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PRELIMINARY STUDIES OF THE PERSISTENCE OF TRITIUM AND ^{14}C IN THE PACIFIC PROVING GROUND*

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(*Presented by* J. J. KORANDA)

Abstract—The results of a preliminary survey of detonation environments in the Pacific Proving Grounds indicate that residual tritium and ^{14}C are present in relatively high concentrations in soil materials of the detonation sites at times up to 12 years after the event. Exchange of soil-bound tritium with the available soil water takes place at a slow but significant rate and tritium is detectable in plants growing in the detonation environments. ^{14}C is also elevated in the terrestrial plants. The basis for the elevated ^{14}C is not implicit in these preliminary data. Tritium and ^{14}C are also present in elevated concentrations in marine organisms. However, due to the high rate of exchange of the lagoon waters with the open sea, these elevated concentrations are highly localized in the vicinity of the detonation site (Mike Crater).

INTRODUCTION

THE FATE of residual radioactivity from nuclear detonations in natural environments has been studied with increasing intensity since the beginning of testing in the S.W. Pacific in 1946. Much of the biological information on the local effects of residual radioactivity is about either the atoll environments of the Pacific Proving Grounds in the Marshall Islands or the desert areas of the Nevada Test Site. Much of this information pertains to fission products and their movement in soils, plants and animals of the detonation environment although some recent research has been concerned with induced radioactivities.

With the advent of thermonuclear devices and the subsequent lowering of the fission–fusion ratios, induced radioactivities have grown in importance relative to fission products. Tritium and ^{14}C represent a significant portion of the residual radioactivity from a thermonuclear detonation. Hydrogen and carbon are ubiquitous elements in the biosphere and are included in every significant biological molecule. Tritium and ^{14}C, therefore, will be incorporated into the basic structures of organisms in the detonation environment.

While tritium and ^{14}C have been detected and studied in worldwide fallout, the fate of these two radioelements in local detonation environments and in the biosphere in general has received little attention. Recent evidence from the Sedan cratering detonation in southern Nevada indicates that tritium is present in high concentration in the geological materials displaced and modified by the detonation 2 years after the detonation. Therefore, a study was initiated to investigate the persistence of tritium and ^{14}C in detonation environments. This paper presents the results of a preliminary survey for tritium and ^{14}C in an atoll environment of the Pacific Proving Grounds. Detonations occurred from 6 to 12 years ago in the sites sampled, and tropical vegetation has reinvaded most of the areas affected. Tritium and ^{14}C activity was determined in plants, animals and soils from a variety of habitats. The effects of indigenous substratum, climatology, and uptake by the biota on the persistence of tritium and ^{14}C are discussed.

Evidence indicates that both tritium and ^{14}C have been retained by the physical materials in the detonation environment in relatively high

* This work performed under the auspices of the U.S. Atomic Energy Commission.

concentrations, and that they are presently detectable at elevated levels in the biota.

DESCRIPTION OF THE STUDY

Eniwetok Atoll is in the northern portion of the Marshall Islands in the S.W. Pacific Ocean (11°30′ N lat, 162°15 E long) and was a proving ground for nuclear device testing from 1948 to 1958 when the last detonations took place. The detonations occurred in various environmental situations within or near the atoll, but for the purposes of surveying for residual tritium and ^{14}C, the crater-forming shots were considered most appropriate. Preliminary studies of a large cratering detonation at the Nevada Test Site have indicated that a significant fraction of the residual tritium produced becomes associated with the geological materials in and displaced by the explosion.[1]

The detonations which produced a distinct crater at Eniwetok Atoll were few (many shots being made from barges within the atoll lagoon), but two areas were selected for survey on the basis of their physical and biological characteristics. Cactus Shot (1958 Hardtack Series) produced a terrestrial crater at the north end of Runit Island on the eastern edge of the atoll. The crater has a distinct lip and a throwout zone of mounded coral sand and debris. Vascular plants have become reestablished on the crater materials, and trees of *Scaevola frutescens* and *Messerschmidia argentea* with diameters up to 4 in. at the crown are growing within 15 ft of the crater lip. Mike Shot (1952 Ivy Series) created a large submarine crater adjacent to Sanildefonso Island. The crater is actually in the coral reef that forms the oval platform on which the islands of the atoll have formed. The crater has partially filled with coral sand since the shot date.

Other islands in the atoll not involved directly in any nuclear detonation (but which undoubtedly received close-in fallout of large particles) were also sampled to provide information on the general tritium background of the atoll. Igurin Island at the southern end of the atoll and Japtan Island on the eastern edge were islands farthest from the detonation sites which were sampled in this series. Engebi Island at the northeast corner of the atoll was also sampled. This island was adjacent to many barge shot sites in the 1958 series, and in previous series tower shots were fired on this island. Collections were also made on Aomon, Biijiri and Aaranbiru Islands on the eastern edge of the atoll. These islands were adjacent to shot areas and received close-in fallout from barge shots in the lagoon. Aomon Island had a 49-kton tower shot in 1948. A map of Eniwetok Atoll with the island names and crater locations is given in Fig. 1.

