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FALLOUT RADIONUCLIDES IN PACIFIC OCEAN TUNA

by

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ABSTRACT

Samples of light and dark muscle from tuna obtained in 1968 and 1969 from the Japanese tuna fishery in the Pacific, and the same tissues plus liver from tuna from the vicinity of Bikini Atoll, were analyzed for gamma-emitting radionuclides, iron-55 and stable iron. Tuna from the southern hemisphere tended to have lower concentrations and specific activities than tuna from the northern hemisphere. There were no significant trends in the data when ^{55}Fe specific activities were compared with species, month of catch, location of catch, or size of fish. There was a close correlation of ^{55}Fe specific activity in light muscle, dark muscle and liver and of ^{55}Fe concentration between dark muscle and liver. Yellowfin tuna caught near Bikini Atoll contained ^{60}Co believed to be derived from the atoll. There was a close correlation of ^{60}Co concentrations between dark muscle and liver. Cesium-137 concentrations were lower than those reported for fish from middle and northern latitudes by three to five orders of magnitude.

INTRODUCTION

Iron-55 is currently the most abundant fallout radionuclide in marine organisms. The radionuclide has been studied intensively because of its widespread occurrence since 1963, following the nuclear test series in 1962. Brill (1968) summarized reported data on ^{55}Fe levels in the environment, emphasizing dosimetric considerations for human populations. Persson (1969) reviewed the mode of production of ^{55}Fe, its distribution in the biosphere, and its dosimetry. Preston (1970) reviewed the occurrence of ^{55}Fe in marine fish and discussed its distribution and possible stratospheric anomalies leading to heavy depositions of ^{55}Fe relative to other fallout radionuclides in northern latitudes. Jennings (1968) suggested that much of the ^{55}Fe from the USSR tests of 1962 was deposited as tropospheric fallout in narrow bands related to the test sites at 75°N and 52°N.

In a review of radionuclides in marine organisms at the Pacific test site in 1956 to 1968, Lowman (1960) reported ^{55}Fe in plankton and fish. Other early reports of ^{55}Fe in marine organisms were those of Rama, Koide and Goldberg (1961), who found the radionuclide in the livers and hearts of Pacific Ocean tuna in 1961, and Seymour (1963), who reported its presence in in skipjack tuna from the North Pacific.

The present paper presents data obtained from analyses of Pacific Ocean tuna collected in 1967, 1968 and 1969. The occurrence of ^{60}Co and ^{137}Cs as well as ^{55}Fe in tuna is discussed.

MATERIALS AND METHODS

Tissues of both light and dark muscle from tuna caught between July 1967 and June 1969 were obtained at the Tokyo port of landing of the Japanese fishery. The samples were oven-dried at the National Institute of Health, Department of Food Control, Tokyo, under the supervision of Dr. T. Kawabata and shipped to Seattle for analysis. Dates and locations of catches were obtained from the vessels' logs. The species sampled were yellowfin (*Thunnus albacares*), bluefin (*Thunnus thynnus*), and big eye (*Thunnus obesus*).

The samples were redried in Seattle and prepared for analysis by gamma-ray spectrometry by compressing the tissues into standard counting geometries. After analysis for gamma-emitting radionuclides, portions of the samples, usually 20 grams per sample, were taken for analyses of iron-55 and stable iron, in duplicate. The light muscle and dark muscle obtained from the first seventeen fish were both analyzed for iron-55, but since the specific activities of the two tissues were similar, as would be expected, and the iron-55 and stable iron concentrations were greater in the dark muscle, only the dark muscle from fish subsequently obtained was analyzed.

In addition to the tuna from the Japanese fishery, sixteen yellowfin tuna, believed to be from a single school, were caught off Bikini Atoll in the Marshall Islands in September 1969. Liver samples, not obtainable from the Japanese-caught fish, and light muscle and dark muscle from these fish were analyzed for gamma-emitting radionuclides, iron-55 and stable iron.

The samples were counted for 900 minutes each with 3-inch x 3-inch NaI(Tl) crystals used in conjunction with 256-channel analyzers. Spectrum reduction was done by Schonfeld's (1965) method of least squares, and all values were corrected for decay to the date of collection.

Iron-55 was separated and purified by a combination of solvent extraction and electrodeposition techniques (Palmer and Beasley, 1967). Recoveries generally exceeded 90%. Counting was done by X-ray spectroscopy with a proportional counter used in conjunction with a multichannel analyzer. Some duplicate samples were analyzed by different persons using different methods, and the results were in close agreement.

RESULTS

The values for ^{55}Fe, stable iron and specific activity in tuna had a wide range. The ranges for dark muscle were ^{55}Fe, 3.3 to 1600 pCi/g dry weight; iron, 49 to 500 ppm; and specific activity, 0.02 to 8.6 nCi ^{55}Fe/mg Fe.

