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RADIOACTIVITY OF OCEAN SEDIMENTS. IV.
THE RADIUM CONTENT OF SEDIMENTS OF THE CAYMAN TROUGH.

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ABSTRACT. The radio-elements are not in equilibrium in the uppermost layers of the sediments at the bottom of the ocean. Of these elements uranium, ionium, and radium have a sufficiently long half-life to be of importance. A history of these three elements is reflected in the variation of the radium content of ocean sediments with the depth below the ocean floor. Measurements of this variation demonstrate that the concentrations of uranium, ionium, and radium at any given time are established by the usual laws of radioactivity governing the growth and decay of radio-elements in a system that is not in radioactive equilibrium. The experimental results must be adjusted to the conditions that pertained to the undisturbed sediments. This requires a knowledge of the history of the specimens from the time that the sediment was taken by the core-sampler to the time at which the specimens were analyzed. The relation between radium content and depth in a given ocean sediment promises a method of determining the rate of accumulation of the deposit at that place.

INTRODUCTION.

DETERMINATIONS of the radium content of the sediments lying below the ocean bottom in the Cayman Trough between Cuba and Jamaica reveal the interrelations of the radio-elements in a far more detailed manner than do the earlier results from a North Atlantic core.[1] This North Atlantic core, P-124, is an unusual case of the general relations that can now be deduced from radium measurements of sediments from lower latitudes. Consequently, the tentative conclusions that were drawn from the results for this core must be partly revised.

During a systematic study of cores from the North Atlantic obtained by Piggot aboard the Cable Ship *Lord Kelvin* in 1936,[2] it became apparent that, though the radioactive rela-

tions were in agreement with the hypothesis derived from the measurements reported here, the alternating zones of sediments deposited in warm and cold water[3] resulted in complications that will necessitate a special study of the constituents of these zones.

The sediments at lower latitudes should be free of such complications. Cores of the ocean bottom in the Cayman Trough of the Caribbean Sea were obtained by Piggot in 1937 aboard the research ketch *Atlantis* of the Woods Hole Oceanographic Institution. The apparatus to secure vertical sections of the ocean bottom in the form of cores has been fully described in previous publications.[2, 4]

DESCRIPTION OF CORES.

Two cores from the Cayman Trough were taken from the vicinity of the Bartlett Deep. South of the Bartlett Deep the bottom of the Trough forms an extensive plateau lying between 4600 and 5100 meters below the surface of the Caribbean Sea. Core P-136 was obtained from about the center of this plateau west of Jamaica. Core P-135 was obtained from the same plateau at the eastern end of the Cayman Trough just south of the Oriente Deep which parallels the south coast of Cuba. The gradient of the north slope of the Bartlett Deep is steep. Core P-137 was secured from a point well down on this slope between Little Cayman Island and the bottom of the Bartlett Deep, which is about 7000 meters.

The cores from the Cayman Trough have not been examined lithologically and chemically in as great detail as were those from the North Atlantic suite,[3] but the foraminifera have been carefully studied by Cushman.[5] The sediment is a Globigerina ooze. Cores P-135 and P-136 each contain a stratum composed almost entirely of foraminifera shells, which occurs in each core at about 66 cm. below the surface of the bottom and is about 2 cm. thick. The North Atlantic core P-124 that will be discussed in the light of new data has been fully described.[1]

RADIUM MEASUREMENTS.

The apparatus and technique for the determination of small quantities of radium has been described elsewhere.[6]

The experimental results are expressed by weight of air-dried sediments. Preliminary "water minus" analyses made at

120° C. for core P-137 in the air-dried state show a nearly constant free-water content of between one and two per cent at various depths in the core. The effect of changes of humidity during storage, if significant, can be avoided by determining the water content when the samples are removed for radium analyses.

The radium results discussed here are based on an apparatus constant determined with the standard solution containing 10^{-11} g. Ra which has been prepared by the National Bureau of Standards.

TABLE I.

The Radium Content of Ocean-Bottom Cores from the Cayman Trough.

