UNIVERSITY OF WASHINGTON SEATTLE, WASHINGTON 98105 410335 College of Fisheries September 30, 1970 Laboratory of Radiation Ecology Mr. Tommy McCraw Nuclear Explosives Environmental Safety Branch Division of Operational Safety U. S. Atomic Energy Commission Washington, D. C. 20545 Dear Tom: The draft of the report on iron-55 in Rongelap people is enclosed. Your comments would be appreciated. Tom Beasley and I feel that there should perhaps be some additional information, especially about current sources of iron-55 at Rongelap, before this is published.

we can obtain additional funds we will try to go to Rongelap with Bob Conard and coordinate that trip with

With best regards,

Research Professor

Edward E. Held

51/

a Bikini resurvey.

EEH:ah

Enc.

IRON-55 in Rongelap People,

Fish and Soils

T.M. Beasley and E.E. Held
Laboratory of Radiation Ecology
College of Fisheries
University of Washington

and

R.M. Conard .

Medical Department

Brookhaven National Laboratory

Upton, L.I. New York

BEST COPY AVAILABLE '

September 1970

Since 1965, the distribution of <sup>55</sup>Fe in the biosphere has been studied both in the United States and in the Scandinavian countries. Initially, <sup>55</sup>Fe concentrations were determined in Alaskan Eskimos, residents of Richland, Washington, and in representative foodstuffs of both (1). Subsequently, <sup>55</sup>Fe concentrations in environmental samples and in residents of Finland (2) and Sweden (3) were reported which generally confirmed the findings of the earlier study. Additional research shows that (i) marine organisms and people whose diet is largely seafood contain the highest concentrations of <sup>55</sup>Fe (4); (ii) residents of the northern hemisphere have higher <sup>55</sup>Fe body burdens than those of the southern hemisphere (5); and (iii) the <sup>55</sup>Fe levels in people reached peak concentrations in 1966 and continue to decrease (6,7).

We determined the <sup>55</sup>Fe body burdens of natives at Rongelap Atoll in the Marshall Islands. Not only is their diet high in seafoods (8), but the atoll received high-level fallout following the detonation of a thermonuclear device at Bikini Atoll in 1954 (9). We considered it probable that retention of <sup>55</sup>Fe at the atoll from that event, coupled with world-wide fallout from large-scale nuclear device testing in 1961-62 could lead to unusual body burdens of this radionuclide in the Rongelapese.

The method of separation of <sup>55</sup>Fe was identical to that previously described in this journal (1). The counting technique

was changed slig. ly; a gas filled (Xe) proportional counter operating in anticoincidence with an umbrella of nine Geiger-Muller tubes was used to detect the 5.9-kev x-ray emitted in the electron capture decay of <sup>55</sup>Fe. Pulses from the proportional counter were recorded in a 512-channel multichannel analyzer. The detection system was surrounded by four inches of lead shielding and the resultant background counting rate under the <sup>55</sup>Fe photopeak was 1.7 counts min<sup>-1</sup>. Disintegration rates of <sup>55</sup>Fe in the samples were determined by comparison with <sup>55</sup>Fe electrodeposited from a standard solution obtained from the National Bureau of Standards. Correction was made for the self-absorption of the X-rays in the electrodeposited iron. Stable iron was determined colorimetrically, using o-phenanthralein as the color-forming agent (10).

Body burdens were estimated by first measuring the <sup>55</sup>Fe in a known volume of blood (4-26 ml). Total blood volume was estimated using body weight and average blood volumes of 82 ml blood per kg body weight for males and 74 ml blood per kg body weight for females. The iron content of blood was assumed to be 65 per cent of the total body iron. This method of calculating total blood volume is that used by Persson (11) in his estimate of <sup>55</sup>Fe body burdens in Lapps of Northern Sweden. Previous estimates of body burdens from composite blood specimens (1,5) were made assuming that the average total blood volumes of 5 liters and that 60 per cent of the total iron is in the blood.

Using this method with the Rongelap data, the average <sup>55</sup>Fe body burdens agreed within 15 per cent of those calculated using body weights.

Table 1 gives the average body burdens of <sup>55</sup>Fe in a selected group of Rongelapese sampled in March, 1970, and Figure 1 shows a frequency distribution of the body burdens of males and females. Iron-55 levels in the blood samples were sufficiently high to permit count rate measurements to ± 5 per cent at the 95 per cent (20) confidence level. Not all donors were weighed in 1970, therefore body weights from previous years were used to compute total blood volumes. However, weights from previous years applied mostly to younger donors. Since weight generally increases with age, some individual estimates of <sup>55</sup>Fe body burdens, and therefore the averages shown in Table 1, are likely to be conservative.