Sampling procedure

This survey is intended to be an initial examination of the Eniwetok environment to determine if tritium and ^{14}C are present in biological and soil materials at times up to 12 y after detonation. Because of the preliminary nature of the survey, it was desirable to collect a wide variety of materials without acquiring excessively large numbers of samples for analysis. In some cases a sample series may be incomplete because of sample number limitations or its unavailability at the time of collection. In spite of some shortcomings and the preliminary nature of this survey, the data enable us to describe in some detail the persistence of tritium and ^{14}C in the physical and biological materials of the atoll.

The samples were collected on eight islands on Eniwetok Atoll during July and August 1964. This was the beginning of the rainy season in the northern Marshall Islands. The University of Washington, Laboratory of Radiation Biology scientific staff was conducting a resurvey of Eniwetok and Bikini Atolls at this time and assisted in collecting the biological specimens. It would not have been possible to obtain the wide range of samples reported in this study without their valuable assistance.

At Japtan, Runit and Engebi Islands, wood, green leaves, litter and a soil sample were collected as an ecological series from the same sample site. A wood sample from the south end of Runit Island was compared with the sample from the north end at Cactus Crater. Wood samples were also obtained from four other islands.

Two plant species were often present in the habitats of the detonation sites. These were *Triumfetta procumbens*, a prostrate vine in the *Tiliaceae*, and *Lepturus repens*, a short grass. These species were collected on Runit Island adjacent

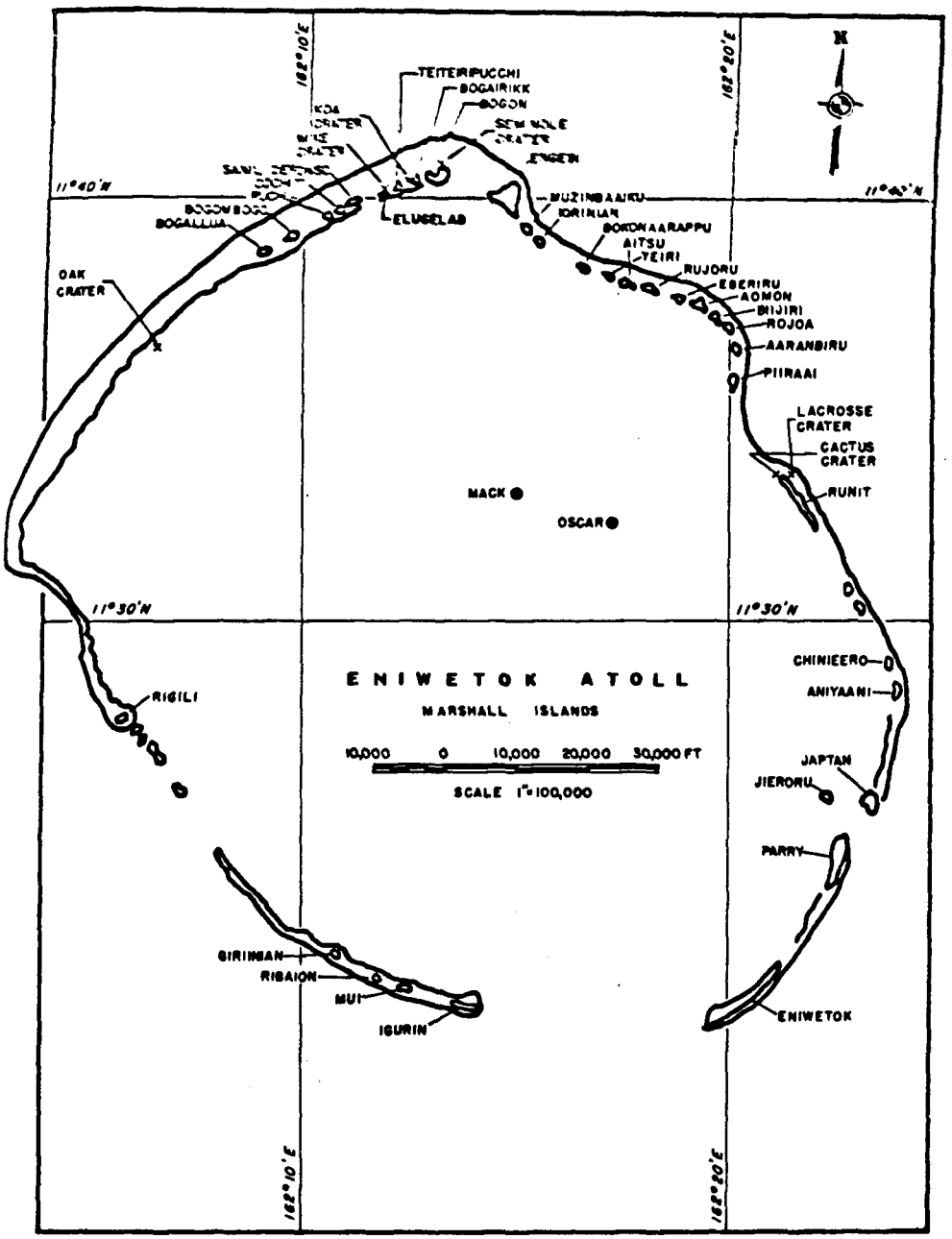

FIG. 1. Map of Eniwetok Atoll.

to Cactus Crater and on Sanildefonso Island near Mike Crater.