Core No. P-135. Depth of water 4900 meters.
Latitude 19° 18′ N. Longitude 76° 48′ W.

Specimen No.	Depth cm.	Radium Content in 10^{-12} g./g.
P-135-0	0- 1	0.95±0.01
P-135-5	5- 6	1.30 0.02
P-135-14	14- 15	1.60 0.02
P-135-20	20- 21	1.83 0.02
P-135-36	36- 37.5	2.11 0.03
P-135-55	55- 57	1.25 0.02
P-135-75	75- 78	1.24 0.02
P-135-82	82- 83	1.20 0.02
P-135-90	90- 91	1.35 0.03
P-135-99	99-100	1.31 0.02
P-135-109	109-110	2.18 0.03

Core No. P-136. Depth of water 4650 meters.
Latitude 18° 38′ N. Longitude 79° 12′ W.

P-136-0	0 - 1	1.42±0.03
P-136-4	4 - 5	1.99 0.02
P-136-15	15 - 16	2.57 0.04
P-136-20	20 - 21	2.73 0.03
P-136-31	30.5- 32	2.52 0.03
P-136-46	46 - 47	2.31 0.03
P-136-59	59 - 62	2.28 0.02
P-136-71	71 - 72	2.01 0.03
P-136-83	83 - 86	2.13 0.03
P-136-100	100 -102	2.23 0.03
P-136-115	115 -116	2.30 0.04
P-136-133	133 -134	1.69 0.02
P-136-158	157.5-159.5	1.25 0.02
P-136-189	189 -190	0.96 0.01

Core No. P-137. Depth of water 4890 meters.
Latitude 19° 14′ N. Longitude 80° 20′ W.

P-137-0	0 - 1	1.40±0.02
P-137-2	2 - 3	1.64 0.02
P-137-8	8 - 10	2.21 0.04
P-137-13	13 - 14	2.34 0.03
P-137-19	19.5- 20.5	2.68 0.03
P-137-29	29 - 30	2.48 0.03
P-137-59	59.5- 60.5	1.88 0.02
P-137-105	105 -107	1.21 0.02
P-137-144	144 -145	0.90 0.01
P-137-188	188 -190	0.54 0.01

DISCUSSION.

Distribution of Radium. Table I presents the results obtained from the three cores of the Cayman Trough. The graphical representation in Fig. 1 for core P-137 will serve for a discussion of the radioactive relations. The outstanding features of the distribution of radium with depth, below the surface of the ocean bottom, are a rapid rise and pronounced maximum of the radium content at a depth of about 20 cm., followed by a gradual decrease to the bottom of the core. Such a maximum radium content has been found at various depths in several cores from the North Atlantic ocean.

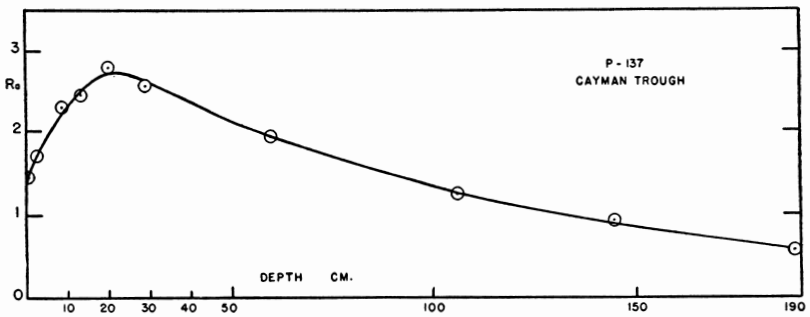

Fig. 1. The radium content in 10^{-12} g. per g. plotted as a function of depth in a core of Globigerina ooze. The radium values plotted here are 4.1 per cent higher than will be found in Table I. This represents the difference between our laboratory radium standard and the new standard prepared by the National Bureau of Standards.