The frequency distribution of the specific activity of ^{55}Fe in tuna caught north of the equator and of tuna caught south of the equator in 1967 and 1968, and in tuna, presumably from a single school, caught at approximately 11°N in June 1969 are shown in Figure 1. The distributions have a strong positive skewness, with a definite tendency for a greater proportion of higher specific activities in tuna caught north of the equator. The tuna presumed to be from a single school off Bikini Atoll show a similar frequency distribution, although the range of specific activity values was less than in the other groups.

Attempts were made to correlate various parameters such as species, month of catch, location of catch, and size of fish with ^{55}Fe concentrations or specific activities; however, no significant relationships were found other than the tendency for fish caught north of the equator to have higher concentrations of ^{55}Fe and higher specific activities than fish caught south of the equator. Values at the extremes of the ranges for both ^{55}Fe concentration and specific activity, were found in tuna caught near the equator, 8°N for example, and in tuna caught at 39°N.

Results of comparisons made of the ^{55}Fe content and specific activities of the light muscle, dark muscle, and liver from the sixteen yellowfin tuna caught off Bikini are given in Table 1. Clearly, determination of ^{55}Fe specific activity in any of the tissues gives essentially the same results. However, greater concentrations of ^{55}Fe in the dark muscle and liver tissues than in light muscle make the former tissues more desirable for ^{55}Fe analysis. The ^{55}Fe concentrations in dark muscle and liver from single fish are approximately the same.

The average ^{137}Cs concentration in the light muscle of tuna caught north of the equator was 0.15 pCi/g dry, with a range from background to 0.43 pCi/g. As with ^{55}Fe, there appeared to be a tendency for lower ^{137}Cs concentrations (0.065 pCi/g, range, background to 0.27 pCi/g) in fish caught south of the equator, but the differences were not statistically significant. The tuna caught near Bikini Atoll contained almost the same average ^{137}Cs concentration (0.16 pCi/g, range, background to 0.31 pCi/g as other tuna caught north of the equator.

Cobalt-60 was not found in light or dark muscle samples of tuna from the Japanese fishery. The limit of detectability with the method used was estimated to be approximately 0.07 pCi/g dry weight. The yellowfin tuna caught off Bikini all contained small, but measurable, amounts of ^{60}Co in the dark muscle and liver tissues. The average value for dark muscle was 1.0 pCi/g dry, with a range of 0.08 to 4.6 pCi/g, and for liver, 1.3, with a range of 0.21 to 5.0 pCi/g. The correlation coefficient for ^{60}Co concentration in dark muscle and liver was 0.966 and the slope of the linear regression was 0.964. Hence, either liver or dark muscle tissue can be used for ^{60}Co determinations with essentially the same results.

DISCUSSION AND CONCLUSIONS

It is evident that species such as big eye, blue fin and yellowfin tuna, which migrate through large areas of the Pacific, are poor indicators of the geographical distribution of radionuclides in the Pacific. On the other hand, they are probably good integrators of ^{55}Fe from wide areas, and they contain both greater concentrations of ^{55}Fe and higher specific activities than species that have been successfully used to demonstrate latitudinal differences in ^{55}Fe content of fish from the northern hemisphere. Preston (1970) found higher specific activities of ^{55}Fe in cod from northern latitudes (65°-78°N) than in the middle latitudes (45°-52°N) of the Atlantic in 1967, 1968, and 1969. The highest value found in cod was 220.8 pCi ^{55}Fe/mg Fe at Iceland in 1969 and the average level in the liver of tuna from 11°N in 1969 was 1360 pCi ^{55}Fe/mg Fe. Maximum levels in tuna caught during 1967 and 1968 exceeded 8000 pCi ^{55}Fe/mg ~~dry~~ Fe, but the levels in most tuna during that period were less than 2000 pCi ^{55}Fe/mg Fe. Extrapolation of mean values to 1964, the peak year for ^{55}Fe levels in Pacific salmon (Jennings, 1968), gives values of approximately 5000 pCi ^{55}Fe/mg Fe. This value is approximately one-third the value reported by Jennings (1968) for chum and sockeye salmon from Petersburg, Alaska, in 1964. Yet, the levels reported by Jennings (1968) for sockeye and Chinook salmon from Bristol Bay, Alaska, in 1967 and by Jenkins (1968) for chum salmon from Kotzebue, Alaska, in 1968 are of the order of 500 Pci ^{55}Fe/mg Fe, or less than the levels in Pacific tuna from the northern hemisphere by a factor of more than two. Iron-55 in salmon from Alaska and the Washington coast appeared to decrease from 1964 to 1967 more rapidly than expected from decreases in the rate of fallout (Jennings, 1968; Jenkins et al., 1968). There was no significant decrease in ^{55}Fe specific activity during the years 1967, 1968, and 1969 in either Atlantic cod or Pacific tuna. Since the salmon were generally from areas of maximum rates of ^{55}Fe fallout in 1963 and 1964, it is possible that the decrease was a relatively localized phenomenon and that the ^{55}Fe levels in fish from the North Atlantic and from latitudes south of 40°N in the North Pacific approached equilibrium with the rate of fallout of ^{55}Fe. ~~Although~~ The large variability of the data casts doubts on the validity of such arguments, nevertheless, they are useful as working hypotheses.