The maximum body burden in the males was 0.85 μCi, while three females had body burdens greater than this value. The maximum observed female body burden was 1.0 μCi, approximately 1/100th of the maximum permissible body burden which has been established for non-occupationally exposed individuals considering the total body as the critical organ (18). Previous measurements of <sup>55</sup>Fe body burdens during a period of increasing <sup>55</sup>Fe fallout generally showed that <sup>55</sup>Fe body burdens of females were higher than those of males (4,5). Presumably this is due to higher †10.000

rates of iron in remales than in males, with the result that females are more nearly at equilibrium with their environment. As environmental levels of <sup>55</sup>Fe decrease, females should, on the average, reflect this change by exhibiting lower <sup>55</sup>Fe body burdens than those of males. Figure 1 shows that more female body burdens tended toward values <0.4 µCi, while male body burdens were more normally distributed, about a mean of 0.43 µCi. Regression analysis of age on body burdens showed a significant correlation (P(0.001); older individuals had higher <sup>55</sup>Fe body burdens. This observation is consistent with earlier data from U.S. citizens (5).

Comparison of the <sup>55</sup>Fe body burdens of peoples of different countries (5) requires knowledge of the turnover rates of <sup>55</sup>Fe in the environment and in humans. Jennings (12) has shown that the <sup>55</sup>Fe specific activities of salmon taken from the northeast Pacific Ocean decreased eightfold between 1964-1967. Assuming that a first order reaction governed the removal of <sup>55</sup>Fe from the mixed layer of the ocean (upper 100 m) he calculated the effective half-life for <sup>55</sup>Fe loss as 11 months. Measurements in cattle and rain waters show decreases, but at lesser rates (6). Iron-55 body burdens of adult males in Richland, Washington, decreased approximately fourfold between 1967 and 1970 (7), corresponding to an effective <sup>55</sup>Fe half-life of 1.5 years. If the <sup>55</sup>Fe turnover rates of Richland, Washington, residents are

from maritime cultures would exhibit similar and perhaps faster turnover rates of <sup>55</sup>Fe because of the short "ecological half-life" (13) of this radionuclide in the marine environment.

The highest <sup>55</sup>Fe body burdens previously measured were in female natives at Bethel, Alaska, during 1966 (5). The average body burden of eighteen females was 1.1 µCi. During the same year, the average <sup>55</sup>Fe body burden of females and males at Tokai-Mura, Japan, was 0.92 µCi and 0.63 µCi respectively (5). If the effective half-life of <sup>55</sup>Fe in Richland, Washington, males and in Tokai-Mura males is comparable, by 1970 the latter group would be expected to have body burdens of approximately 0.1 µCi, four times lower than those measured in Rongelapese males. Iron-55 body burdens of females at Tokai-Mura might be expected to be comparable to those of males for reasons discussed earlier.

All of the donors of the Rongelap study were subjected to external radiation during the accidental contamination of Rongelap Atoll in 1954. Because of the high levels of radioactivity at the Atoll, the Rongelap natives were moved to Majuro Atoll where they resided for three and one-half years. Following exposure in 1954, whole body counting and urinalysis disclosed measurable quantities of internally deposited fallout radionuclides. By 1957, however, the only radionuclides present in the Rongelapese in significantly measurable quantities were 65 Zn, 137 Cs and 90 Sr (9). No 55 Fe analyses were performed at that time so body burdens

of this radionuclide are not known. However, based on the levels of  $^{65}$ Zn,  $^{137}$ Cs and  $^{90}$ Sr observed,  $^{55}$ Fe body burdens were probably small (9). The Rongelapese returned to the atoll in mid-July, 1957. Whole body counting and urinalysis measurements one year later disclosed significant body burdens of several radionuclides, the most notable being  $^{137}$ Cs. It is probable that  $^{55}$ Fe body burdens increased similarly.

The concentrations of <sup>55</sup>Fe in a selected species of fish and in soils are listed in Table 2. Activity in the fish is based on wet weights for comparison with previous work. Pooled samples were used in the analyses to reduce the effect of within-sample variation. The <sup>55</sup>Fe values for soil samples collected in 1963 at both Kabelle Islet and Rongelap Islet are averages of samples collected at depth increments of 0-1.3 cm and 1.3-2.5 cm. Specific activities are not given for soils since varying amounts of pre-1954 plant detritus could significantly alter the stable iron content but not the <sup>55</sup>Fe activity. In this instance, activity per unit weight of soil is a better index of changes which occur as a result of input or loss than is specific activity.

The decrease in Fe specific activities in Mulloidichthys, sp. (goatfish) at Rongelap between 1959-1961 corresponds to an ecological half-life of 11 months, identical to that observed by Jennings (11). Higher specific activities may have occurred

at earlier times, however, estimates based only on exponential loss would not take into account possible retention and cycling of <sup>55</sup>Fe within the lagoon, or the time lag between deposition and maximum specific activity in the aquatic biota.

