A Symposium on

RADIOISOTOPES IN THE BIOSPHERE

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PREFACE

A symposium on the ecology of radioisotopes was held at the University of Minnesota from October 19-23, 1959. In addition to the University, the sponsors of the meeting were the National Science Foundation, the Atomic Energy Commission, and the Agricultural Research Service, USDA.

The demonstration that the debris from atomic explosions and from the peaceful uses of atomic energy included radioisotopes that were, biologically, of consequence to man has resulted in studies on numerous aspects of their presence in the biosphere. In this regard, the symposium was concerned with a discussion of the facts and deficiencies in our knowledge relating to the pathway of radioisotopes from the time they enter soils until they are excreted from living systems. Particular emphasis was placed on the uptake of radioisotopes by plants, animals, and man and the genetic consequences of their ingestion.

In planning the symposium, numerous members of the University faculty gave generously of their time. In addition, it is a pleasure to acknowledge the valuable contributions of the following section chairmen: Dr. E. R. Graham and Dr. T. C. Broyer, Section 1; Dr. E. L. Powers, Section 2; Dr. Bernard H. Trum, Section 3; Dr. Wallace D. Armstrong, Section 4; Dr. Joseph H. Ross, Section 5; and Dr. William P. Norris, Section 6.

April 25, 1960

Symposium Organizing Committee

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Section I

RADIOISOTOPES IN SOILS AND PLANTS

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Radioisotopes in Soils: Particularly with Reference to Strontium[90]

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Radioisotopes have been recognized constituents of soils for many years. In particular K^{40}, Rb^{87}, C^{14}, Th^{232}, U^{238} and the associated daughter products of the latter two, may be found in a great many soils throughout the world. Radium[226], which is a daughter product found in the uranium series, was given attention as a soil element by Moore (1914).

These naturally occurring radioisotopes are present in soil because their half-lives are comparable to the age of the earth or because they are continually being produced by the decay of unstable parent nuclei or by nuclear reactions from cosmic rays. The types of deposits in which these radioisotopes occur therefore, differ considerably. Thorium is abundant as a rock component, but uranium is found in a much greater variety of chemical combinations. This fact is responsible for the major differences in distribution that do exist.

For reference we have listed in table 1 (Dudley, 1957) known naturally occurring radioisotopes of half-life greater than a few days, and in table 2 (Dudley, 1957) the abundance of the corresponding naturally occurring inert elements appear. It is interesting as a basis for later comparisons with man-made radioisotopes to estimate the abundance of radioactive K^{40} in soil. Using table 1, the isotopic abundance of K^{40} in natural potassium is, expressed in activity units, about 8.4×10^{-4} μc/g potassium. Converting to per cent abundance, K^{40} is about 0.01 per cent of natural potassium. From table 2, the abundance of inert potassium in igneous rock in the earth's crust is about 2.6 per cent of the rock by weight. Potassium[40] then, would be about 0.026 per cent of the rock by weight. Or, returning to activity units, the activity of K^{40} in igneous rock would be about 0.032 μc/g of rock. Let us go on to a hypothetical calculation assuming that we have a soil containing K^{40} of the abundance just computed. Very roughly, a 1-foot depth of soil runs about 35