It is clear that latitudinal differences occur in the North Atlantic and that at least hemispheric differences occur in the Pacific. Folsom and Young (1965) sampled distinct North Pacific and South Pacific populations of albacore (Thunnus alalunga) in

1964 and found that the concentration of ^{60}Co in the northern population was approximately five times that in the southern population; this supports the conclusion that ^{55}Fe concentrations are generally higher in northern hemisphere tuna than in southern hemisphere tuna.

The results of analyses of tissues from the yellowfin tuna caught off Bikini Atoll in 1969 show the large variability that can be encountered even with fish of approximately the same size, caught in the same place within a period of approximately an hour and believed to be from the same school. Certainly the analytical method is not suspect since the correlation for specific activities between tissues is high. One cannot be positive that these fish spent their entire life spans together and were thus exposed to the same water masses, but assuming that they were, there is no evident explanation for the large variability that was found.

Blood and liver usually contained the highest concentrations of ^{55}Fe, but in the highly vascular dark muscle of the tuna the levels were as high as they were in the liver. This may be of some importance in future sampling, as concentrations decrease, since it is often easier to obtain large samples of dark muscle than of liver.

Folsom and Young (1965) reported a ^{60}Co concentration of 0.16 pCi/g wet in the liver of albacore from the North Pacific, and Seymour (1963) reported an average ^{60}Co concentration of 0.68 pCi.g wet in the liver of eleven tuna from the Central Pacific. We found no detectable ^{60}Co in tuna from the Japanese fishery in 1967 and 1968, but did find significant concentrations of ^{60}Co in yellowfin tuna collected near Bikini Atoll in 1969. It appears that the source of ^{60}Co in these yellowfin tuna was the atoll. This is consistent with Welander's (1967) report of relatively high concentrations of ^{60}Co in marine organisms at Bikini Atoll in 1964. Liver of fish from different islets in the lagoon contained average ^{60}Co concentrations ranging from 79 to 330 pCi/g dry. Held (1971) reported values as high as 260 pCi/g dry in fish livers collected at Bikini in 1969. If the source of ^{60}Co for the yellowfin tuna sampled is indeed Bikini Atoll, it would appear that the tuna stay in the vicinity of the atoll for an appreciable period of time.

It would be pertinent to investigate Bikini and Eniwetok Atolls, sites of nuclear and thermonuclear tests from 1946 to 1958, as possibly significant point sources, essentially, of radionuclides in the North Equatorial Current.

Cesium-137 concentrations in the Pacific tuna, including those from the Bikini area, were of the same order of magnitude as the values reported by Seymour (1963) and by Folsom and Young (1965), average values of 0.03 to 0.09 pCi/g wet muscle tissue.

However, the values reported by Preston (1970) for cod and plaice from northern latitudes were higher by from three to four orders of magnitude. Pacific salmon sampled in 1967 (Jenkins, 1969) contained ^{137}Cs concentrations generally in the same order of magnitude as those of the cod but in some cases even an order of magnitude greater. These differences between ^{137}Cs concentrations in tuna and those in the fish from the middle and northern latitudes reflect the well-known latitudinal differences in world-wide fallout (Joseph et al., in press) and probably also reflected the contributions of ^{137}Cs from surface runoff in some areas, as Preston has pointed out.

In recapitulation: The tuna from the southern hemisphere tended to have lower concentrations and specific activities than tuna from the northern hemisphere. There were no significant trends in the data when ^{55}Fe specific activities were compared with species, month of catch, location of fish, or size of fish. There was a close correlation of ^{55} specific activity in light muscle, dark muscle and liver and of ^{55}Fe concentration between dark muscle and liver. Yellowfin tuna caught near Bikini Atoll contained ^{60}Co believed to be derived from the atoll. There was a close correlation of ^{60}Co concentrations between dark muscle and liver. Cesium-137 concentrations were lower than those reported for fish from middle and northern latitudes by three to five orders of magnitude.