As has been pointed out,[1] it is possible to explain such a distribution of radium in the ocean sediments by assuming a rate of deposition of radium from the ocean water that varies with time, according to a function that must bear a relation

complementary to the summation of the two exponential terms represented by the curve in Fig. 1. There is little likelihood of such a special variation of the rate of deposition with time.

Interpretation. The distribution shown in Fig. 1 may be interpreted on the basis of the growth and decay of radio-elements in systems not in equilibrium. The variation of the radium content with depth in the sediment reflects the change in relation to time of the radium content of any particular level. There is some radium being deposited from the water, and also more ionium than is necessary to maintain this particular radium concentration, but much less uranium than is required to maintain this ionium concentration. The radium content therefore increases until equilibrium with the ionium is established.[a] It then decreases, as the ionium decreases, until the ionium content reaches that value fixed by equilibrium with the uranium present. The half-life of uranium-238 being 4.5×10^9 years, the radium content should remain essentially constant below the level at which the latter equilibrium is established. The ionium-uranium equilibrium has not been attained at the depths in the sediment reached by the cores described here. Preliminary measurements of the uranium content of core P-137[7] indicate that the constant radium concentration below the bottom of this core should be about 0.35×10^{-12} g. Ra per g. of air-dried sediment, a value which is in agreement with the radium content of many sedimentary rocks. This fact is important in considering the flow of heat beneath the ocean basins.

Such an interpretation of the variation of the radium content below the surface of the ocean bottom appears to apply, generally, to the bottom sediments of such widely separated regions as the North Atlantic, the Caribbean Sea, and the Pacific Ocean, and to sediments of such varied characteristics as Blue mud, Globigerina ooze, and Red clay. A Red clay sediment exhibits a variation of this interpretation in that the ionium is deposited at a lower concentration than that required to maintain the initial radium content. Therefore there is no maximum radium content but a steep initial fall in radium concentration followed by a more gradual decrease which is controlled by the ionium of longer life.

This hypothesis, stated qualitatively above, can be given

[a] This is the conclusion represented by curve IV in discussing the radium content of a North Atlantic core. Reference 1.

rigid mathematical treatment by expressions involving the three exponential functions governing the growth and decay of these three radio-elements. By applying these expressions to the relation of radium content with depth, information on the rate of deposition of ocean sediments can be obtained.

Results, Expressed on a Volume Basis. An analysis of these results, to determine rates of deposition, requires a conversion of the measurements to the radium content per unit cross-section of sediment (cm.^{2}) per unit of depth (cm.). The conversion factor may vary with respect to the character of the sediment and its depth below the ocean floor. One would expect the weight of solid material per centimeter cube to increase somewhat with the depth because of compaction. This is true, for example, in the core of Blue mud from the North Atlantic P-124 (see Fig. 2), but Pratje reports a few measurements that indicate the converse.[b] Moreover, variations in the character of the sediment may produce fluctuations of the weight of solid matter per centimeter cube superimposed on the general trend. Thus Fig. 2 indicates a rough correlation between the weight of solid matter per centimeter cube and the proportion of clay and silt in core P-124.[c]

The determination of the weights of solid material and of water in a section of a recently secured core is easy and accurate, because the technique of core-sampling prevents any dehydration and shrinkage prior to opening the container in the laboratory.[4] Unfortunately, all the cores so far analyzed for radium had been opened and dried for a considerable time and one must resort to an indirect method of arriving at the conversion factor. This method, which is not particularly accurate, consists of tracing the roughly semi-circular cross-sections[d] of both ends of a known length of dried specimen and weighing the specimen. The areas of these tracings are measured with a planimeter and averaged, and the volume of the dried specimen V_d is calculated. The original volume of the

[b] Pratje's cores are from the very shallow basin of the Frisches Haff in East Prussia and are probably exceptions to the rule. Pratje, O.: 1934, *Annalen* Hydrographie u. Maritimen Meteorologie, 62, 129.

[c] Chemical and mechanical analyses of the North Atlantic cores were made on the basis of three fractions: carbonate, sand and gravel $>$ 0.074 mm., clay and silt $<$ 0.074 mm. See reference 3, Plate 3.

