Health Physics Pergamon Press 1972. Vol. 22 (March), pp. 245–250. Printed in Northern Ireland

IRON-55 IN RONGELAP PEOPLE, FISH AND SOILS

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(*Received* 5 *February* 1971; *in revised form* 12 *April* 1971)

Abstract—The ^{55}Fe body burdens for 60 residents of Rongelap Atoll are reported. The measured burdens are approximately 3 times higher than those of a similar number of residents from Tokai-mura, Japan. Since previous measurements in 1966 revealed substantial ^{55}Fe body burdens in Japanese residents, the current Rongelapese ^{55}Fe body burdens pose interesting questions.

INTRODUCTION

SINCE 1965, the distribution of ^{55}Fe in the biosphere has been studied both in the United States and in the Scandinavian countries. Initially, ^{55}Fe concentrations were determined in Alaskan Eskimos, residents of Richland, Washington, and in representative foodstuffs of both.[1] Subsequently, ^{55}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 ^{55}Fe;[4] (ii) residents of the northern hemisphere have higher ^{55}Fe body burdens than those of the southern hemisphere;[5] and (iii) the ^{55}Fe levels in people reached peak concentrations in 1966 and continue to decrease.[6,7] JAAKKOLA[8] has recently presented an excellent summary of the measurements of ^{55}Fe in Finnish Lapps which includes a valuable bibliography.

We determined the ^{55}Fe body burdens of natives at Rongelap Atoll in the Marshall Islands. Our interest in this particular population derives from two important considerations. First, the Rongelapese are a maritime culture, and they derive a large portion of their diet from the sea.[9] Prominent in this diet are the reef fishes; goatfish (*Mulloidicthys, sp.*), mullet (*Neomyxus, sp.*) and surgeon-fish (*Acanthurus, sp.*). Estimates of fish consumption vary, but daily intakes between 75–150 g appear reasonable. There are no apparent qualitative differences in the diets of males or females. Thus, the determination of ^{55}Fe in this population is of interest for comparison with other maritime cultures. Second, Rongelap Atoll received high-level fallout following the detonation of a thermonuclear device at Bikini Atoll in 1954.[10] We considered it probable that retention of ^{55}Fe at the atoll from that event, coupled with input from world wide fallout from large-scale nuclear device testing in 1961–1962 might lead to elevated concentrations of ^{55}Fe in marine species in the Rongelapese diet and therefore unusual body burdens of this radionuclide in Rongelap residents.

METHODS AND TECHNIQUES

The method of separation of ^{55}Fe was identical to that previously described.[1] The counting technique was changed slightly; a gas filled (Xe) proportional counter operating in anticoincidence with an umbrella of nine Geiger–Müller tubes was used to detect the 5.9-keV X-ray emitted in the electron capture decay of ^{55}Fe. Pulses from the proportional counter were recorded in a 512-channel multichannel analyzer. The detection system was surrounded by 4 in. of lead shielding and the resultant background counting rate under the ^{55}Fe photopeak was 1.7 counts/min^{-1}.

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Disintegration rates of ^{55}Fe in the samples were determined by comparison with ^{55}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*-phenanthroline as the color-forming agent.[11]

Body burdens were estimated by first measuring the ^{55}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% of the total body iron. This method of calculating total blood volume is that used by Persson[12] in his estimate of ^{55}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 volume was 5 l. and that 60% of the total iron is in the blood. Using this method with the Rongelap data, the average ^{55}Fe body burdens agreed within 15% of those calculated using body weights.

RESULTS AND DISCUSSION

Table 1 gives the average body burdens of ^{55}Fe in the group of Rongelapese sampled in March 1970, and Fig. 1 shows a frequency distribution of the body burdens of males and females. The Rongelapese which were included in this study consisted entirely of those individuals who were subjected to external radiation in 1954. Iron-55 levels in the blood samples were sufficiently high to permit count rate measurements to $\pm 5\%$ at the 95% (2σ) 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 ^{55}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 3 females had body burdens greater than this value. The maximum observed female body burden was 1.0 μCi,

Table 1. *Average* ^{55}Fe *body burdens of Rongelapese*

Date sampled	No. of subjects/sex	^{55}Fe (μCi)*
March 1970	28/M	0.43 ± 0.17
March 1970	32/F	0.40 ± 0.27

* Standard error (1σ) of the mean.