TABLE 1. Data on naturally occurring radioisotopes

Isotope	Long-lived ancestor	Half-life	Particulate radiation	Average energy (Mev)	Natural activity (μc/g)	Comment
C^{14}	.....	5.6×10^{3} yr	beta	0.05	7×10^{-6}	Activity of living carbon
K^{40}	.....	1.2×10^{9} yr	beta	0.4	8.4×10^{-4}	(beta activity)
					0.9×10^{-4}	(gamma activity)
Rb^{87}	.....	6×10^{10} yr	beta	0.1	1.9×10^{-2}	
La^{138}	.....	2×10^{11} yr	beta	0.3	7×10^{-7}	
Sm^{147}	.....	1.4×10^{11} yr	alpha	2.2	2.6×10^{-3}	
Lu^{176}	.....	7×10^{10} yr	beta	0.1	8×10^{-4}	
Re^{187}	.....	4×10^{12} yr	beta	0.1	3×10^{-4}	
Pb^{210} (RaD)	U^{238}	22 yr	beta	0.006	8×10^{-2}	Natural activity was computed from average Pb and U concentration in earth's crust, and is therefore not applicable to Pb ore deposites
			(beta	0.3)	(average)	
			(alpha	5.3)		
			(decay products	)		
Pb^{212} (ThB)	Th^{232}	10.6 hr	beta	0.1	8×10^{-2}	Natural activity was computed from average Pb and Th concentration in earth's crust, and is therefore not applicable to Pb ore deposites
			(beta	0.7)	(average)	
			(alpha	8.9)		
			(decay products	)		
Po^{210}	U^{238}	140 d	alpha	5.3	.....	No long-lived isotope. Therefore Po^{210} found carrier free in equilibrium with U^{238}
Rn^{222}	U^{238}	3.8 d	alpha	5.5	.....	
			(beta	1.5)		
			(alpha	19.0)		
			(decay products	)		
Ra^{226}	U^{238}	1600 yr	alpha	4.8	1.0×10^{6}	
			(beta	1.5)		
			(alpha	24.5)		
			(decay products	)		

TABLE 1 (continued)

Isotope	Long-lived ancestor	Half-life	Particulate radiation	Average energy (Mev)	Natural activity (μc/g)	Comment
Ra^{228}	Th^{232}	6.7 yr	beta (beta (alpha (decay products	0.01 0.5) 32))	1.0×10^6 (average)	Natural activity based on abundance of Ra element and is therefore only an average value
Ac^{227}	U^{235}	22 yr	beta (beta (alpha (decay products	0.01 0.5) 32))	.....	No long lived isotope
Th^{232}	.....	1.4×10^{10} yr	alpha (beta (alpha (decay products	4.0 1.3) 32))	0.11	
Th^{228} (RdTh)	Th^{232}	1.9 yr	alpha (beta (alpha (decay products	6.4 1.3) 27))	0.11	Natural specific activity based on abundance of Th^{232}
Th^{230}	U^{238}	8×10^4 yr	alpha	4.7	0.1 (average)	Natural specific activity based on abundance of Th^{232} and is therefore an average value
U^{233}	.....	4.5×10^9 yr	alpha	4.20	0.335	
U^{234}	U^{238}	2.5×10^5 yr	alpha	4.76	0.335	Natural specific activity based on abundance of U^{238}
U^{235}	.....	7.1×10^8 yr	alpha	4.4	0.015	

TABLE 2. Abundance of elements in various rocks (ppm)

Element	Earth's crust Igneous	Sedimentary		
		Sandstone	Shale	Limestone
K	2.6×10^4	1.1×10^4	2.7×10^4	2.7×10^3
Ca	3.6×10^4	3.9×10^4	2.2×10^4	3.0×10^5
Rb	3.1×10^2	2.7×10^2	3×10^2	0
Sr	3×10^2	26	1.7×10^2	$4 \times 10^2 - 8 \times 10^2$
Ba	2.5×10^2	1.7×10^2	4.6×10^2	1.2×10^2
La	18	.....	.....	.....
Sm	6	.....	.....	.....
Lu	0.7	.....	.....	.....
Re	10^{-3}	.....	.....	.....
Pb	16	20	20	5 - 10
Th	12	6	10	1
U	4	1.2	1.2	1.3

kilograms per square foot. On an area basis, therefore, the above K^{40} abundance amounts to 21,300 millicuries per square mile.

For Ra^{226}, using radium assay data in soils (Lowder, 1956) one can estimate that between 80 and 800 millicuries per square mile can be found in the surface foot of various United States soils.

There are interesting possibilities for study of the naturally occurring radioisotopes in soil. For example, the use of soil carbon as a basis for dating has hardly been explored. A few measurements made on some of our grassland soils indicate carbon ages of from a few hundred to about 1,000 years.