[d] The sample and its brass-tube container are cut longitudinally on a diameter which results in two samples of semi-circular cross-section. Piggot, C. S.: Smithsonian Rept. for 1936, pp. 207-216. See, particularly, Plate 5.

specimen in the wet state V_w is known from the inside dimensions of the brass tube and the length between the transverse cracks in the sample. These cracks are located by a mark left by the shrinking material on the brass sample tube. The bulk density of the dried material D_d is computed from the weight W_d and V_d.

The volume shrinkage S is defined by

$$S=(V_w-V_d)/V_w \text{ or } V_d/V_w=(1-S) \quad \ldots\ldots\ldots (1)$$

Replacing V_d by W_d/D_d, the weight of the dried material in a unit volume of the original wet material, ρ, is given by

$$\rho=W_d/V_w=D_d(1-S) \quad \ldots\ldots\ldots\ldots\ldots (2)$$

Figure 2 shows the variation of ρ, expressed in g. per cm.^{3}, with the depth in three cores.

Values of S for core P-124 have been measured elsewhere[3] and only D_d was determined in this laboratory. The shrinkage S for the Blue mud of core P-124 varies from as high as 0.51 in the surface of the ocean bottom to 0.33 at 280 cm. below this surface. Globigerina ooze, represented by cores P-136 and P-137, shows considerably less shrinkage; values of S for P-136 lie between 0.17 and 0.28, while the values for P-137 show a greater spread from S = 0.04 to 0.24 with most of the values around 0.12.

The proportion of the solid matter ρ in the Blue mud increases considerably from top to bottom, whereas in the Glo-

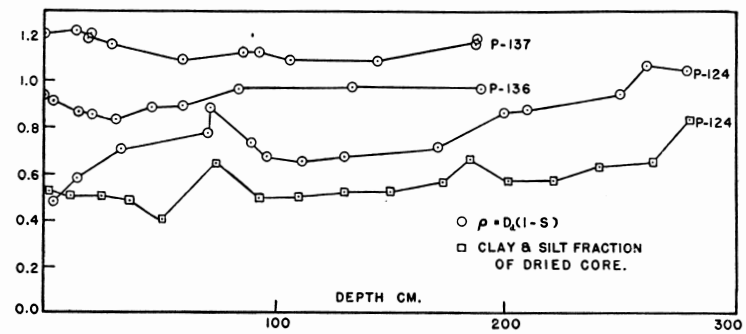

Fig. 2. The weight of dried material, ρ, per unit volume of core sample in the original state, plotted against depth in cores of Globigerina ooze, P-136 and P-137, and in a core of Blue mud, P-124; and the proportion of clay and silt in the core of Blue mud.

bigerina ooze the change is small and the fluctuations less pronounced. In P-137 there is a decrease in the weight of solid matter per unit volume at intermediate depths, and also this weight is always greater than in P-136. An explanation of these differences must await the completion of the lithological and chemical analyses. It should be remembered that while the two cores were obtained at stations only 72 nautical miles apart, there exists the important topographical feature of the Bartlett Deep between them.

The radium content R, per unit volume of the sediment in its original state, is given by

$$R = Ra \cdot \rho \quad \ldots\ldots\ldots\ldots\ldots\ldots (3)$$

where Ra is the radium content per gram of dried material, as given in Table I. Values of R are given in Table II, together with values for P-124 from previously published data[1] and new results. The earlier results[1] for P-124 have been corrected for the apparatus constant determined with the new radium standard. It is difficult to appraise the uncertainty of the values of R, but they are probably accurate to ± 10 per cent.

TABLE II.

The Radium Content of Ocean-Bottom Cores on a Volume Basis.