Marine organisms were collected at three locations within the lagoon. Fishes were obtained near Igurin Island, Runit Island and off Sanildefonso Island at the edge of Mike Crater. Small killer clams were collected near Runit Island on the reef. Two types of land crabs were obtained: the coconut crab, *Birgus latro*, on Igurin Island and hermit crabs, *Coenobita perlatus*, on Sanildefonso Island.

Seawater samples were obtained at two locations in the lagoon. A surface sample (−10 ft) and a deep-water sample (−180 ft) were obtained from near the deepest part of the lagoon, using a polyvinylchloride water bottle. A seawater sample was also collected 1 m from the bottom of Mike Crater. Rainwater was collected in plastic trays on Eniwetok Island in August.

The taxonomy of the plants considered in this study is based on ST. JOHN.[2]

Analytical procedures

All of the biological samples were analyzed for loose-water and tissue-bound tritium. The loose water of the plant and animal tissues and the soil samples was extracted by freeze drying at 0.1 μ of vacuum. RANEY[3] demonstrated that freeze-drying plant tissues was equivalent to oven-drying at 105°C for 24 h. Loose water of plants extracted by cryo-sublimation represents all interstitial water, vacuolar and protoplasmic water, and loosely held water of hydration in the cell walls. Because of the various complex states in which water is held in plant cells, it is probably more realistic to consider this water as *cryo-sublimed water* than to attempt to describe its exact physiological nature.

The loose water obtained by freeze-drying soils is all of the capillary water and most of the hygroscopic water. Hygroscopic water is removed from soils by oven drying for 8–10 h at 100–110°C. Plants are generally able to extract water from soils at tensions up to 15 atm or about 50 μ of vacuum. Therefore, the water freeze dried from soils (and defined here as loose water) may represent more than that actually available to the plant.

The tissue-bound water of the animal and plant tissues, and the mineral-bound water of the soil particles (in this case, almost $CaCO_3$ and $Ca(OH)_2$) were obtained by ... ing the freeze-dry residues in a ... containing copper oxide at 600–800°C. ... is collected from the effluent gas by a freeze ... and CO_2 by bubbling the effluent gas thr... KOH. The extracted water was then anal... for tritium by either gas-counting or scintill... methods. The tritium and ^{14}C determina... were performed under contract by Isotopes I... of Westwood, N.J.

The analytical results are expressed in Trit... Units (T.U.). A Tritium Unit is defined as ... atom of tritium per 10^{18} atoms of hydrogen a... therefore, is an expression of specific activi... The tritium content of the samples will b... reported in T.U. for loose- and tissue- ... mineral-bound water, and the ^{14}C is expressed ... the percentage of ^{14}C above that of wood (fro... the year 1890) which is equivalent to 0.95 of th... National Bureau of Standards oxalic acid ^{14}C standard.

RESULTS

The analytical results of the plant and so... samples from seven islands on Eniwetok At... are given in Tables 1, 2 and 3. Data in thes... tables are presented as an ecological series representative of the habitats being studied. The wood, leaves, litter and soil from a single sampling station make up an ecological series. Table 1 contains the tritium values of plants and a soil collected at Cactus Crater on Runit Island. Table 2 lists the tritium content of plants and a soil sample from Engebi and Sanildefonso Island, adjacent to Mike Crater. Table 3 contains the tritium values for plants and soils from those islands which were not intimately associated with the Cactus or Mike Shots. These samples were mainly wood, but a complete ecological series was collected on Japtan Island.

Duplicate samples were also analyzed to provide an internal check on the analytical methods and to determine the variation present within a single sample collection. A duplicate sample of water from the bottom of Mike Crater was reanalyzed 1 month after the first analysis, and the sample was stored in glass between analyses. Four duplicate wood samples were analyzed. The wood samples had been electrically sealed in double polyethylene bags (6 mil) and kept at below freezing temperatures between

Table 1. *Tritium content of samples collected on Runit Island at Cactus Crater* (in T.U.)

Sample	Wood		Leaves		Litter		Soil	
	Loose water	Bound water	Loose water	Bound water	Loose water	Bound water	Loose water	Bound water
Messerschmidia argentea, inner wood	2970 ± 50	4600 ± 140	442 ± 27	1070 ± 45	152 ± 17	1800 ± 80	509 ± 30	3.54 ± 0.10 × 10⁶
outer wood	3230 ± 60	2740 ± 100						
Messerschmidia argentea,*	410 ± 40	620 ± 40						
Messerschmidia argentea, S. end of Runit)	≤70	220 ± 26						
Scaevola frutescens	540 ± 32	1360 ± 40	119 ± 20	750 ± 50	59 ± 19	7100 ± 250†		
Lepturus repens			265 ± 28	2840 ± 90				
Triumfetta procumbens			150 ± 28	600 ± 40				

* (second sample one month later).
† Second analysis—10,000 ± 300 T.U.

