Report on Naturally Occurring Radionuclides in Human Tissues

by

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Remarks

The present report was prepared to update the former report on naturally occurring radionuclides in human tissues (A/AC 82/G/L.1045) with the addition of newly acquired information and to include some old data which was taken from the 1962 UNSCEAR report for the purpose of comparison. However, only limited data was included here from these old publications because of the following reasons.

Some data is concerned with only total alpha activity. Other with nuclide content in whole body ash. And, some data on nuclide content in bone ash is included again in more recent reports by the same authors.

The information in high natural radiation areas is still scanty because of the difficulty of sampling human tissues in such areas. Several old figures on Ra^{226} in the bones of inhabitants in Kerala, India, showed a much higher value than in normal bones. But there is little data available. (table 1)

The recent data on samples of teeth collected in the Araxá and Tapira region of Brazil, where high radioactive intrusives are found, revealed a tendency toward a higher content of Ra^{226} than in other regions. (table 1) Further, the teeth of two Indian tribes in Brazil that eat Brazil nuts, were found to show a several times higher content of Ra^{226} than in those of the inhabitants of New Jersey which were analyzed at the same time. (table 2)

Pb^{210} levels in bones of Canadian Eskimos who had been permanent residents in the Arctic were found to be considerably higher than the levels in bones of Canadian Eskimos who had been residing in southern Canada for some time prior to removal of the bone. (table 4) This high value is probably due to the special food-chain of lichen-reindeer-Eskimos. The same phenomenon was also found for soft tissues as the result of high Po^{210} content in placenta of Canadian Arctic Eskimos. (table 7)

Attention has been paid recently to the effect of cigarette smoking on the Po^{210} content in human tissues. It was found through research on various parts of lung tissue of smokers that there is a considerably higher level of Po^{210} content than in those of non-smokers. (table 7)

References cited

(1) Hallden, N.A., I.M. Fisenne, J.H. Harley, Radium -226 in human diet and bone. Science 140: 1327-1329 (1963)

(2) Hallden, N.A., J.H. Harley, Ra226 in diet and human bone from San Juan, Puerto Rico. Nature 204: 240-241 (1964)

(3) Holtzman, R.B., Measurement of the natural contents of RaD (Pb^{210}) and RaF (Po^{210}) in human bone- estimates of whole-body burdens. Health Physics 9: 385-400 (1963)

(4) Holtzman, R.B., Pb-210 (RaD) in inhabitants of a Caribbean island. Health Physics 11: 477-480 (1965)

(5) Hursh, J.B., Arvin Lovaas, Radium 226 in bone and soft tissues of man. Nature 198: 265-268 (1963)

(6) Jaworowsky, Z.S., Content of Radium D in human bones and hair. Nukleonika 10: 297-301 (1965)

(7) Osborne, R.V., Lead-210 and Polonium-210 in human tissues. Nature 199: 295 (1963)

(8) Owers, M.J., A. Parker, Radioactivities in human and animal bones. AERE-R-4466, 19p. (1964)

(9) Stahlhofen, W., Measurement of the natural content of Th228, Ra226 and their daughters in the human body. Assessment of Radioactivity in Man. IAEA Vol. II., 505-519 (1964)

(10) Rajewsky, B., W. Stahlhofen, Zur Bestimmung der natürlich vorkommenden alphastrahlenden Nuklide im menschlichen Knochen. Naturwissenschaften 49: 607 (1962)

(11) Rajewsky, B., W. Stahlhofen, Naturally occurring alpha-emitting nuclides in the human body. Nature 198: 960-962 (1963)

(12) Rajewsky, B., V. Belloch-Zimmermann, E. Löhn, W. Stahlhofen, ^{226}Ra in human embryonic tissues, relationship of activity to the stage of pregnancy, measurement of natural ^{226}Ra occurrence in the human placenta. Health Physics 11: 161-169 (1965)

(13) Hill, C.R., Polonium -210 in Man. Nature 208: 423-428 (1965)

(14) Rivera, J. Cesium -137, Stable Strontium and Radium -226 in two human skeletons. HASL-149: 134-137 (1964)

(15) Little, J.B., et al. Polonium -210 in lungs and soft tissues of cigarette smokers. Rad. Res. 22: 209 (Abstract) (1964)

(16) Segall, A., Radiogeology and Population exposure to background Radiation in Northern New England. Science 140: 1337-1339 (1963)