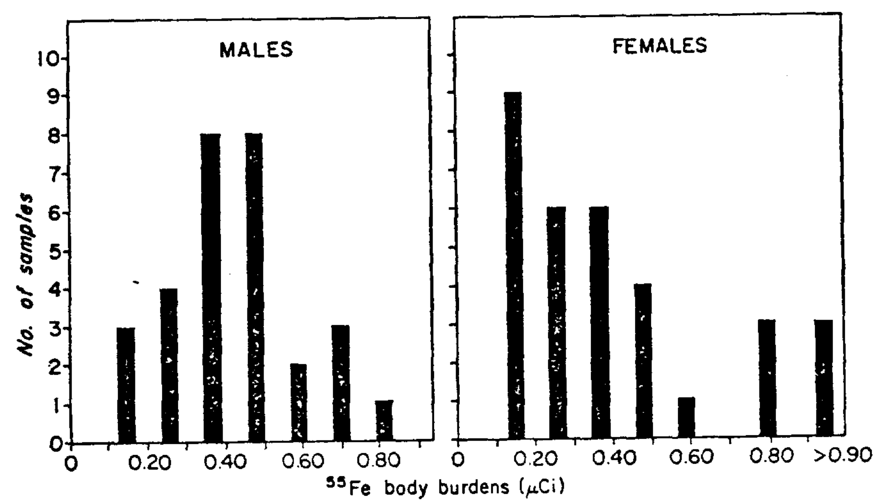

Fig. 1. Frequency distribution of ^{55}Fe body burdens in Rongelap males and females. Body burdens have been grouped into classes. Example: 3 Rongelap males had ^{55}Fe body burdens between 0.10–0.20 μCi while nine females had ^{55}Fe body burdens in the same activity interval.

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.[13] Previous measurements of ^{55}Fe body burdens during a period of increasing ^{55}Fe fallout generally showed that ^{55}Fe body burdens of females were higher than those of males.[4,5] Presumably this is due to higher turnover rates of iron in females than in males, with the result that females are more nearly at equilibrium with their environment. As environmental levels of ^{55}Fe decrease, females should, on the average, reflect this change by exhibiting lower ^{55}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 ^{55}Fe body burdens. Table 2 shows a tabulation of the average ^{55}Fe body of males and females by age groups. The number of samples per age group is admittedly small yet the general increase of ^{55}Fe body burden with age appears qualitatively consistent with earlier data obtained by analyzing blood from Seattle, Washington males in 1966.

Comparison of the ^{55}Fe body burdens of peoples of different countries[5] requires knowledge of the turnover rates of ^{55}Fe in the environment and in humans. JENNINGS[14] has shown that the ^{55}Fe specific activities of salmon taken

Table 2. Average body burden of ^{55}Fe in Rongelapese residents of different ages

	Age	Number of samples	Body burden (μCi)
Males	16–20	8	0.31
	21–31	4	0.33
	32–42	5	0.52
	43–53	2	0.58
	54–64	6	0.53
	>64	3	0.48
Females	16–20	6	0.23
	21–31	12	0.34
	32–42	5	0.33
	43–53	7	0.66
	54–64	2	0.57
	>64	2	0.66

from the northeast Pacific Ocean decreased eightfold between 1964–1967. Assuming that a first order reaction governed the removal of ^{55}Fe from the mixed layer of the ocean (upper 100 m) he calculated the effective half-life for ^{55}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 4 fold between 1967 and 1970,[7] corresponding to an effective ^{55}Fe half-life of 1.5 yr. If the ^{55}Fe turnover rates of Richland, Washington, residents are similar to those of insular populations, we conclude that people from maritime cultures would exhibit similar and perhaps faster turnover rates of ^{55}Fe because of the short "ecological half-life"[15] of this radionuclide in the marine environment.