Table 2. *Tritium content of samples collected on Engebi and Sanildefonso Islands* (in T.U.)

Samples	Wood		Leaves		Litter		Soil	
	Loose water	Bound water	Loose water	Bound water	Loose water	Bound water	Loose water	Bound water
Messerschmidia argentea, Engebi Is.	65	170 ± 20	60	530 ± 60	60	160 ± 30	60	3.68 ± .10 × 10⁵
Scaevola frutescens, Engebi Is.	120 ± 15	170 ± 20	60	560 ± 40	60	1960 ± 60		
Messerschmidia argentea, Sanildefonso Is.	80 ± 30	200 ± 40	60	180 ± 60				
Lepturus repens, Sanildefonso Is,			60				60	7.50 ± .05 × 10⁴
Triumfetta procumbens, Sanildefonso Is.			70	180 ± 30				

Table 3. Tritium content of samples from Igurin, Japtan, Biijiri and Aaranbiru Islands (in T.U.)

Samples	Wood		Leaves		Litter		Soil	
	Loose water	Bound water	Loose water	Bound water	Loose water	Bound water	Loose water	Bound water
Messerschmidia argentea, Igurin	118 ± 22	190 ± 24					50	3.00 ± .10 ×
Pisonia grandis, Igurin Is.	62 ± 26	391 ± 28						
Cocos nucifera, old nut, Igurin	990 ± 34	(Husk) 740 ± 90						
young nut, same tree	80 ± 40	150 ± 30	(Meat) 160 ± 30	84 ± 24				
Messerschmidia argentea, Japtan	147 ± 26	240 ± 34						
Pisonia grandis, Japtan Is.	80	240 ± 30	116 ± 19	330 ± 40	(young) 132 ± 23	300 ± 30	50 ± 25	3.44 ± 0.10 × 10
					(old) 61 ± 15	277 ± 32		
Ochrosia oppositifolia Japtan Is.	76 ± 26	160 ± 30						
Cocos nucifera, young nut, Japtan	40	(Husk) 580 ± 40	(Meat) 330 ± 20	70				
Guettarda speciosa, Aaranbiru Is.	75 ± 30	70 ± 40						
Pisonia grandis, Aaranbiru Is.	130 ± 40	40						
Messerschmidia argentea, Biijiri	69 ± 15	170 ± 20						

analyses. Duplicate sample results are given in Table 4.

Table 5 presents the tritium content of water samples collected at Eniwetok Atoll. The tritium content of a late 1963 seawater sample from the Marshall Islands was reported by the IAEA[4] and is included in this table.

The tritium content of animals collected at four locations on Eniwetok Atoll is given in Table 6. ^{14}C values are also included in this table.

In order to characterize the soil-bound source of tritium and ^{14}C, the values for five soils are given separately in Table 7, and have appeared in the tables with the ecological series. Tables 8 and 9 contain the ^{14}C values for plant samples collected on Eniwetok Atoll and are arranged in the same ecological sequence that was used in Tables 1 through 3.

DISCUSSION

Background information

The mode of entry of water into plants is mainly through absorption of water from the soil by roots. The mechanism involves both active and passive absorption although the passive route is the most important in satisfying the water requirements of the plant. Large amounts of water are absorbed each day by the root systems of plants, transported through the stems, and released to the atmosphere by transpiration. Most of the water absorbed is lost in transpiration, and less than 1 per cent will be used in the synthesis of new organic matter.[5]

Table 4. Duplicate samples

Sample	First analysis (T.U.) Loose water	First analysis (T.U.) Bound water	Second analysis (T.U.) Loose water	Second analysis (T.U.) Bound water
Seawater, Mike Crater	820 ± 40		840 ± 40	
Livermore tapwater*	1410 ± 50		1460 ± 70	
Messerschmidia argentea, wood, Cactus Crater	2970 ± 50	3350 ± 50	3100 ± 100	6100 ± 200†
Messerschmidia argentea, wood, second sample from Cactus Crater area	410 ± 40	620 ± 40	330 ± 32	500 ± 50
Messerschmidia argentea, S. end of Runit Island	≤70	220 ± 26	≤60	180 ± 24
Messerschmidia argentea, Engebi Island.	≤65	170 ± 20	270 ± 32	180 ± 22

* Livermore tapwater (Hetch Hetchy Aqueduct water which represents surface runoff of precipitation) was utilized for duplicate sample analysis because of its ready availability. This tritium content is within the range of values occurring in North American precipitation since the beginning of nuclear testing.

† Two subsequent analyses yielded values of 3100 ± 50 and 2960 ± 50 T.U.