(17) PennaFranca, E. et al. Status of investigations in the Brazilian areas of high natural Radioactivity. Health Physics 11: 699-712 (1965)

(18) Wallace, D.E. Th^{228} and Ra^{226} analysis of bones from Kerala, India. ANL-6398: 67-70 (1961)

(19) Petrow, H.G. et al. Radiochemical analysis of Brazilian samples from regions of high natural radioactivity. NYO-3086-1 Radioactivity Studies Part VI. 1-6 (1965)

(20) Penna Franca, E. et al. Radiochemical and radioecological studies on Brazilian areas of high natural radiation. NYO-3273-6 Vol. I & II. (1965)

(21) Watanabe, H. unpublished

(22) Lucas, H.F. Jr. Correlation of the natural radioactivity of the human body to that of its environment: Uptake and retention of Ra^{226} from food and water. ANL-6297: 55-56 (1961)

(23) Walton, A., R. Kologrivov, J.L. Kulp. The concentration and distribution of radium in the normal human skeleton. Health Physics 1: 409-416 (1959)

(24) Muth, H., B. Rajewsky, H.J. Hantke, et al. The normal radium content and the Ra^{226}/Ca ratio of various foods, drinking water and different organs and tissues of human body. Health Physics 2: 239-245 (1960)

(25) Hill, C.R., Z.S. Jaworowski. Lead -210 in some human and animal tissues. Nature 190: 353-354 (1961)

(26) Hunt, V.R., E.P. Radford Jr., A.I. Segall. Comparison of concentrations of alpha-emitting elements in teeth and bones. Int. J. Rad. Biol. 7: 277-287 (1963)

(27) Lucas, H.F. Jr., R.B. Holtzman, D.C. Dahlin. Radium -226, radium -228, lead -210 and fluorine in persons with osteogenic sarcoma. Science 144: 1573-1575 (1964)

(28) Hill, R.C. Po^{210} content of human placentas in relation to dietary habit. Nature, in press.

Table 1. ^{226}Ra in human bone

LOCALITY	NUMBER OF SAMPLE	pCi/g (10^{-3}) fresh weight	pCi/g (10^{-2}) ash	pCi/g (10^{-2}) Ca	Sample type	Reference
U.S.A. (mostly Illinois)	128		3.7 (0.5 - 28.6)		rib, skull, tibia, vertebrae femur, etc.	(3)
U.S.A. (Chicago residents last 15 yrs. or more)	14		1.5 (0.5 - 2.8)		rib	(3)
U.S.A.						
New York	64			3.2	vertebrae	(1)
San Francisco	71			2.6	"	
U.S.A. (Rochester)	9	3.4 ± 0.4 (1.8 - 4.8)			clavicle	(5)
		1.1 ± 0.3 (0.36 - 2.9)			vertebrae	
U.S.A.	42		1.2 (0.5 - 2.7)		rib	(22)

foot note: (1) ± SE.

(2) figures in brackets indicate the range (Min. - Max.)

^{226}Ra in human bone (continued)

Locality	Number of sample	pCi/g(10^{-3}) fresh weight	pCi/g(10^{-2}) ash	pCi/g(10^{-2}) Ca	Sample type	Reference
U.S.A. (New York City)	140		0.8		total skeleton	(23)
U.S.A. (Wisconsin)	1 1		2.9 1.5		total skeleton	(14)
Puerto Rico (San Juan)	27			1.7	vertebrae	(2)
U.S.A. (New England)			1.4			(26)
U.S.A. (Illinois)	32		2.8 (0.2 - 7.5)			(27)

^{226}Ra in human bone (continued)

LOCALITY	NUMBER OF SAMPLE	pCi/g (10^{-3}) fresh weight	pCi/g (10^{-2}) ash	pCi/g (10^{-2}) Ca	Sample type	Reference
U.K.	1		0.8	2	femur	(8)
	1		0.8	2		
	1		2.0	5		
Germany	22	3.9 (1.8 - 7.8)	1.46 (0.7 - 2.9)		femur tibia	(10)
Germany	25	3.8 ± 1.2	1.4 ± 0.4		femur	(11)
Germany	47	1.30 ± 0.90 (0.10 - 4.77)	1.30 ± 0.60 (0.39 - 3.16)	4.0 ± 2.0 (1.1 - 9.5)	foetal bone, 4-10 months	(12)
Germany	1	5.4	1.2	3	tibia (composite sample of 56 individuals)	(24)
	1	4.8	1.1	5	femur (" 37 individuals)	
Japan age (7-19)	12		0.4 (0.0 - 1.0)		rib, etc.	(21)
(20-70)	27		1.4 (0.2 - 4.8)			

