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⁵⁵Fe CONCENTRATION AND SPECIFIC
ACTIVITIES IN NORTH PACIFIC MARINE ORGANISMS

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ABSTRACT

Marine organisms have been shown to have up to 1000-fold higher specific activities of iron-55 than that in seawater. It was also shown that iron-55 specific activities increase 10 to 30-fold in salmon between mid and northern latitudes. These observations are currently explained on the basis of a different chemical form for iron-55 in seawater and greater dilution of the iron-55 by stable iron at mid-latitudes than at the northern latitudes. The half-time for iron-55 concentrations in mature harvested salmon at all latitudes is approximately 10 months, which is comparable to the half-time for movement of stratospheric debris to the earth's surface. This observation precluded the possibility that early tropospheric deposition of iron-55 from the 1961-62 Russian nuclear tests contributed significantly to the long-term concentrations of iron-55 in marine biota.

INTRODUCTION

Our previous studies of iron-55 in the marine environment have shown a marked departure from what one would anticipate if it were assumed that the radio and stable iron behaved in the same manner. Two very significant observations which have prompted a continuation of our iron-55 studies in the marine environment are (1) the specific activity of iron-55 is 10 to 1000-fold higher in some ocean organisms than in seawater,^{1,2} and (2) the concentrations and specific activity are much higher in salmon from northern Alaska than they are from off the contiguous U. S. west coast.² During the 1961-62 U. S. and Russian nuclear testing, the iron-55 increased from a relatively minor constituent of fallout to the most abundant long-lived radionuclide.³ Iron-55 is formed through neutron capture by iron-54 and iron-56 in (n, γ) and in (n,2n) reactions, respectively. It has a half-life of 2.4 years and decays by emission of a very weak energy 5.9 keV X-ray. It is of particular interest its stable element counterpart is a biologically essential element and it deposits its energy mainly within blood cells and the blood forming cells of the human body.⁴

In considering food pathways of ⁵⁵Fe to man, marine food chains have been shown to be far more important than terrestrial food chains.^{1,3,5} It has been shown that whole population groups which consume large amounts of seafood, including the Japanese, Scandinavian and the southeast Alaskan Eskimos, have far higher iron-55 burdens than the average of other people in the world.^{1,6} The environmental chemistry of iron-55 has been of particular interest, since it is a striking example of where a radionuclide behaves grossly different in biological uptake rates than its stable element counterpart. The problem of 10 to 30-fold higher ⁵⁵Fe specific activities

in salmon from northern latitudes than those from the mid-latitudes has been difficult to explain, since the normal radionuclide deposition pattern results in higher deposition at the mid-latitudes. Jennings^{7,8} and Preston⁹ have suggested that the high iron-55 specific activity observed in marine organisms in northern latitudes resulted from high tropospheric deposition of nuclear debris from the 1961-62 USSR nuclear tests. Our most recent observations and the data included in this report refute their conclusion and present evidence for other mechanisms which seem to satisfactorily explain these apparent anomalies.

ANALYTICAL METHOD

The only radiation associated with the decay of ⁵⁵Fe is a 5.9 keV X-ray. Its low energy requires a chemical separation from the sample to permit its sensitive quantitative measurement. This method is well documented^{5,8} and includes its separation and electro-deposited on a copper planchet using the method of Maletskos and Irving.¹⁰ The ⁵⁵Fe X-rays are counted on a 90% argon - 10% methane gas flow proportional counter that is placed between large plastic scintillation detectors which are operated in anticoincidence to reduce the background. This assembly is shielded by 4" thick lead. The background is approximately 1 count per minute while the minimum detectable amount of ⁵⁵Fe is 18 disintegrations at the 99% confidence level in a 60 minute count.

RESULTS AND DISCUSSION

In our early studies of the concentrations of radionuclides in various seafoods, it was observed that tuna and salmon contained much higher concentrations of iron-55 than any of the terrestrial foods. Subsequent measurements of iron-55 specific activities in seawater showed that the specific activities in some marine organisms were 1000-fold higher than in seawater. This, together with the observations that much higher concentrations existed at the northern latitudes, spurred our interest in elucidating the mechanisms responsible for these phenomena. During 1968 a spectrum of marine organisms was sampled off the coast of Washington and in the Columbia River estuary. The specific activities (dpm iron-55 per mg of iron) are compared with the specific activities of iron-55 in air and seawater (Figure 2). This data does not give a clear indication of food chain relationships which bear on the specific activity of iron-55 in the marine organisms; however, it does provide some essential basic information. It is apparent that the specific activity in the lower trophic levels (euphausiids) are not nearly as high as in some of the predaceous fish such as sea bass, salmon, and rock fish.