Table 5. Tritium content of rain and seawater at Eniwetok Atoll

Sample	Location	T.U.
Lagoon seawater	10 ft below surface near center of lagoon	≤60
Lagoon seawater	180 ft below surface at bottom of lagoon	≤60
Lagoon seawater	1 m from bottom of Mike Crater	820 ± 40
		840 ± 40
Pacific Ocean seawater	Marshall Islands, December 1963*	7.9
Rainwater	Eniwetok Island, August 1964	86 ± 3

* IAEA[4]

Table 6. Tritium and ^{14}C content of animals collected at Eniwetok Atoll

Specimen	Location	Tritium units Loose water	Tritium units Bound water	^{14}C (Δ‰)
Coconut crab *Birgus latro*	Igurin Island reef	81 ± 30	≤95	20 ± 1
Hermit crab *Coenobita perlatus*	Sanildefonso Island Mike Crater	≤60	270 ± 30	136 ± 6
Killer clam *Tridacna crocea*	Runit Island reef	96 ± 21	≤80	−1 ± 3
Groupers *Epinephalus merrai*	Igurin Island reef	84 ± 27	170 ± 20	16 ± 4
Groupers *Epinephalus merrai*	Runit Island reef	187 ± 27	110 ± 32	24 ± 4
Surgeonfish *Acanthurus triostegus*	Runit Island reef	≤39	400 ± 80	20 ± 4
Groupers *Epinephalus merrai*	Sanildefonso Island Mike Crater	≤60	≤60	216 ± 7
Surgeonfish *Acanthurus triostegus*	Sanildefonso Island Mike Crater	≤60	90 ± 33	185 ± 7

Table 7. Soil-bound sources of tritium and ^{14}C at Eniwetok Atoll

Area	Tritium (T.U.) Loose	Bound	^{14}C (Δ%)
Cactus Crater	509 ± 30	3.54×10^6	2920 ± 65
Runit Island			
Engebi Island	≤60	3.68×10^5	1110 ± 30
Sanildefonso Island	≤60	7.50×10^4	840 ± 23
Mike Crater area			
Japtan Island	≤50	3.44×10^5	25 ± 5
Igurin Island	≤50	3.00×10^4	−40 ± 2

Table 8. ^{14}C content of plants on Runit, Engebi and Sanildefonso Islands

Sample and area	^{14}C (Δ%) Wood	Leaves	Litter	Soil
Cactus Crater, Runit Island				
Messerschmidia argentea,				
inner growth rings	59 ± 4	125 ± 6	204 ± 7	2920 ± 65
outer growth rings	78 ± 5			
Scaevola frutescens	69 ± 5	76 ± 5	87 ± 6	
Lepturus repens		102 ± 5		
Triumfetta procumbens		134 ± 6		
Engebi Island				
Messerschmidia argentea	41 ± 4	75 ± 5	58 ± 4	1110 ± 30
Scaevola frutescens	51 ± 5	60 ± 5	71 ± 4	
Sanildefonso Island				
Messerschmidia argentea	57 ± 4	70 ± 4		840 ± 23
Lepturus repens		102 ± 5		
Triumfetta procumbens		78 ± 5		

Table 9. ^{14}C content of plants on Japtan, Biijiri, Igurin and Aaranbiru Islands

Sample and area	^{14}C (Δ%) Wood	Fruit	Leaves	Litter	Soil
Japtan Island					
Messerschmidia argentea	7 ± 4				
Pisonia grandis	3 ± 4		69 ± 5	78 ± 5*	25 ± 5
				70 ± 4†	
Ochrosia oppositifolia	54 ± 5				
Igurin Island					
Messerschmidia argentea	16 ± 4				
Pisonia grandis	24 ± 1				
Cocos nucifera		64 ± 5			
Biijiri Island					
Messerschmidia argentea	41 ± 4				
Aaranbiru Island					
Messerschmidia argentea	54 ± 4				
Pisonia grandis	−6 ± 3				
Guettarda speciosa	34 ± 4				

* Young litter.
† Old litter.

some will be retained as interstitial water. Tritiated water (THO) is absorbed by the vascular plant along with stable water (HHO), although isotopic discrimination must occur at various levels in the plant metabolic processes that regulate the mass movement of water in plants. RANEY[3] reviewed the literature concerned with plant–water relationships, and also those relationships concerned with the movement of deuterated (DHO) and tritiated water (THO) into and within plants. He grew tobacco plants in tritiated Hoagland's solution for 80 days, and the following levels of equilibration with the nutrient solution were observed in the various plant tissues:

Tissue	*Per cent equilibration with the root solution*
Stem	97
Leaf veins	92
Mesophyll of leaf	62

The stem of this plant was 48 cm long. He also demonstrated similar results in an HHO-grown sunflower plant; in this case, the tritiated water was shown to be 92 per cent equilibrated with the root solution in 8 h. Therefore, in herbaceous plants, at least, we can expect relatively rapid absorption of tritiated water and movement into the shoot tissues after its appearance in the root zone. This rapid near-equilibration was also demonstrated by CLINE[6] who found that *Phaseolus vulgaris* shoot tissues contained 45–65 per cent of the tritium concentration in the nutrient solution within 12 h. Tissue-bound tritium in all shoot tissues was only about 1 per cent of the nutrient solution at 72 h.