^{226}Ra in human bone (continued)

Locality	Number of samples	pCi/g(10^{-3}) fresh weight	pCi/g(10^{-2}) ash	pCi/g(10^{-2}) Ca	Sample type	Reference
Canada (Vancouver)	7		0.6		single	
Chile (Santiago)	3		3.0		bone ash	(23)
Switzerland (Zurich)	1		1.6			
U.S.A. (Denver)	1		1.1			
Venezuela (Caracas)	4		0.3			
Germany (Koln & Bonn)	5		2.0			
Various countries	15		1.2 (0.4 - 3.6)		composite bone ash	(23)
India (Kerala)	1		7.6		cortical bone	(18)
	1		10.5			
	1		12.7			

Table 2 ^{226}Ra in human teeth

Locality	Number of sample	pCi/g(10^{-2}) ash	pCi/g(10^{-2}) Ca	Reference
U.S.A. (northern New England)	25	1.4		(16)
	15	2.0		
	20	0.9		
	20	1.0		
	20	1.8		
	20	1.6		
	20	1.4		
	20	2.5		
U.S.A. (New Jersey) each sample includes 20 - 40 teeth	Age 1 (0 - 10 yrs)		7.8	(19)
	1 (10 - 20 yrs)		1.2	
	1 (20 - 30 yrs)		0.6	
	1 (30 - 40 yrs)		3.8	
	1 (40 - 50 yrs)		1.7	
	average		3.0	

^{226}Ra in human teeth (continued)

Locality	Number of sample	pCi/g(10^{-2}) ash	pCi/g(10^{-2}) Ca	Reference
Brazil				
Indian, Caiabis	1		14	
Indian, Caneoiro	1		27	(19)
(Brazil nut eater)	average		21	
Brazil				
Guarapari	23	3.6 ± 2.3 (0.6-10.4)		
Meaipe	15	2.3 ± 1.9 (0.6-7.7)		
Vitoria	14	3.0 ± 2.2 (0.8-7.9)		
Rio de Janeiro	13	3.7 ± 2.9 (0.6-12.3)		(17)
Poços de Caldas	13	1.5 ± 0.8 (0.6-3.1)		and
Araxa	24	8.0 ± 5.0 (0.8-20.4)		(20)
Tapira	12	6.0 ± 4.2 (1.8-16.0)		
Ax. and Tp. (high activity area only)	16	8.5 ± 4.8 (2.1-18.8)		

Table 3. ^{228}Ra or ^{228}Th in human bone

Locality	Number of sample	pCi/g(10^{-3}) fresh weight	pCi/g(10^{-2}) ash	pCi/g(10^{-2}) Ca	Sample type	Reference
Germany (Th)	25	1.4 ± 0.5	0.5 ± 0.2		femur	(11)
U.K. (Ra)	1 1 1			1 1 2	femur	(8)
India Kerala (Th)	1 1 1		2.8 1.1 1.1		cortical bone	(18)
U.S.A. (Ra)	32		0.7 (0.2 - 1.9)			(27)

Table 4. ^{210}Pb (RaD) or ^{210}Po (RaF) in human bone

Locality	Number of sample	pCi/g(10^{-2}) fresh weight	pCi/g(10^{-2}) ash	Sample type	Reference
U.S.A. (mostly Illinois)	128		14.6 (3.7 - 45.4) trabecular(67)* 18.4 male (47)* 19.6 female (20)* 15.6 Cortical (61)* 10.5 male (36)* 11.5 female (25)* 9.0	rib, skull, tibia, vertebrae, femur, etc.	(3)
U.S.A. (Chicago residents last 15 yrs. or more)	14		17.7 (6.3 - 35.5)	rib	(3)
U.S.A.	5	(Po) 1.8 (1.2 - 2.7)		lower thracic vertebrae	(15)
U.S.A. (Illinois)	32		8.0 (1.9 - 18.2)		(27)
U.S.A. (New England)	25		14.2		(26)

Foot note: (1) * number of samples

(2) data are ^{210}Pb unless otherwise indicated

Table 4. ^{210}Pb (RaD) or ^{210}Po (RaF) in human bone (continued)

