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Large quantities of ^{55}Fe, formed by nuclear testing during 1961 and 1962, have been deposited on the North Pacific Ocean as fallout and concentrated by marine organisms. As a result of this high ^{55}Fe fallout and of low concentrations of iron in the ocean, marine organisms are the site of high ^{55}Fe specific activity (^{55}Fe activity/weight of total iron).

Iron-55 specific activity was seen to vary among five species of Pacific salmon, with sockeye salmon showing highest and chinook salmon showing lowest ^{55}Fe specific activity. Variation of ^{55}Fe specific activity among species of salmon appears to result from their migration routes and feeding habits. In 1964 at Strait of Juan de Fuca, Canada, where the largest ^{55}Fe specific activity difference between the chinook and sockeye salmon was observed, the ^{55}Fe specific activity of chinook salmon was 0.745 $\mu Ci/g$ Fe and that of

sockeye salmon was 28.7 μCi/g Fe.

In subsequent years ^{55}Fe in salmon decreased, and from this decrease an eightfold decrease in ^{55}Fe in the surface layer of the ocean, corrected for physical decay to the time of initial input, was observed. Assuming an approximately instantaneous input of fallout ^{55}Fe and a first order reaction, the rate constant for the removal of ^{55}Fe from the surface layer of the ocean was calculated to be 0.0603 $month^{-1}$.

Iron-55 removed from the surface layer of the ocean does not reappear in high specific activity in sediments or in benthic organisms, probably because the high stable iron content of sediments dilutes the ^{55}Fe, thus yielding a low ^{55}Fe specific activity.

The accumulated input of ^{55}Fe into central Alaska, measured in lichen, was found to be 209 nCi/m^2 and a calculation of the ^{55}Fe present in the North Pacific Ocean from ^{55}Fe specific activity in salmon was found to be 6.1×10^5 nCi/m^2. The assumption that ^{55}Fe is uniformly distributed throughout the ocean, used in the calculation of ^{55}Fe present in the North Pacific Ocean, accounts for part of the discrepancy between these figures. The remaining discrepancy is apparently due to differing amounts of ^{55}Fe laid down as fallout, with the North Pacific Ocean receiving at least ten times as much as Alaska.

Iron-55 in Pacific Ocean Organisms

by

Charles David Jennings

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INTRODUCTION

General Remarks

Utilization of nuclear energy, both in national defense and in peaceful applications, has raised the question of what effect radioactive contaminants in the environment have on man. Comar and Lengemann (1967) visualize the entire scope of problems concerned with environmental contamination by radioactive materials in terms of systems analysis with the input as contamination of the environment and the output as harm to man. Between radioactive contamination and harm to man lies a series of interrelated "black boxes," such as uptake and turnover of radionuclides by various members of the food web, exchange of radionuclides with sediments as well as many other processes. The interrelationships among these black boxes are depicted in Figure 1. Through research these black boxes are elucidated one by one with the ultimate goal being a complete picture of the consequences to man of radioactivity in the environment.

Another approach which can be taken in the study of radioactive contamination does not concern itself with possible harm to man, but rather considers the contaminant a tag which can be