Although, as will appear in the coming discussion, the activity levels of man-made isotopes such as Cs^{137}, Sr^{90}, Zr^{95}, etc. are far lower in soils than that of some of the natural radioisotopes, our effort has been strongly fixed on these man-made isotopes for two reasons: first, because activity level alone is not a valid measure of potential biological hazard, and second, because nation-wide concern has been focused on the hazard to man from such man-made isotopes as occur in radioactive fallout. This paper will henceforth discuss in detail, the purpose and results of the work on environmental Sr^{90} levels and distribution patterns over large areas, and will include as well, some measurements on gamma emitters in fallout.

CRITERIA FOR SAMPLING AND MEASURING STRONTIUM^{90} IN SOILS

In our world-wide fallout program, there are two basic reasons for measuring Sr^{90} in soils. In the first place one wants to relate levels in soils to levels in plants, animals, human foods and human beings. Secondly, one wants to relate levels in soils to amounts produced in weapons testing and to amounts existing elsewhere in the environment in order to determine a world-wide material balance. It is clear that there is a difference in the approach to sampling and

analysis depending upon which goal is in mind. Thus for soil sampling in relation to up-take, one must be assured that the samples represent what the plants "see" and, similarly, that the analytical methods do not measure Sr^{90} that would be completely unavailable to a plant.

For soil sampling to determine total deposition of Sr^{90}, the objective is to obtain a soil sample from an area having all the Sr^{90} that ever fell out on it but no other. It is to be remembered that rainfall is undoubtedly the principle mechanism that brings Sr^{90} to the ground from the air. Hence, a suitable spot would be level and sufficiently permeable to absorb all of the precipitation that falls on it. But it must not receive water from higher ground by run-off. It should have base exchange capacity adequate to keep the Sr^{90} ions from being readily leached into the ground water. The area should not, of course, be sheltered by buildings or trees. A good grass turf aids in absorption of the water and reduces the likelihood of run-off.

The extent to which vegetation enhances the removal of Sr^{90} from the air is not known. It is our opinion that the Sr^{90} fallout on a bare ground surface is less than on an area covered by a dense stand of alfalfa, for example. In our work we have standardized on short-cropped grass sods as the most suitable sampling areas. Here is a subject for which our knowledge is scant and research work is clearly needed for studying the effect of different kinds and densities of vegetation with respect to impaction or screening of Sr^{90} deposition from the air.

World-wide Sampling Programs for Strontium^{90}

Limited soil sampling activities began in 1953. Since then, the work has expanded greatly to the point where we now sample regularly over the United States and over much of the free world's land area. For special studies of, for instance, the relationship of Sr^{90} deposition to rainfall, more intensive sampling in selected regions has been done.

The following are the soil collection and analysis programs which have been carried out recently by the U. S. Department of Agriculture and the Atomic Energy Commission (Alexander, 1959).

1. 1958 Outside U. S. soils (62 localities, 78 sites).
2. 1958 United States soils (17 localities, 23 sites).
3. 1958 Special soil sampling along a 32 inch rainfall contour in central United States (18 localities).
4. 1957 Special soil sampling along a 40 inch rainfall contour on the west coast of the United States and Canada (11 localities).

Results of Strontium^{90} Soil Sampling

The 1958 sampling provided us with a much better estimate than we had previously of what errors might be involved in the sample-taking and the

sample processing and radiochemical analysis. A study of the total Sr^{90} in a number of paired sites (that is, localities sampled in duplicate) in 1958 in various parts of the world outside the United States, showed an average of about 1.6 millicuries per square mile difference between the two sites at any one locality. This was for sites from one to twenty-five miles apart but still sampled as one locality (table 3). Examination of data from six Health and Safety Laboratory localities in the United States sampled in duplicate in 1958 showed an average site difference of 4.8 mc of Sr^{90} per square mile (table 4). The analytical duplicate error was 1.1 mc per square mile. Including both sample replication and analytical error the difference between the paired sites is about 10 to 12 per cent. This result comes in spite of the fact that for these United States locations the second site in each locality was originally selected because of some doubts regarding the validity of the original sampling site.