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Table 1. Comparison of iron-55 concentrations and specific activities of three yellowfin tuna tissues.

		pCi/g dry		nCi ^{55}Fe/mg Fe	
	N	$\bar{x}$	σ	$\bar{x}$	σ
Light muscle	16	29.1	± 16.2	1.18	±0.72
Dark muscle	16	334	±193	1.16	±0.56
Liver	16	374	±234	1.36	±0.66

Correlation coefficient

Dark muscle vs liver (pCi/g dry)	0.886
Light muscle vs dark muscle (nCi ^{55}Fe/mg Fe)	0.900
Light muscle vs liver (")	0.838
Dark muscle vs liver (")	0.977

Regression coefficient (b) and intercept (a)

	b	a
Dark muscle on liver (pCi/g dry)	1.07	15.2
Light muscle on dark muscle (nCi ^{55}Fe/mg Fe)	0.706	0.332
" " " liver (")	0.764	0.455
Dark " " " (")	1.14	0.033

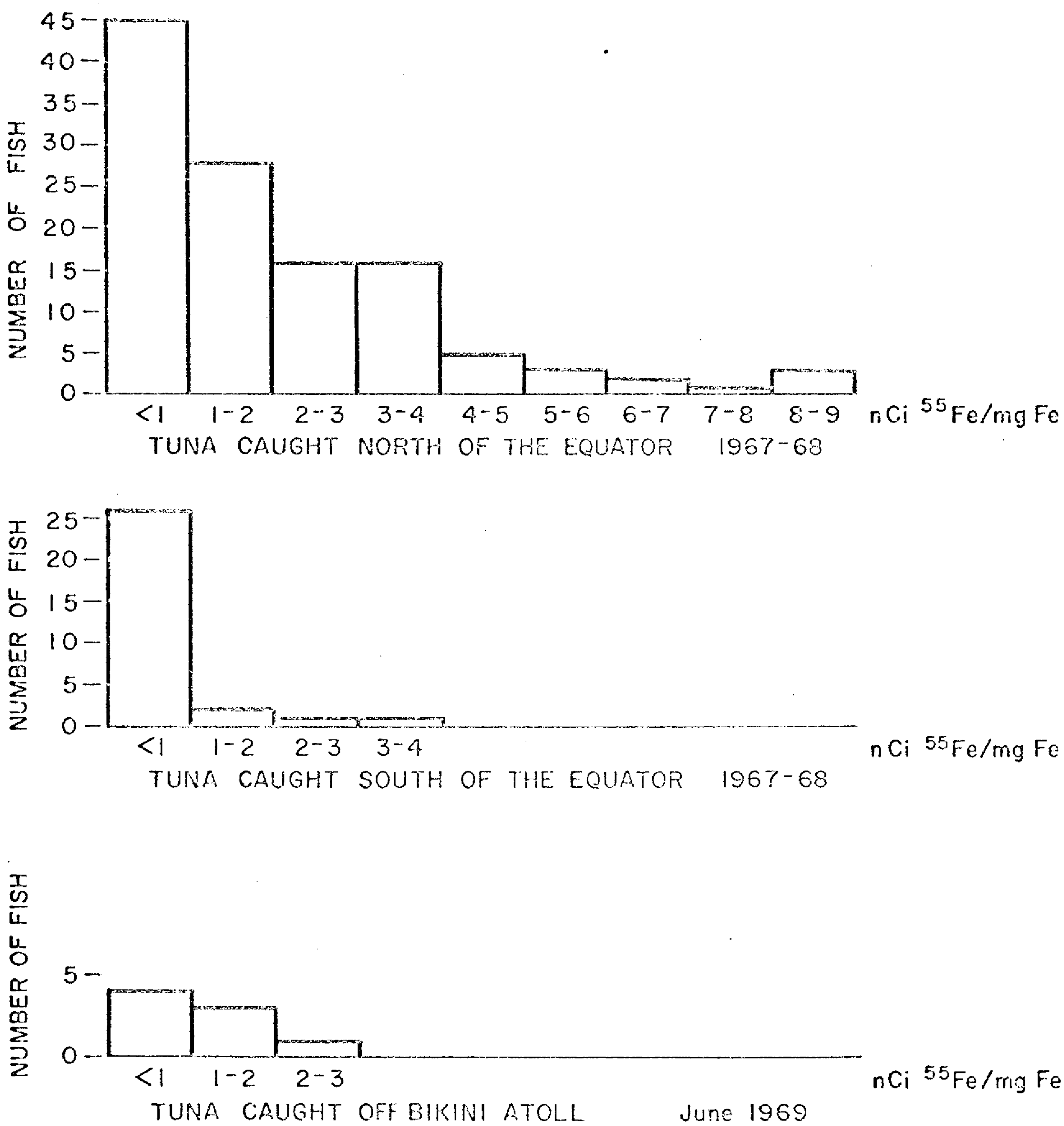

Fig. 1. Frequency distribution of ^{55}Fe specific activity in Pacific Ocean tuna.