Locality	Number of sample	pCi/g(10^{-2}) fresh weight	pCi/g(10^{-2}) ash	Sample type	Reference
Puerto Rico (Caribbean Island)	28		11.8 (4.9 - 23.7) male (19) 13.3 (6.4-23.7) female (9) 8.8 (4.9-12.6)	vertebrae	(4)
Poland (Warsaw)	20	4.05 (1.52 - 5.9)		vertebrae	(6)
Germany	20	(Pb) 3.2 ± 1.7 (Po) 3.1 ± 1.0	11 ± 4 13 ± 5	femur tibia	(11)
U.K.	9	(Po) 1.7		vertebrae	(7)

^{210}Pb(RaD) or ^{210}Po(RaF) in human bone (continued)

Locality	Number of Sample	pCi/g(10^{-2}) fresh weight	pCi/g(10^{-2}) ash	Sample type	Reference
U.K.	6	2.6 (2.1 - 3.4)		5 vertebrae 1 tibia	(25)
Canada (Eskimos)					
Igloolik, NW Territory		71			
unknown		38			
Carberry, Man.		0.8			
Winnipeg, Man.		1.3			
Edmonton, Alta.		2.7			
unknown		1.2			(13)
Pakatawagan, Man.		3.7			
Nelson House, Man.		4.2			
Nelson House, Man.		4.0			
unknown		10			

Table 5 ^{210}Po in human teeth

Locality	Number of Sample	pCi/g (10^{-2}) ash	Reference
U.S.A. (Northern New England)	25	5.0	
	15	5.7	
	20	5.2	
	20	4.7	
	20	5.6	(16)
	20	6.1	
	20	5.0	
	20	5.9	

Table 6. ^{226}Ra in human soft tissues

Locality	Tissue	Number of sample	pCi/g (10^{-4}) fresh weight	pCi/g (10^{-2}) ash	pCi/g Ca	Reference
Germany	kidney	3	1.0	1.0	0.50	(12)
	testicles	3	0.9	0.9	0.46	
	spleen	3	1.0	0.9	0.98	
	intestine	3	1.0	2.2	0.83	
	liver	3	1.6	1.3	1.60	
	muscle	3	0.5	0.5	0.48	
	pancreas	3	0.7	1.2	0.38	
	average		0.9	1.1	0.75	
Germany	foetal soft tissue (4-10 months)	15	1.1 (0.5-1.9)	1.2 (0.6-2.3)	0.70 (0.31-1.49)	(12)
	Placenta	9	1.6 (0.7-3.1)	1.4 (0.7-2.4)	0.35 (0.21-0.56)	
U.S.A. (Rochester)	liver	9	1.8 ± 1.0			(5)
	skeletal muscle	11	0.5 ± 0.2			
	spleen	9	1.0 ± 0.4			
	kidney	11	1.2 ± 0.3			
	heart	9	0.7 ± 0.2			

PLEASE NOTE

The column at the bottom of page 13 (Po^{210} in placenta) is replaced by the following table.

	Locality		No. of sample	pc/g(10^{-3}) fresh weight	Reference
Canada	Hudson's Bay Coast	(a)	6	114	
	"	(b)	7	115	
"	inland, rural	(a)	2	34.1	
	"	(b)	4	14.4	(28)
"	Yellowknife, N.W.T	(a)	1	24.1	
	"	(b)	1	3.5	
	"	(c)	11	3.6	
U.K.	London	(c)	10	3.3	

Diet Class (a) much reindeer and caribou

(b) some " "

(c) normal diet

Table 7. ^{210}Po in human soft tissues

Locality	Tissues	Number of sample	pCi/g(10^{-3}) fresh weight	Reference
U. K.	liver	4	10	
	kidney	2	7.1	
	spleen	3	3.2	(7)
	lung	2	3.0	
	skeletal muscle	5	1.1	
	testis	4	3.3	
Germany	liver	3	13	
	spleen	3	3	
	muscle	3	2	(9)
	kidney	3	5	
U. S. A.	liver	4	11	(3)
	muscle	2	6	
Canadian Arctic	placenta	9	59.0	
Southern Canada	"	9	5.0	(28)
United Kingdom	"	10	3.3	

^{210}Po in human soft tissues (continued)