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Bottom dwelling organisms such as the crab, sole and cod also have specific activities which are comparable to those in seawater. The rosy rock fish which had the highest specific activity is known to feed on crustacea such as the euphausiids, copepods and amphipods as well as small fish. An analysis of the data in Figure 2 simply does not provide sufficient information to suggest a mechanism for the very high specific activities in the predaceous fish. A second spectrum of marine organisms was collected during 1971 from locations along the west coast which are indicated in Figure 3. This sampling included phytoplankton which, of course, are the very basis of the food chain for essentially all of the ocean organisms. As shown in Figure 4, the specific activity of iron-55 in phytoplankton is only about twofold greater than that in seawater and it would appear difficult to explain specific activities which are 10 to 100-fold higher on the basis of this primary food material for ocean organisms. Phytoplankton samples are a mixture of actual floating plant organisms together with a variety of inorganic and detrital matter. It seems possible that this mixture could have a relatively low specific activity, whereas the parts actually metabolized by euphausiids and other filter-feeding organisms would have a substantially higher specific activity. Based on all the observations, we believe that this is the case. The "phytoplankton mixture" which we have been able to sample contains a substantial amount of stable iron which is either not consumed by filter-feeding organisms, or is not metabolized. There are other peculiarities in the food chain; for example, pink shrimp are known to include filter-feeding crustacea in their diets. They do, however, feed on or near the bottom during the day and then rise to near the surface at night. Measurements of their gut content show large amounts of sediment material containing high concentrations of stable iron which apparently serves to dilute iron-55 taken in during their feeding on crustacea. We acknowledge that large variations in the iron-55 specific activity of phytoplankton could exist and that our observed measurement in Figure 4 may not be typical. However, phytoplankton measurements elsewhere have also shown a similar low iron-55 activity.¹¹ The sampling of phytoplankton is normally done in phytoplankton blooms which are fed by nutrient-rich upwelling waters. These would be expected to be low in iron-55.

Differences in Specific Activity as a Function of Latitude

Our previous observations as well as those reported here show a 10 to 30-fold higher specific activity in northern salmon than in those collected at mid-latitudes. This is contrary to what one would predict from fallout patterns. Measurements during the past decade at Pt. Barrow, Alaska (70°N latitude) and Richland, Washington (46°N latitude) indicate that fallout rates of iron-55 and other radionuclides have been consistently two to threefold higher at 46°N than at 70°N latitude.¹² A possible explanation for the higher specific activity of iron-55 in marine organisms in the northern latitudes is that there is simply less stable iron available for uptake. To determine

if larger amounts of iron are entering the ocean at mid-latitudes than at the northern latitudes, stable iron has been measured on air filters at Pt. Barrow, Alaska and Richland, Washington. These concentrations, which are summarized in Figure 5, suggest that the stable airborne iron is at least an order of magnitude higher in the mid-latitudes. If a fraction of this stable iron were biologically available to the marine organisms, then it could certainly account for the lower specific activities in the mid-latitude salmon. Evidence that this is the case has recently been developed in other studies at our laboratory. It has been found that most of the stable iron in mid-latitude aerosols is associated with particles of less than 1μ and essentially all of this iron is from anthropogenic sources.¹³

The specific activities of iron-55 in salmon which were harvested off the west coast from mid-California through northern Alaska during 1968 are summarized in Figure 6. The specific activities show a progressive increase from mid-California through northern Washington and an additional increase in one of the salmon runs from Kotzebue, Alaska (68°N latitude). From tagging studies, salmon from Alaska, Canada and Washington rivers are known to mix in an ocean area that extends along the Aleutian chain and out into the Pacific Ocean.¹⁴ This common migration area may explain the comparable concentrations in some salmon runs which were sampled between northern Washington and Kotzebue, Alaska. The threefold higher concentration in chum salmon from the August run (see Figure 6) suggests another feeding area, perhaps to the north of the Aleutian chain in the Bering Sea.¹⁵