The highest ^{55}Fe body burdens previously measured were in female natives at Bethel, Alaska, during 1966.[5] The average body burden of 18 females was 1.1 μCi. During the same year, the average ^{55}Fe body burden of females and males at Tokai-mura, Japan was 0.92 μCi and 0.63 μCi respectively.[5] We determined the ^{55}Fe body burdens of 32 females and 37 males from Tokai-mura from blood collected in October 1970; the average values were 0.12 and 0.17 respectively. Thus, not only do the Rongelapese have significantly higher ^{55}Fe body burdens than those of the Tokai-mura residents, but the decrease in the ^{55}Fe body burdens of this latter group from 1966 to 1970 appears comparable to that for Richland, Washington, males.

As previously stated, 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 3.5 yr. 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.[10] No ^{55}Fe analyses were performed at that time so body burdens of this radionuclide are not known. However, based

Table 3. *Iron*-55 *content in goatfish* (*Mulloidichthys, sp.*) *and soil samples from Rongelap Atoll. Number of fish livers contributing to single, pooled sample analyzed are in parentheses; stable* Fe *and* ^{55}Fe *are expressed per kilogram of wet tissue or per kilogram dry soil*

Location	Sample	Collection date	Content		
			^{55}Fe (μCi)*	Stable Fe (g)	$\frac{^{55}Fe}{Fe}$ $\frac{(\mu Ci)}{g}$
		Biological samples			
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		

* Sample counting times were arranged to determine ^{55}Fe in biological samples to $\pm 5\%$ at the 95% confidence level (2σ); soil samples to ± 5–20% at the 67% confidence level (1σ). Stable Fe determination, $\pm 10\%$ at the 67% confidence level (1σ). Activities are corrected to collection date.

on the levels of ^{65}Zn, ^{137}Cs and ^{90}Sr observed, ^{55}Fe body burdens were probably small.[10] The Rongelapese returned to the atoll in mid-July 1957. Whole body counting and urinalysis measurements one year later disclosed increased body burdens of several radionuclides, the most notable being ^{137}Cs. It is probable that ^{55}Fe body burdens increased similarly.

The concentrations of ^{55}Fe in a selected species of fish and in the calcareous soils are listed in Table 3. Activity in the fish is based on wet weights for comparison with previous work. The amount of sample that was available for analysis was small and therefore it was necessary to combine individual samples. This prevents an estimate of the range of ^{55}Fe concentrations which occur between fish, i.e. the within sample variation. The number in parenthesis in Table 2 for the biological samples indicates the number of fishes which contributed to the pooled, single sample analyzed. The ^{55}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 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 ^{55}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 ^{55}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.[14] 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 ^{55}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 ^{55}Fe resulting from testing nuclear devices in 1961–1962. 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

1961–1962 tests were maximal.[16] Speeds of surface currents in both the California and North Equatorial Currents are sufficiently large to account for transport of waters from 30 to 40°N latitude to Rongelap Atoll[17] in the time period 1961–1963; similarly, maximum surface deposition of ^{55}Fe occurred in 1963,[16] and thus the data of Table 2 probably reflect contributions from both sources. 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.

Our measurements of ^{55}Fe in soils collected from the same sites between 1959–1963 do *not* clarify either of the input sources mentioned above; the changes in concentrations are greater than can be accounted for by physical decay. Natural processes which remove ^{55}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 ^{55}Fe soil concentrations present at the collection time.