Locality	Tissues	Number of sample	pCi/g(10^{-3}) fresh weight	Reference
U. K. (cigarette smoker & non-smoker)	liver	4 (6)	20 (14.8)	(13)
	bronch. tree	4 (6)	7.3 (3.1)	
	alveolae	4 (6)	9.9 (3.4)	
	mean lung	4 (6)	8.6 (3.2)	
	kidney	4 (5)	20.5 (15)	
	gonad	4 (2)	3.9 (2.8)	

figures without brackets are for cigarette smokers and those in brackets are for non-smokers

^{210}Po in human soft tissues (continued)

Locality	Tissue	Number of sample	pCi/g (10^{-3}) fresh weight	Reference
U.S.A.	Peribronchial lymph node (smokers + non-smokers)	17	11 (6-20)	
	lung parenchyma			
	(smokers	12	8 (2-20)	
	(non-smokers	5	1.8 (1-2)	
smokers	(major bronchi	12	28	(15)
smokers	(segmental bronchi	12	53	
smokers	(upper lobe segmental bifurcations	12	189	
smokers	(lower lobe segmental bifurcations	12	318	
	renal cortex	5 (3 smokers + 2 non-smokers)	12 (8-20)	
	liver	5 (")	12 (8-21)	
	spleen	5 (")	2 (1-3)	
	urinary bladder	5 (")	1 (0.5-2)	

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Common Strontium Content of the Human Skeleton

The geochemica and biogeochemical behavior of the dement strontium is important in undestanding the movement of fission-procuct strontium-90 into man (1). Several investigators (2, 3) have analyzed human bone from different locations for srontium. The availability of a large collection of bones from the study of world-wide fallout of strontium-90 made convenient the examination of this parameter in greater detail. This report (4) is concerned with (i) the distributionof strontium among the different bone in an individual skeleton, (ii) the distribution of strontium in the populaion of a single city, and (iii) the extesion of information on geographical vaiation. Samples consisted of a variety of bones from eleven individuals, whole skeleton ash from 133 New York City cadavers, and composites from 16 localitie each representing equal weights of bne ash from 4 to 38 individuals.

The analyses we performed by an emission spectrographic technique modified from that o Turekian and Kulp (2). The standard used to define the working curves we actual samples of bone ash which are analyzed by the isotope dilution method (accurate to within 5 percent) All samples were run in duplicate and re reported as parts of strontium per milion. The reproducibility of these analyses is estimated to be about ± 10 percent.

The average strtium content of additional samples fm previously investigated areas (2) waound to be about 30 percent lower. In cer to check this discrepancy, some of the original samples were reanalyzed by the present method. The new analyses were also about 30 percent lower in ch case. Synthetic standards similar those used by Turekian and Kulp (2) were analyzed, using the present working curve defined by isotope dilution alyses of bone ash. The results indicathat a matrix difference between bs and chemically precipitated phosph is responsible for the higher values rrted in the earlier work (2). In view this observation, the samples of Tur n and Kulp were composited by lety and redetermined.

The distribution of common strontium among the different bones of individuals was examined by analyzing the femur, tibia, fibula, humerus, ulna, radius, hand or foot bones, skull, pelvis-sternum, vertebrae, ribs, clavicle, scapula, and knee-elbow from eleven skeletons (5). Although the average strontium content of the whole skeleton varied by a factor of 3 among these individuals, there was no systematic difference in strontium content between any two bones of the body outside of the experimental error (standard deviation of 10 percent). Thus a single bone can give a valid estimate of the common strontium content of the body at this level of certainty. This would also be the case for strontium-90 distribution if a population ingested a diet with a constant Sr^{90}/Ca ratio throughout the lifetime of the individuals.

Table 1. World survey of common strontium in human bone.

Location	No. of samples	Sr in bone ash	
		ppm	Av
	North America		
Boston	37	101	
Boston	38	109	105
Boston	62*	117	
New York	134*	162	162
Houston	14†	125	152
Houston	12	190	
Denver	33†	203	203
Vancouver	17†	164	144
Vancouver	12	117	
San Juan	5†	179	179
Guatemala	29	156	156
	South America		
Recife	6†	344	344
Guayaquil	17	179	179
Cordoba	18	160	160
Santiago	37†	160	160
Santiago	24	160	
Caracas	37†	187	187
	Europe		
West Germany	30†	137	137
Copenhagen	2†	242	253
Copenhagen	4	256	
Zurich	1†	140	140
Rome	9†	160	206
Rome	10	258	
London	4†	187	160
London	21	156	
	Asia		
Tokyo	36	206	
Tokyo	21*	199	206
Tokyo	5†	203	
Taiwan	19†	191	187
Taiwan	6	179	
India	30†	176	187
India	12	214	
	Africa		
Durban	13	195	195
Liberia	1	324	324
World av			172

* Samples run individually. † Samples reported by Turekian and Kulp (2) rerun as composites.