According to RANEY,[3] the lack of complete equilibration (20–30 per cent below root solution THO concentrations) is attributed to the inflow of unlabeled water from the atmosphere and the dilution of the THO arriving from the root system by metabolic water formed in respiration in the leaves. Tests carried out in a growth chamber having predried atmosphere gave THO levels in leaf tissues which were near equilibrium (95 per cent) with the root solution concentration.

Studies by RANEY,[3] CLINE,[6] RANEY and VAADIA[7] and BIDDULPH and CORY,[8] which have produced much useful information on the movement of tritiated water in vascular plants, have been concerned mainly with herbaceous species for practical experimental reasons. Woody plants present more complex problems in their secondary tissues and pathways of water movement. WOODS and O'NEAL[9] injected 5 Ci of tritium in 500 ml of water into sandy soils at three depths and followed its uptake by various species of oak in North Carolina. They collected transpired water from leaves by enclosing them in plastic bags. The largest amount of tritium recovered from one branch (5–20 leaves) in 1 day was 4 μCi/ml of transpired water. The amount of dilution of the injected tritium by stable soil water was not measured. In the shallowest depth of tritium application, the transpired water of the trees reached approximately 400 times the background level on the day of application, and by the end of the second day, the high value of 4 μCi/ml was attained in the transpired water vapor.

These data point out that in woody plants as well as in herbaceous species a rapid movement of tritiated soil-water takes place into the root and shoot system, and that changes in soil-water tritium levels would be reflected in the loose-water tritium content of leaves in a very short time, probably within a day. These results were also demonstrated by LEWIS and BURGY[10] who injected microcurie amounts of tritium into wells at depths up to 83 ft in northern California. They detected the tritium activity in transpired water vapor from oak trees located 55 ft from the point of injection in 1 day.

In general, the results of these physiological studies indicate that tritiated water absorption and movement in plants approximates that of stable water and, for gross movements of water in plants, tritium may be regarded as an excellent tracer for loose or unbound water within the plant. Furthermore, the rapid rate of equilibration with the THO in the root zone indicates that the loose water of the plant is representative of the tritium concentration of the current ground water in the root zone. Changes in levels of tritium in the soil water will show in the loose water of plant tissues within 1–2 days.

With respect to the tissue-bound water of plants, isotopic effects are certainly present in the many biochemical reactions that involve the synthesis of new organic matter. Furthermore,

recent studies at the Chalk River Laboratories of the Atomic Energy of Canada Limited,[11] concerned with dating tritium dispersal in ground water by tree-ring analysis, indicate that part of the tissue-bound tritium of cellulose is held as exchangeable hydroxyl ions. The effect of these two processes on the tissue-bound tritium data reported in this paper has not been evaluated at this time.

The state of tritium in soil and geological materials, especially those modified and displaced by a nuclear detonation, has not been adequately described. According to RANKAMA and SAHAMA,[12] hydrogen is found in mineral structures in the four following states: independent hydroxyls, structural water, hygroscopic water and hydrides. Rock and soil materials also contain interstitial water which is held by capillary forces. Structural water and hygroscopic water, in addition to interstitial or capillary water, are the most important to an understanding of the problem considered. The water usually most available to plants from the soil is capillary and hygroscopic in form. Hygroscopic water is generally assumed to be available to plants to tensions of 31 atm (hygroscopic coefficient) and capillary water is readily available for absorption by roots. Structural water cannot be removed without breaking down the structure of the mineral, but there is biological evidence in this paper and elsewhere that structural water exchanges at some unknown rate with the free capillary and hygroscopic water of the soil or mineral materials.

The movement of tritiated water through geological strata has been studied at the Idaho Reactor Test site in connection with the monitoring of waste disposal wells.[13] An attenuation of tritium concentrations in the ground water was partially attributed to the exchange of tritiated water, with water of hydration or similarly bound water in the rock strata. RHODES and WILDING[14] conducted experiments with exchange columns of various rock types and alluvial sediments, and passed tritiated water through them. Even with small columns, an attenuation of tritium concentration occurred as the solution passed through the columns.

The Eniwetok soil materials are composed almost entirely of coral fragments and debris ($CaCO_3$ and $Ca(OH)_2$). The work of ADAMS, FARLOW and SCHELL[15] leads to the conclusion that these two compounds are the source of the high residual tritium and ^{14}C in the soil samples from Eniwetok Atoll. ADAMS, FARLOW and SCHELL studied large particles formed in nuclear detonations at Eniwetok, Bikini, and the Nevada Test Site.

Facts from the above data will help to interpret the preliminary data in Tables 1–9. These facts are:

(a) loosely-held tritiated water present in the root zone of vascular plants enters and moves within the plant along conventional physiological pathways,

(b) various tissues of the plant exhibit a dynamic near-equilibrium relationship with the tritium (THO) of the root zone,

(c) changes in the tritium concentration of the available soil water will be reflected in a short time in the loose-water tritium content of some plant tissues, and

(d) tritium may become bound in fallout and substratum materials of the detonation site in a tightly-held hydrated or chemically bound state because of the fireball and post-detonation physicochemical phenomena.