Core No. P-135 Globigerina ooze			Core No. P-136 Globigerina ooze		
Mean depth cm.	ρ g./cm.^{3}	R Radium in 10^{-12} g./cm.^{3}	Mean depth cm.	ρ g./cm.^{3}	R Radium in 10^{-12} g./cm.^{3}
0.5	1.08	1.03	0.5	0.93	1.33
5.5	1.08	1.40	4.5	0.91	1.81
14.5	1.09	1.74	15.5	0.86	2.21
20.5	1.09	1.99	20.5	0.85	2.32
37	1.05	2.22	31	0.83	2.09
56	1.32	1.65	46.5	0.88	2.03
76.5	1.33	1.65	60.5	0.89	2.03
82.5	1.33	1.60	71.5	0.98	1.97
90.5	1.18	1.59	84.5	0.96	2.04
99.5	1.18	1.55	101	0.97	2.16
109.5	1.05	2.29	115.5	0.92	2.12
			133.5	0.97	1.64
			158.5	1.20	1.50
			189.5	1.20	1.15

Core No. P-137
Globigerina ooze

Mean depth cm.	ρ g./cm.^{3}	R Radium in 10^{-12}g./cm^3
0.5	1.20	1.68
2.5	1.20	1.97
9	1.20	2.65
13.5	1.20	2.81
20	1.19	3.19
29.5	1.15	2.85
60	1.09	2.05
106	1.08	1.31
144.5	1.08	0.97
189	1.17	0.63

Core No. P-124. Depth of water 4700 meters.
Blue mud. Latitude 46° 03′ N. Longitude 43° 23′ W.

Specimen No.	Mean depth cm.	Ra Radium in 10^{-12} g./g.	ρ g./cm.^{3}	R Radium in 10^{-12} g./cm.^{3}
P-124-0	1	1.16 $\pm$ 0.02	0.48	0.56
P-124-25	26.5	1.10 0.01	0.66	0.73
P-124-74	75.5	1.19 0.02	0.87	1.04
P-124-90	91.5	1.57 0.02	0.72	1.13
P-124-130	132	1.68 0.03	0.68	1.14
P-124-171	172	1.79 0.03	0.72	1.29
P-124-207	209	1.78 0.02	0.86	1.53
P-124-257	258	1.34 0.02	1.06	1.42
P-124-283	284	1.39 0.02	1.04	1.45

The shape of the curve in Fig. 1 for core P-137 is not changed appreciably by this conversion of units. The peak is slightly accentuated, and the radium content per unit volume of original material is higher than that per gram of dried core ($\rho > 1$). For core P-136, ρ is less than 1.0, and the radium content, on a volume basis, is lower than that on a dry-weight basis. A plateau is developed below the peak, as shown in Fig. 3. This plateau is probably related to the stratum of pure Globigerina, referred to in the description of the cores, which occurs at 64 to 66 cm. below the top of this core. This feature is also evident in core P-135. It suggests a very rapid deposition at this period in the history of the sediment.

The fluctuations of the radium content per gram of dried Blue mud from the North Atlantic core P-124 largely disappear when the results are reduced to a volume basis (see Fig. 3). The maximum radium content, which is attained on the establishment of equilibrium with the ionium present, is probably reached

near the bottom of the core, which implies a much more rapid deposition in terms of centimeters per year than that of the Globigerina ooze in the Cayman Trough. Rapid deposition of P-124 is also consistent with the fact that the instrument failed to penetrate a cold water zone, which was pierced at relatively shallow depths in all the other cores of the North Atlantic suite.[2] Moreover, the station that yielded core P-124 was just