The increase in specific activity of goatfish liver between late 1961 and mid 1963 reflects the increased environmental concentrations of <sup>55</sup>Fe resulting from testing nuclear devices in 1961-62. Introduction of this radionuclide to Rongelap Atoll can occur both by atmospheric fallout and by water transport of radioactivity from oceanic regimes. The westward-moving North Equatorial Current is comprised of waters from northern latitudes where fallout from the 1961-62 tests were maximal (14). Speeds of surface currents in both the California and North Equatorial Currents are sufficiently large to account for transport of waters from 30-40° N latitude to Rongelap Atoll (15) in the time period 1961-63; similarly, maximum surface deposition of <sup>55</sup>Fe occurred in 1963 (14) and thus the data of Table 2 probably reflect contributions from both sources (16).

Our measurements of <sup>55</sup>Fe in soils collected from the same sites between 1959-1963 do not clarify either of the input sources mentioned above; the change in concentrations are greater than can be accounted for by physical decay. Natural processes which remove <sup>55</sup>Fe from the upper 2.54 cm of soil may preclude its use as a precise collector, and therefore the results are

useful only to indicate order of magnitude values of <sup>55</sup>Fe soil concentrations present at the collection time.

Unfortunately it is not possible to offer a clear argument in explanation of the <sup>55</sup>Fe body burdens of the Rongelapese presented here at this time. Samples from 1963 through 1969 would have shed light on the problem, but none are available for analysis. The possibility of Rongelap lagoon acting as a nutrient and trace-element "trap" similar to estuaries (17) is intriguing. Removal and retention of both stable Fe and <sup>55</sup>Fe from the North Equatorial Current could lead to high specific activities of the radionuclide in species important in the Rongelapese diet. The fact that livers from mature goatfish contain between 2-3 times as much stable iron per unit wet weight as do livers of mature salmon (4) is consistent with this argument.

Marshallese in general show a tendency toward anemia (9) and thus may absorb more iron from their diet than do non-anemic individuals. The <sup>55</sup>Fe body burdens of the Rongelapese may therefore only reflect more complete uptake of iron rather than uptake of iron of high specific activity. It is clear that further measurements of the specific activities of <sup>55</sup>Fe in the diets of the Rongelapese and the effective half-life they display for this radionuclide will be needed to clarify these possibilities.

## REFERENCES AND NOTES

- 1. H.E. Palmer and T.M. Beasley, Science 149, 431 (1965).
- T. Jaakkola, in Radioecological Concentration Processes,
   B. Aberg and F.P. Hungate, Eds. (Pergamon, Oxford, 1967),
   pp. 247-251.
- B. Persson, in Radioecological Concentration Processes,
   B. Aberg and F.P. Hungate, Eds. (Pergamon, Oxford, 1967),
   pp. 253-258.
- 4. H.E. Palmer and T.M. Beasley, Nature 211, 1253 (1966).
- 5. H.E. Palmer and T.M. Beasley, Health Physics 13, 889 (1967).
- 6. H.E. Palmer, et. al., Radiological Health Data Reports 9, 387 (1968).
- 7. C.E. Jenkins and J.C. Langford, Pacific Northwest Lab. Rep. 1051, Part 2 (1969), pp. 73-74.
- 8. D. Chakravarti and E.E. Held, J. Food Science 38, 221 (1963).
- 9. R.A. Conard, et. al., Brookhaven National Lab. Rep. 50029

  (T-446) (1967). This report contains a brief review of past medical surveys on Rongelapese natives and a bibliography listing medical reports, radioecological studies, and graduate theses which deal with work conducted at Rongelap Atoll.
- 10. E.B. Sandell, in Colorimetric Determination of Traces of Metals, B.L. Clarke, P.J. Elving and I.M. Kolthoff, Eds. (Interscience, New York, 1959). p. 541.

- 11. R.B.R. Persso Health Physics 16, 69 ( 59).
- 12. C.D. Jennings, Iron-55 in Pacific Ocean Organisms, Ph.D. Thesis, Oregon State Univ. (1968), 77 p.
- 13. E.E. Held, Pacif. Sci. 14, 18 (1960).
- 14. E.P. Hardy and J. Rivera, HASL Tech. Rep. 193 (1969), p. II-9.
- 15. J.P. Tully, in Studies on Oceanography, K. Yoshida, Ed. (Univ. of Wash. Press, 1965), pp. 68-94.
- 16. The higher specific activity noted in goatfish liver in August 1963 may reflect more nearly the specific activity of waters at Rongelap Atoll than does the sample of March 1963.