Variation in individuals from a given species provides an indication of the complexity of the food web. As shown in Figure 6, the ⁵⁵Fe specific activity of mature salmon collected off the west coast near the mouth of the Columbia River showed a relatively wide range compared with the high specific activity chum salmon collected at Kotzebue, Alaska (68°N). The wide range in specific activity of the former group may well be due to the large variety of organisms which comprise their diet. A much narrower range in Kotzebue may reflect a simpler food web. The immature salmon collected near the mouth of the Columbia River (see Figure 6) showed much lower specific activities than mature salmon. This is apparently due to higher concentrations of stable iron in the Columbia River and in coastal waters where these organisms had developed. In Figure 7 the changes in specific activity with time are plotted for salmon which are believed to come from three different migration areas. The half-times of the slopes of these curves are similar and average about 10 months. This is comparable to the half-time of stratospheric debris. The nearly parallel nature of these curves verifies that the input rates of ⁵⁵Fe into the migration areas of these three groups of salmon have been relatively constant over the last several years. The decrease in these salmon specific activities with time parallels the decrease in input rate of ⁵⁵Fe from the atmosphere; therefore, major tropospheric deposition at

northern latitudes (from the 1961-62 nuclear tests) could not have produced a significant long-term increase in northern versus mid-latitude marine organisms. Further evidence that ^{55}Fe specific activity in ocean organisms reflects atmospheric concentrations is evidenced by the similar increase in contiguous west coast and Alaskan salmon during the last few years. These increases, of course, are due to fallout from the Chinese nuclear tests. Aquarium studies have shown that when iron salts are added to seawater, they are initially taken up at a high rate by marine organisms; however, after a few weeks this iron becomes essentially unavailable for biological uptake.¹⁶ These studies confirm our actual ocean observations where the evidence shows that the ^{55}Fe specific activities in organisms simply reflect atmospheric input rates and show no evidence of cumulative effects from past ^{55}Fe additions. One might expect to find significantly different ^{55}Fe specific activities in different species of salmon. We have measured the specific activity of ^{55}Fe in four salmon species which were sampled during August, 1972 at Cook Inlet, Alaska. These data, shown in Figure 8, indicate relatively little difference between species. The sockeye do show the lowest concentrations and this may be due to the fact that they are predominately plankton feeders. As previously indicated, planktonic material does have a considerably lower specific activity than other organisms which comprise the foods utilized by salmon.

SUMMARY AND RECOMMENDATIONS

The ^{55}Fe from nuclear weapons testing which enters the ocean is in a chemical form which is much more readily available for biological uptake than is stable oceanic iron. This property of ^{55}Fe results in up to 1000-fold higher ^{55}Fe specific activities in marine organisms than is present in seawater. Dilution of ^{55}Fe by stable iron from anthropogenic sources or natural sources appears to be responsible for the lower specific activity in mid-latitude marine organisms than in organisms from high latitudes. The half-time for ^{55}Fe in harvested salmon from the west coast is essentially the same as the half-residence time for stratospheric radioactive debris. This fact suggests that ^{55}Fe reaching the ocean surface is available for biological uptake for a relatively short time. Measurements of ^{55}Fe specific activity in the various trophic levels of marine organisms does not give a clear picture of the reason for the much higher ^{55}Fe specific activities in certain crustacea and predacious fish than in the phytoplankton. A more detailed food web and chemical form study is recommended to elucidate this apparent anomaly. What we have learned about the behavior of the radionuclide ^{55}Fe in the ocean environment may well apply to other fallout radionuclides, and a consideration of this fact should be included in estimating the potential hazard of the release of radionuclides to the marine environment. For example, these data suggest that the release of radioiron from a reactor to the oceans at northern latitudes may have a 10 to 30-fold greater impact than releases of radioiron at mid-latitudes, and this may be true for other radionuclides.

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FIG. 1

Sampling Locations for the Collection of Columbia River Estuarine and Adjacent Pacific Ocean Marine Organisms

FIG. 2

55 ⁵⁵Fe SPECIFIC ACTIVITIES IN AIR, SEAWATER, AND MARINE ORGANISMS-COLUMBIA RIVER REGION 1968

FIG 3

SAMPLING STATIONS FOR COLLECTION OF MARINE
ORGANISMS R/V THOMPSON - OCTOBER, 1971

FIG. 4

⁵⁵Fe/Fe SPECIFIC ACTIVITIES IN AIR, SEAWATER, AND MARINE ORGANISMS OREGON-WASHINGTON COAST OCTOBER 1971

FIG. 5

AVERAGE YEARLY STABLE IRON CONCENTRATIONS
IN POINT BARROW, ALASKA, AND RICHLAND, WASHINGTON
AEROSOLS-mg Fe/10³ SCM

$^{55}\text{Fe}/\text{Fe}$ SPECIFIC ACTIVITY IN PACIFIC SALMON-1968

FIG 7

^{55}Fe CONCENTRATIONS IN PACIFIC SALMON

1972-COMPARISON OF $^{55}\text{Fe}/\text{Fe}$ SPECIFIC ACTIVITIES IN SALMON

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