Interpretation of results

The reproducibility of the data reported in this paper is illustrated by the values in Table 4. These data were obtained from the analysis of duplicated samples from the same specimen of original sample. The water samples show excellent agreement and are both within the prescribed errors. The wood samples, in general show good agreement except for the one sample which shows an error by a factor of 2. Since it was not possible to obtain duplicate samples in every case, the data must be interpreted with this variation in mind.

The data in Tables 1, 2 and 3 indicate that in most of the environments from which the samples were obtained, a considerable source of tritium exists in a bound state within the soil materials and is slowly exchanging with the loose water of the soil. As expected, an increased level of tritium in the loose soil water produced an increased level of tritium in the loose water of the leaves of plants growing on that soil. The loose water tritium of *Messerschmidia* wood (3230 T.U

at Cactus Crater (Table 1) does not appear to be related to the current level of tritium in the soil water (509 T.U.) beneath that plant. Because the total loose water of a woody stem may contain water that is not involved in the current transpirational stream, the loose-water tritium value for wood is not expected to be in equilibrium with soil-water tritium levels. However, the loose water of the leaves of the *Messerschmidia* tree at Cactus Crater contained 442 T.U. which is within the range of expected equilibrium with the soil-water tritium (509 T.U.) indicated by experimental studies.

The variation in the bound tritium content of the *Messerschmidia* and *Scaevola* trees growing in the Cactus Crater area within 30 ft of each other is no doubt related to the usual variation in soil moisture conditions (whenever they are considered micro-topographically).

The high tissue-bound tritium values of plant samples at Cactus Crater indicate that not only is tritium present under the current environmental conditions, but also that it occurred in higher concentrations in the available soil water in periods preceding this survey and subsequent to the detonations. The higher level of bound tritium in the inner growth rings or older wood of *Messerschmidia* indicates greater concentrations at earlier times, even if exchange mechanisms are considered. The tissue-bound tritium content of the Cactus Crater litter samples, when compared to the tissue-bound tritium values for green leaves from the same tree, also indicates a varying concentration of tritium during the 12–15 months.

On Engebi and Sandildefonso Islands, loose water in plants and the soil had a low tritium content also. Previous to the sampling, however, available water must have contained more tritium because tissue-bound tritium levels are as much as thirty times (*Scaevola* litter) the current loose soil water.

Because of the low level of tritium in the available soil water in the presence of the large sources bound in the soil, the levels occurring in the organic matter as tissue-bound tritium seem to be anomalous. The following sequence of events should explain the conditions described above. The tritium levels in the soil water will steadily increase if it is not moving through the soil rapidly, as it would conceivably be in the rainy season. After heavy precipitation and in very porous soil materials such as coral sand, rainwater moves quickly through the soil profile and washes the shallow root zones of the atoll plants, lowers the loose-water tritium content, and leaves it with the same tritium content as the rainwater. The shallow root system of *Messerschmidia* was described by KENADY[16] who found that a 4 ft high tree had lateral roots which extended 60 ft from the crown within 2–6 in. of the surface of the ground. Because the plant absorbs large quantities of water daily, the loose water of the leaves will quickly adjust to the low tritium content of the new soil water, while the tissue-bound tritium of the same plant confirms that previously, tritium was more abundant in the water of its root zone. BLUMENSTOCK and REX[17] give an average value of 6.93 in. of rain on Eniwetok Island for August which was the time of sample collection. This would account for the lower loose-water tritium content in soil water at this time.

In Table 3, the tritium analyses of samples collected on four other islands on Eniwetok Atoll are reported. These islands have been contaminated by close-in fallout which contained large particles. Like the other islands, a large source of tritium is present, bound in soil materials. Also the loose soil water has a lower tritium concentration, and loose-water tritium values in plants, except for coconuts, were generally between 50 and 150 T.U. Higher levels were present in the tissue-bound tritium of the plants. The elevated tritium concentrations in the coconuts from Japtan and Igurin Islands would indicate that the tree had been exposed to water of much higher tritium concentration during the formation of the coconut.

The tritium content of water samples collected at Eniwetok Atoll is given in Table 5. Because of the frequent exchange of the lagoon water with the open sea and the vigorous mixing of the lagoon itself, the water in the lagoon is apparently uniform in its tritium content from surface to bottom. VON ARX[18] estimated the exchange of Bikini lagoon water with the open seas as once every 13–39 days depending upon the season. In spite of the frequent exchange of the lagoon with the open sea and the occurrence of intra-lagoon mixing, there is a local concentration of

tritium in the Mike Crater area. Since the substratum materials are the same in Mike Crater as they are on the islands, the same bound source of tritium exists in the coral sand of Mike Crater. The exchange rate of the tritium from those particles is adequate to produce a detectable local concentration in spite of the mixing which would tend to disperse it. Pacific Ocean seawater in the Marshall Islands area was reported by IAEA[4] as containing 7.9 T.U. in December 1960. The level of tritium in Mike Crater, therefore, represent a local concentration of at least 100 times that of the lagoon waters. Duplicate analyses were made of the Mike Crater water sample and are listed in Table 4.