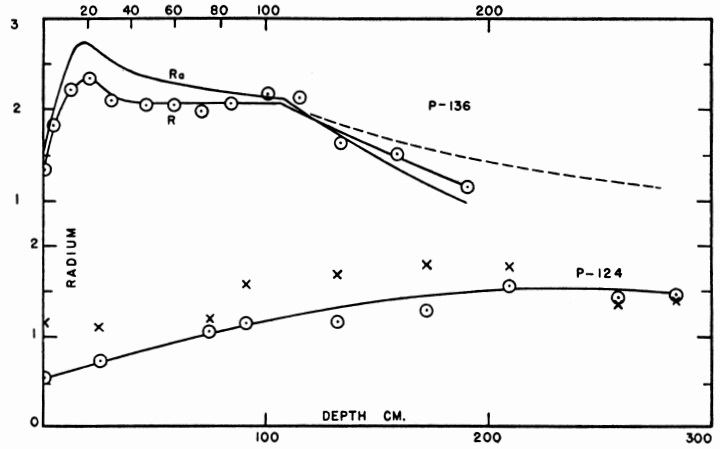

Fig. 3. A comparison of the radium content, R, per unit volume of core sample in the original state, with the radium content, Ra, per gram of dry sample, plotted against the depth in the cores.

Upper curves. Globigerina ooze. Dashed curve is the plot of R, corrected for distortion.

Lower curve. Blue mud. Crosses indicate the values of Ra, and circles the values of R.

over the foot of the Continental Shelf where comparatively high rates of deposition may be expected. In correcting the previous conclusions drawn from the results for P-124, it is now found that an excess of ionium is deposited with the radium, and hence there is no longer any evidence *against* a purely chemical deposition of radium and ionium from the ocean water.[1]

Distortion. Reference has been made to compaction in considering the increase in the proportion of solid material in the sediments with increasing depth. In order to avoid ambiguity, this natural compaction will be called consolidation, and the term distortion will be reserved for the effects produced by the process of sampling. The consolidation may be considered for

the present as being included in the results. The interpretation of clay cores taken by a great many techniques from both land and under-water formations is a current phase of research.[8, 9] Recent experiments with the Piggot core-sampler in the varved-clay pits of East Windsor Hill, near Hartford, Connecticut, demonstrate a non-linear relation between the depth of a given sample in the core and the depth of the same sample in the undisturbed sediment.[11] The equation of this relation might be applied to the ocean-bottom sediments, although so far there exists no satisfactory connection between its parameters and such variables as grain size, water content, mineralogical and chemical characteristics. An application of this distortion correction will have the effect of a progressively increasing extension of the depth axis in Fig. 3. In cores P-135, P-136 and P-137 the shift of the peak-radium values to the right will be negligible. The most important correction will be a reduction in the gradient of the ionium decay curve below the peak as illustrated by the broken line in Fig. 3, which has been computed provisionally on the basis of the quantitative relation found for the varved clays. The curve for P-124 in Fig. 3 will be attenuated without alteration of its general shape. The distortion relations are very important when these results are applied to the determination of the rate of deposition.

Other Results. Among the 28 determinations of the radium in ocean-bottom samples by Hans Pettersson[10] there occur measurements in six short cores collected by the Prince of Monaco on a voyage of the *Princess Alice II.* Four out of the six cores are designated as Red clay and will be referred to in a discussion of our Red-clay analyses. Radium determinations at two and three depths, respectively, in Globigerina ooze are given for Pettersson's cores PA 2868 and PA 1798, the maximum depths being 18 and 23.5 cm. The three determinations in core PA 1798 (south of the Azores) show an increasing radium content with depth similar to the initial steep rise for the cores from the Cayman Trough. The two determinations in core PA 2868 (west of Portugal) show no decided change in the radium content, but there is no record between 2 and 18 cm. Moreover, the samples in both cores represent appreciable fractions of the whole core and vary from 3.5 to 5.5 cm. in length. Apparently, only the shortness of the cores available to Pettersson for his pioneer work prevented his arrival at the conclusions presented in this paper.

If cores can be obtained of such length that the ionium-uranium equilibrium has been attained in their lower portions, the radium analyses will provide further confirmation of the interpretation of the radium relations developed here, will determine the degree of constancy of the uranium concentration, and furnish a value for the uranium content throughout the core. A knowledge of the uranium concentration is necessary in order to calculate the rate of deposition, particularly in the section where the radium content is controlled by the ionium decay. The uranium is now being measured by an independent method.[7]

An effort will be made to obtain longer cores from the more remote and quieter depths of the ocean where deposition may be very much slower than in the regions so far sampled.

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