(T) - 29	3500	3.6	1.6	4.2	4.0	0.36	0.024	0.33	0.13	0.96	0.33		
(O) - 6	3100	38	7.9	20	3.5	0.87	0.053	2.3	1.7	5.2	2.5		
(T) - 6	13000	7.5	0.25	0.45	1.9	0.098	0.018	0.48	0.31	0.16	0.069		
(O) - 56	1400	37	6.2	7.6	29	0.70	0.10	3.1	6.5	6.1	2.1		
(T) - 56	1800	33	6.6	8.5	38	0.73	0.11	3.0	2.8	6.1	2.3		

TABLE IV - B

	$(K_{\text{radionuclide}} / K_{\text{stable element}}) \times 100\%$					
	Sr	Cs	Fe	Zn	Co	Ce
(O) - 47	7.6	0.20	0.03		0.22	0.28
(T) - 47	12.9	0.15	0.025		0.17	0.23
(O) - 25	42.0	0.32	0.08		3.6	0.32
(T) - 25	42.0	0.42	0.17		4.6	0.35
(O) - 29	5.5	0.50	0.51		1.75	0.85
(T) - 29	5.3	0.67	0.58		1.90	1.20
(O) - 56	8.8	0.06	0.71		0.08	7.1
(T) - 6	2.1	0.32	26.5		3.6	13.1
(O) - 56	270	63	1.47	81	4.15	0.93
(T) - 56	210	70	1.35		3.76	0.72

- 6T -