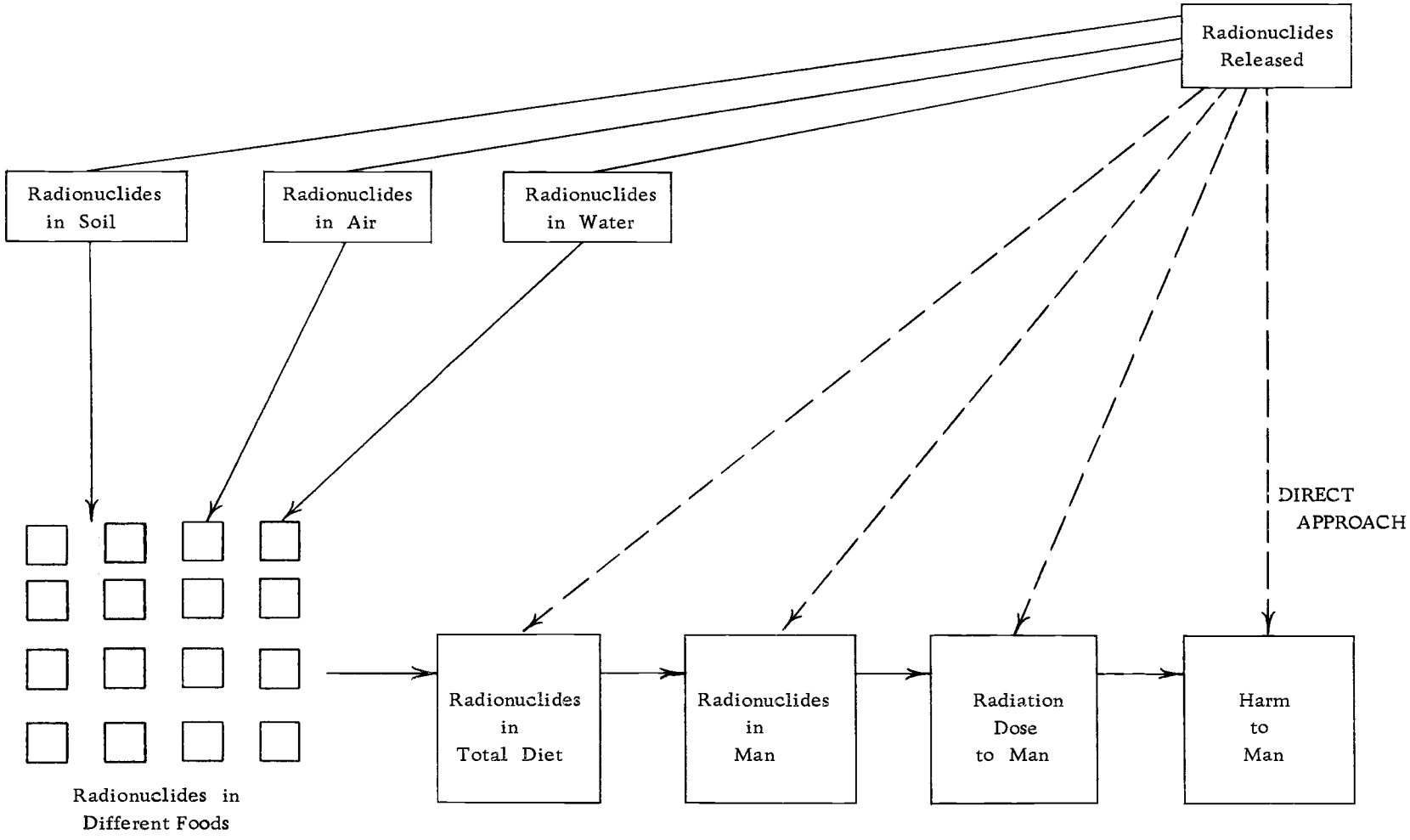


Figure 1. The major step-wise approaches to investigation of environmental contamination with radioactivity (after Comar and Lengemann, 1967). Solid lines represent the major pathways to be studied.

followed through the environment. In this way artificial radionuclides can serve as environmental tracers and thus can yield information relative to the movements of ocean water and migrations of organisms in the ocean.

This study contains some features of both approaches. Iron-55 in salmon, a food fish, is discussed in terms of migration routes and feeding habits of salmon which in turn relate to its potential harm to man. In addition, the vertical movement of ^{55}Fe in the ocean is considered by measuring concentrations in benthic organisms. This gives some insight into the mixing of the deep layers of the ocean and into methods of vertical transport of ^{55}Fe in the ocean.

Source of Radioactivity

Iron-55, formed by (n, 2n) and (n, γ) reactions on ^{56}Fe and ^{54}Fe, respectively (Rama, Koide and Goldberg, 1961), can be produced by any thermal neutron source. The two important neutron sources which produce ^{55}Fe in the environment are nuclear reactors and nuclear explosions. The large thermonuclear explosions provide many neutrons (~0.5 kg neutrons per megaton explosion) (Libby, 1956) and thus form large amounts of ^{55}Fe in the structural material of thermonuclear devices. Much greater production of ^{55}Fe occurred in the recent large thermonuclear explosions than in the earlier relatively small atomic explosions of the 1940's. Measurements of

^{55}Fe near the Columbia River show that the nuclear reactors at Hanford do not contribute ^{55}Fe even at the level observed in fallout and may be neglected as an insignificant source of ^{55}Fe in the ocean. Apparently, nuclear testing, to which we now turn our attention, is responsible for most of the ^{55}Fe in the North Pacific Ocean.

Two periods of recent testing by the major nuclear powers, the United States and Russia, took place during 1952-58 and 1961-62. The first of these periods of testing ended in 1958 with a moratorium on testing, which continued for about three years until terminated by a Russian weapon test in 1961. The second series of major testing was halted by the signing of the Nuclear Test Ban Treaty in 1962. The amount of fission was roughly equivalent in the 1952-58 series and the 1961-62 series but appeared at an increased rate in the more recent tests (Peirson and Cambray, 1965).

Russian testing during 1961-62 was carried out in the Arctic (75°N., 55°E.) and in Central Asia (52°N., 78°E.) while the American explosions in 1962 were at Johnston Island (17°N., 169°W.) and Christmas Island (2°N., 157°W.) (Peirson and Cambray, 1965).

The major trajectory for the debris from the Russian tests was in a west to east direction, although it was frequently subject to large disturbances. Nuclear debris reaching the British Isles from the Russian tests made nearly a complete circuit of the hemisphere in an interval of time between 12 and 25 days (Perison and Cambray,

1965).