  Livers from immature goatfish were used in the August specimen, while the March sample was obtained from mature fish. Recent measurements of the specific activity of immature and mature goatfish liver collected at Johnston Atoll in 1968 showed that immature fish livers contained less stable iron and more 55 Fe per unit wet weight than did livers from mature fish.

  We assume that the immature fish are in the process of forming their iron stores and therefore more nearly reflect the environmental specific activities than do the mature fish whose iron stores are already formed. In the latter case, exchange rates may be slow.
- 17. T.W. Duke and T.R. Rice, in Proceedings of the Gulf and Caribbean Fisheries Institute, J. Higman, Ed. (Univ. of Miami, 1966), pp. 59-67.

18. International Commission for Radiological Protection,

Recommendations: Report 2, Permissible Dose for Internal
Radiation (Pergamon, Oxford, 1960).

TABLE 1. Average 55 Fe body burdens of Rongelapese

Date No. of subjects/sex 55 Fe (μCi)\*

March 1970 28/M 0.43±0.17

March 1970 32/F 0.40±0.27

<sup>\*</sup>Standard error (11) of the mean.

rable 2. Iron-55 c itent in goatfish (Mulloi chthys, sp.)

and soil samples from Rongelap Atoll. Sample numbers

are in parentheses; stable Fe and <sup>55</sup>Fe are expressed

per kilogram of wet tissue or per kilogram dry soil.

							Content	
Location	Sam	ple		Collection date		55 Fe (μCi)*	Stable Fe (g)	55 Fe (μCi)
		Bi	olog	ical Samples	3	•	•	•
Rongelap Islet	Liver	(20)		September	1959	4.04	0.98	4.1
Kabelle Islet	Liver	(7)		September	1961	0.68	0.76	0.9
Rongelap Islet	Liver	(10)		March	1963	1.40	1.15	1.2
Kabelle Islet	Liver	(71)		August	1963	2.49	0.37	6.7
			•	•		•		• .
				Soils				•
Kabelle Islet	Soil	(0-2.5	cm)	August	1959	0.24		
Kabelle Islet	Soil	(0-2.5	cm)	August	1963	0.06		
Rongelap Islet	Soil	(0-2.5	cm)	March .	1959	0.05		•
Rongelap Islet	Soil	(0-2.5	cm)	August	1963	0.006		
		•			•	•		

<sup>\*</sup> Sample counting times were arranged to determine <sup>55</sup>Fe in biological samples to ± 5 per cent at the 95 per cent confidence level (2¢); soil samples to ± 5-20 per cent at the 67 per cent confidence level (16). Stable Fe determination, ± 10 per cent at the 67 per cent confidence level (1¢). Activities are corrected to collection date.

		• .			•				14.0	10 X 01
.		. •			•					0
		,	•			· · . 15	anti are	: 432 T T T T		
			•	•			ar amas i	ries vizin	XII	•
			•		•	٠.				32
										0.7
1	E E				1 .		-	,	इ <b>ट्स्ट</b> र्स्स	8.0
İ						• •		. !	11221	°
	E X				•	, Periodor water	·			0.50
	12-			-				1,515	4000 ST	٥
			•							3
					<u> </u>	1375 CO.231 Children		2.30°		į
										9
							Services Free	31 T 32 T		4
				How we will			7			97.0
			•							
		Tresser.	GEOGRAPHICA NO post ou octobr		in facilities and	AND THE	<u>जन्म । त</u> ुः	जारण्या संस्थित		
		The second of th				(इस एक प्रा १९७४ (स्ट्रेस		ज्ञाप्य (क्रिके	ra Biratura Ligipapa bira	21.0
		William Stee				१८४१-६८५ स्था १९५८ (१८४८)			er green en	01.0
						નક્ષ્મ દાજાવા જિલ્લાના જિલ્લાના	सामा १६०१ व्याप्त १९८० व्याप्त १९८० व्याप्त			
						१६४१ ६४ ५४ ४४ ४४ १९-५४ १४ ४६			ng grand pa Mareen new	
						(20) torres			e grande	0:-
						(20) torres			e grande	0.10
										0.1 01.0
										0.10 09.0
										0.10 0.90
										0.10 0.00 0.00
							Earline L	eseren eseren		0.10 0.00 0.00
								25.75C		0.1 01:0 08:0 00:0
	RLES							25.75C		0.0 0.0 0.0 0.0
	MRLES							25.75C		0.10 080 01.0 09.0 0.0
	MRLES							25.75C		0.1 010 080 010 090 010
	MRLES				d german de filosofia e en estado en la estado en			25.75C		0.1 0.0 00.0 00.0 00.0
	MRLES							25.75C		0.1 010 080 010 090 010