The world-wide data for soils (figures 1, 2) show a strong peaking of the Sr^{90} deposition in the north temperate latitudes, a low deposition in equatorial regions and a moderate increase from the equator southward to the middle southern latitudes. The contribution of an abnormally high rainfall site in Oahu, Hawaii is shown as a blank or non-cross hatched bar on the graphs and is considered to be an anomalous value since high rainfall areas were not sampled in other latitude belts. Conversely, the fallout at Lima, Peru was so low that considerable doubt is cast on the analytical values. Again the contribution of this site is shown by a blank bar on the graphs but the value is considered an anomalous one. The low amounts found in the area from 70° to 80° north latitude (and presumably for southern latitudes) are largely due to the fact that these areas are very low in total precipitation. In the very cold, very dry northern areas such as found on Ellesmere Island and Cornwallis Island, vegetation is sparse and in small clumps only. The areas in between the vegetation are bare. The soil materials blow and shift about with the winds. In the winter, snow blows from such areas into the clumps of vegetation and also to distant drifts. The clumps accumulate winter precipitation. They represent abnormal accumulations of Sr^{90} rather than the fallout that actually was deposited per square mile. The 2 sites on Cornwallis Island differ from each other by more than 2-fold. Both are high in relation to the sites at Point Barrow, Alaska where there is a complete tundra cover. Perhaps there is not a satisfactory way of sampling the partially vegetated, wind-swept areas. The contribution of the data from Ellesmere and Cornwallis Island is shown as the non-cross hatched section on the bar graph.

A similar situation exists in our southwestern desert areas. Between the clumps of sagebrush and greasewood the soil surface horizon is in frequent motion due to wind-action. Blown materials accumulate in the vegetation at heights greater than those of bare areas. Heavy rains wash some of the soil material in the clumps back into the lower-lying bare spots. Larson (1958) found several-fold differences between replicate samples taken in these virgin areas near the Nevada Test Site. This kind of local variation is to be expected

TABLE 3. Replication—1958 soils collected by L. T. Alexander[a,b]

Location	Distance between sites (mi)	Site A Depth (in.)	Aliquot[c]	mc Sr^{90}/mi^2 (av)		Site B Depth (in.)	Aliquot[c]	mc Sr^{90}/mi^2 (av)	
Japan, Tokyo	3	0-7	a	30.82	30.42	0-7	a	23.47	24.62
			b	30.03			b	25.76	
Philippines, Manila	4	0-6	a	9.24	8.80	0-6	a	7.10	6.70
			b	8.37			b	6.30	
Un. of So. Africa, Durban	2	0-6	a	7.50	6.93	0-6	a	6.23	6.90
			b	6.36			b	7.58	
Canal Zone, Panama	5	0-6	a	6.69	7.11	0-6	a	8.57	9.01
			b	7.53			b	9.45	
Singapore	2	0-6	a	4.20	4.06	0-6	a	4.22	4.18
			b	3.93			b	4.15	
Belgian Congo, Leopoldville	1	0-6	a	5.90	6.02	0-6	a	4.80	4.90
			b	6.15			b	5.01	
Brazil, Belem	4	0-6	a	6.24	6.45	0-6	a	7.25	6.66
			b	6.66			b	6.07	
Southern Rhodesia, Salisbury	1	0-6	a	5.04	4.94	0-6	a	5.24	4.90
			b	4.84			b	4.55	
North Dakota, Mandan	15	0-6	a	44.80	44.80	0-6	a	40.20	40.20
			b	44.80			b	[d]	
Alaska, Barrow	3	0-6	a	4.29	4.29	0-6	a	3.52	3.52
			b	[d]			b	[d]	
Hawaii, Oahu	25	0-2	a	21.15	21.92	0-2	a	22.04	22.14
			b	22.69			b	22.24	
		2-6	a	10.03	9.94	2-6	a	11.51	11.04
			b	9.84			b	10.56	
		Total			31.86	Total			33.18

[a] Average difference between analytical duplicates—0.72 mc/mi².
[b] Average difference between means of site duplicates—1.64 mc/mi².
[c] a and b are analytical duplicates on separate soil aliquots from same soil sample.
[d] Duplicate not analyzed.