Unfortunately it is not possible to offer a clear argument in explanation of the ^{55}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. In a speculative vein, several explanations can be advanced. First, the possibility of Rongelap lagoon acting as a nutrient and trace-element "trap" similar to estuaries[18] is intriguing. Removal and retention within the lagoon of both stable iron and ^{55}Fe from the North Equatorial Current could lead to high specific activities of ^{55}Fe 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. Second, JENNINGS[14] and more recently PRESTON[19] have suggested that large amounts of ^{55}Fe were introduced into the northern hemisphere as tropospheric fallout during the 1961–1962 test series and that deposition may have occurred in rather narrow latitudinal bands (maximum input at 60°–70°N (Preston). JOSEPH *et al.*[20] suggest that subsequent stratospheric deposition of ^{55}Fe from these tests was similar to that observed for ^{90}Sr, i.e. maximum deposition at 45°N. Transport of ^{55}Fe from these high latitudes by major ocean current systems feeding the North Equatorial Current (in addition to biological and physico-chemical factors) will deplete northern waters of this radionuclide. As discussed earlier, transit time for these waters from high latitudes to Rongelap Atoll is measured in year, and it is therefore possible that the higher ^{55}Fe body burdens in the Rongelapese reflects a "lag" time between the ^{55}Fe specific activities of Northern Pacific and Southern Pacific Ocean biota due to this transport. Finally it is possible that the high ^{55}Fe Rongelapese body burdens relative to the Japanese donors results from a combination of dietary intake and uptake and retention differences for iron between the two groups sampled. Unfortunately precise dietary information for both groups is lacking and little is known about the ^{55}Fe specific activities of the foods eaten by both groups. The Rongelapese do not suffer from iron deficiency anemia, so enhanced uptake of iron from the diet is probably inconsequential. It is clear that further measurements of the specific activities of ^{55}Fe in the diets of the Rongelapese and other maritime cultures and the effective half-life they display for this radionuclide will be needed to clarify the questions raised here.

Acknowledgement—We thank Mr. T. A. JOKELA for technical assistance and Dr. M. FUGITA for kindly arranging for the blood specimens from Tokai-mura, Japan. Dr. A. H. SEYMOUR and Mr. H. E. PALMER provided helpful discussions during preparation of the manuscript. Supported by AEC under Contract No. AT(45-1)-2225-14 and AT(26-1)-269.

REFERENCES

1. H. E. Palmer and T. M. Beasley, *Science* **149,** 431 (1965).
2. T. Jaakkola, in: *Radioecological Concentration Processes* (Edited by B. Aberg and F. P. Hungate), pp. 247–251, Pergamon Press, Oxford (1967).
3. B. Persson, in: *Radioecological Concentration Processes* (Edited by B. Aberg and F. P. Hungate), pp. 253–258, Pergamon Press, Oxford (1967).
4. H. E. Palmer and T. M. Beasley, *Nature, Lond.* **211,** 1253 (1966).
5. H. E. Palmer and T. M. Beasley, *Health Phys.* **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, pp. 73–74 (1969).
8. T. Jaakkola, *Suomal. Tiedeatrat. Toim.* Series A. 2. *Chemica* 150 (1969).
9. D. Chakravarti and E. E. Held, *J. Fd. Sci.* **38,** 221 (1963).
10. 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.
11. E. B. Sandell, in: *Colorimetric Determination of Traces of Metals* (Edited by B. L. Clarke, P. J. Elving and I. M. Kolthoff), p. 541, Interscience, New York (1959).
12. R. B. R. Persson, *Health Phys.* **16,** 69 (1969).
13. International Commission for Radiological Protection, Recommendations: Report 2, *Permissible Dose for Internal Radiation*, Pergamon Press, Oxford (1960).
14. C. D. Jennings, Iron-55 in Pacific Ocean Organisms, Ph.D. Thesis, p. 77, Oregon State University (1968).
15. E. E. Held, *Pacif. Sci.* **14,** 18 (1960).
16. E. P. Hardy and J. Rivera, HASL Tech. Rep. p. II-9, 193 (1969).
17. J. P. Tully, in: *Studies on Oceanography* (Edited by K. Yoshida), pp. 68–94, University of Washington Press (1965).
18. T. W. Duke and T. R. Rice, in: *Proceedings of the Gulf and Caribbean Fishieries Institute* (Edited by J. Higman), pp. 59–67, University of Miami (1966).
19. A. Preston, *Mar. biol.* **6,** 345 (1970).
20. A. B. Joseph, E. A. Schubert and H. L. Volchok, in: *Radioactivity in Marine Environment* (Edited by A. H. Seymour), Nat. Acad. Sci.—Nat. Res. Council (in press).