The tritium and ^{14}C content of the animals collected on Eniwetok Atoll are given in Table 6. Tritium values for loose water in animals show a slight increase over the lagoon waters with which they should be in equilibrium.[19] There is an indication that the animals from the Runit reef environment and the Mike Crater area have an enriched tissue-bound tritium content. The hermit crab on Sanildefonso Island and the surgeonfish from the Runit reef area had the highest bound tritium content. More significant in marine animals, however, is the enrichment of ^{14}C in their tissues. All three animals from the Sanildefonso Island area contain elevated ^{14}C levels, with the fishes being the highest. The hermit crabs apparently obtained their ^{14}C from the algal debris which they ate at the water's edge, because their terrestrial diet of *Messerschmidia* litter does not appear to be high in ^{14}C. In the marine environment of Eniwetok Atoll, residual tritium and ^{14}C bound in coral debris are released at a slow but detectable rate and appear in the animals of the marine detonation environment at significant levels. While tritium and ^{14}C are elevated throughout the lagoon, Table 5 demonstrates that there is a definite localization in the area of Mike Crater on the basis of the ^{14}C content of animals living in that environment.

To characterize further the sources of tritium in the Eniwetok environment and to demonstrate the relationship with ^{14}C, the values for the soil samples collected in this survey are compiled separately in Table 7. These relationships may apply only for a substratum high in carbonate such as coral sand. In terrestrial environments the soil-bound tritium exchanges with the [illegible] soil water and is detectable in the plants which rapidly equilibrate with the soil-water tritium levels, but the typical route of entry for carbon into plants is from the atmosphere through the leaves. Nevertheless, as Tables 8 and 9 indicate, ^{14}C content of plant samples is also elevated. In the Cactus Crater area, where the ^{14}C soil was the largest, there is an indication of enrichment of ^{14}C in the litter of *Messerschmidia argentea* and the leaves of *Triumfetta procumbens*. The expected ^{14}C content of Eniwetok plants is some value less than 50 per cent (Δ per cent). KIGOSHI and ENDO[20] gave ^{14}C values for pine and *Paulownia* in Japan as 22.1 and 18.7 per cent for the 1959–1960 wood. These values do not include any increment of ^{14}C added to the world's atmosphere from the 1961 high yield tests. FERGUSSON[21] indicated that by July 1962, the surface ^{14}C in the air had increased to 37.5 per cent and in March 1962, grass in southern California had a ^{14}C content of 20.7 per cent. The low values in Table 9 for wood are in this range, but none of the areas sampled in this survey may be considered as uncontaminated and, therefore, elevated levels of ^{14}C may be expected anyplace within the atoll. Wood from trees growing in the Cactus Crater area have elevated ^{14}C values and recent organic matter, leaves and litter are even higher. The typical route of carbon entry into plants is from the atmosphere through the leaves. The basis of the elevated ^{14}C in the terrestrial plants is not evident in these data but two possibilities are suggested. The physiologically specialized atoll plants may absorb compounds containing ^{14}C through the roots, or there is an increased ^{14}C level in the immediate ground-level atmosphere of the plants due to gaseous release from the soil source.

Further evidence for elevated ^{14}C levels in the Mike Crater area (Sanildefonso Island) was obtained from the analysis of a marine alga collected in the Mike Crater area. Samples of algae from Igurin Island at the southern end of the atoll were also analyzed for ^{14}C content and the values are included for comparison. Dr. RALPH PALUMBO, University of Washing-

Laboratory of Radiation Biology, supplied the algae samples.

	delta % ^{14}C
Boodlea composita, Sanildefonso Is.	1430 ± 26
Halimeda opuntia, Igurin Is.	32 ± 4
Caulerpa serrulata, Igurin Is.	25 ± 3

The Eniwetok Island rainwater sample collected in August 1964 is listed in Table 5 as containing 86 ± 3 Tritium Units. This value was suspected and since the rainwater tritium level will theoretically represent the environmental water tritium base line, the sample was electrolytically enriched 100-fold. This analysis yielded a value of 27 ± 3 Tritium Units. Soil and plant loose-water tritium contents may therefore be compared with this base value.

In summary, the results of this preliminary survey of detonation environments in the Pacific Proving Grounds indicate that residual tritium and ^{14}C are present in relatively high concentrations in soil materials of the detonation sites up to 12 yr after the event. Exchange of soil-bound tritium with the available soil water takes place at a slow but significant rate, and tritium is detectable in plants growing in the detonation environments. Carbon-14 is also elevated in the terrestrial plants. The basis for the elevated ^{14}C is not implicit in these preliminary data. Tritium and ^{14}C are also present in elevated concentrations in marine organisms. However, because of the high rate of exchange of the lagoon waters with the open sea, these elevated concentrations are highly localized in the vicinity of the detonation site (Mike Crater area).

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