The histogram (Fig. 1) of the strontium concentration in 133 individuals (whole skeleton ash) from New York City shows a nearly normal distribution with a standard deviation that is only about ± 32 percent of the mean of 162 parts per million by weight. The narrow spread reflects the averaging of food sources in a city environment.

The data on the concentration of strontium in human bone in various geographical localities are summarized in Table 1. To show that the use of composite samples is valid, the samples from Boston and Tokyo were run individually, and then equal weights of bone ash were combined into composite samples. There appear to be small but significant differences from one locality to the next. The average for any given locality falls within a factor of 2 of the mean of the data (172 ppm). Recent work by Sowden and Stitch (6) on a limited number of samples from England analyzed by neutron activation gives results which are consistent within the experimental and natural variation of those reported here. Their work shows a lower strontium concentration in young children. This is expected as a result of fetal discrimination against strontium (7). An examination of the present analyses shows that for adults there is no age effect.

The average world-wide value of $(\%Sr)/(\%Ca) \times 10^3$ in human bone derived from Table 1 is 0.45 ± 0.1. The value $(\%Sr/\%Ca) \times 10^3$ in average rock or soil is 7 ± 1 (8). The discrimination factor between soil and skeleton for the strontium/calcium ratio is therefore 15 ± 2. The experimentally determined discrimination factor for strontium/calcium between soil and plant is about unity (9), between plant and milk, about 7 (10), and between milk or vegetation and human bone it is about 4 (1). Thus, if in the average urban world population, half of the calcium in the diet comes from milk, and half from

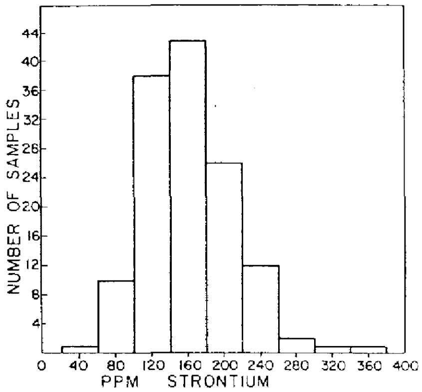

Fig. 1. Histogram of common strontium in ash of whole skeleton from New York City.

vegetables, the predicted over-all discrimination factor would be 16. This figure is in good agreement with the geochemical value of 15 ± 2. If strontium-90 becomes uniformly mixed with the soil, as may occur in tilled fields, this factor will permit prediction of human bone level directly from soil analyses.

The relatively uniform distribution of common strontium in human bone reflects the uniformity in human diet. This observation means that variations in strontium/calcium ratios in different areas will not be an important factor in the distribution of strontium-90 from nuclear tests in the world's population.

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References and Notes

1. W. R. Eckelmann, J. L. Kulp, A. R. Schulert, *Science* 127, 266 (1958).
2. K. K. Turekian and J. L. Kulp, *Science* 124, 405 (1956).
3. R. M. Hodges *et al.*, *J. Biol. Chem.* 185, 518 (1950).
4. Lamont Geological Observatory contribution No. 304. This research was carried out under contract AT(30-1)-1656 between the U.S. Atomic Energy Commission and Columbia University. The criticisms and suggestions of Dr. K. K. Turekian are appreciated. We acknowledge the assistance of Mr. P. Hazlett.
5. A paper describing the details of this work is being prepared by A. R. Schulert, J. L. Kulp and E. Hodges.
6. E. M. Sowden and S. R. Stitch, *Biochem. J.* 67, 104 (1957).
7. C. L. Comar *et al.*, *Proc. Soc. Exptl. Biol. Med.* 88, 232 (1955).
8. K. K. Turekian and J. L. Kulp, *Geochim. et Cosmochim. Acta* 10, 245 (1956).
9. R. Menzel, private communication.
10. W. J. Visek *et al.*, *J. Dairy Sci.* 35, 783 (1952).

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