This general west to east circulation pattern in the high latitudes of the northern hemisphere suggests that much of the ^{55}Fe in the North Pacific is contamination from the Russian tests. Support for this idea comes from the observation of Peirson and Cambray (1965) that no short-lived activity from the American explosions in the tropics was observed in the United Kingdom. Apparently, fallout from the American tests does not quickly reach the high latitudes of the northern hemisphere.

Additional support for the contention that the Russian tests were the major contributors to ^{55}Fe in the North Pacific can be found by comparing tropospheric and stratospheric fallout. Libby (1956) places the limiting time for wash out of most of the radioactivity in the troposphere by precipitation at a matter of weeks following a nuclear explosion. On the other hand, stratospheric fallout is deposited only at a very slow rate corresponding to an average time in the stratosphere of about 10 years.

> As a result of this long residence time in the highest layers of the atmosphere, the winds mix and distribute the radioactive material broadly over the earth and one finds, when the fallout does finally find its way down into the troposphere where rain and snow wash it out, that the rates of precipitation are relatively uniform over the entire earth's surface (Libby, 1956, p. 658).

While evidence points to the fact that stratospheric fallout is highest at mid-latitudes due to breaks in the tropopause (Mauchline

and Templeton, 1964), in a first approximation stratospheric fallout is uniformly distributed and would not seem to account for the high ^{55}Fe occurring in Alaskan waters.

Purpose of the Study

During and following the American tests of the 1950's several surveys of the environmental radioactivity in the Pacific Proving Ground were made (Donaldson et al., 1956; Seymour et al., 1957). In these and other studies prior to resumption of testing in 1961, ^{55}Fe was detected in water (Lowman, 1963a), soil (Palumbo and Lowman, 1958), plankton (Lowman, 1958; Lowman, 1963b), fish (Lowman, 1963b; Lowman, Palumbo and South, 1957; Rama, Koide and Goldberg, 1961) and in other marine organisms (Lowman, Palumbo and South, 1957). Iron-55 contributed a significant portion of the total radioactivity in each of these studies, contributing in one case 74% of the total (Lowman, Palumbo and South, 1957).

Since the 1962 tests, measurements of ^{55}Fe have been made in terrestrial food chains (Jaakkola, 1967; Persson, 1967; Miettinen, 1967; Palmer and Beasley, 1965) and in marine organisms (Palmer and Beasley, 1965, 1967a, b; Palmer, Beasley and Folsom, 1966; Wrenn and Cohen, 1967). These workers report that the highest levels of ^{55}Fe measured in the biosphere have been in ocean organisms and that salmon liver contained the highest ^{55}Fe activity of any

organism (Palmer and Beasley, 1967a, b).

The ^{55}Fe specific activity of salmon liver in 1965 (Palmer and Beasley, 1967a) was about 300 times as high as the ^{55}Fe specific activity of tuna liver reported in 1961 (Rama, Koide and Goldberg, 1961).

It is apparent, then, that ^{55}Fe in biological organisms is higher in the marine environment than in the terrestrial environment, with highest levels occurring after the most recent test series.

A radiological survey in the 1950's near the American test site in the Pacific showed only ^{55}Fe to have a level greater than the maximum permissible concentration in water for human consumption (Lowman, Palumbo and South, 1957). Although the maximum permissible concentration for water cannot be used directly for food items unless the diet of the individual under consideration is known, it does point to the relative importance of ^{55}Fe compared to other fallout nuclides.

Despite the fact that ^{55}Fe is probably the most abundant fallout radionuclide presently contaminating the environment (Palmer and Beasley, 1967b) and an isotope of a biologically important trace element, appallingly little is known about its activity levels or its spatial and temporal variations in the marine environment. No one has reported on ^{55}Fe in a specific organism over a geographical area or across several years. Owing to the half life (2.7 years) of ^{55}Fe

and to its large initial input into the environment, it is still possible to measure easily ^{55}Fe in samples several years old and this has been undertaken in this study.

The answer to why ^{55}Fe has been largely neglected probably lies in detection difficulties. The soft (5.9 keV) X-ray by which it is detected requires that it be chemically separated before counting. By contrast, γ-emitters in fallout, can be evaluated instrumentally with no chemical separation involved, and have been more widely studied than ^{55}Fe.

In this study ^{55}Fe is measured in several marine organisms throughout the North Pacific Ocean with special emphasis given to Pacific salmon samples collected from Bristol Bay, Alaska, in the north to Eureka, California, in the south and to Japan on the west (see Figure 2).

The purposes of this research are set forth below:

1. To study the location of ^{55}Fe once it is introduced into the environment. Does it remain in the surface waters, being recycled among biota, or is it soon lost from the upper regions of the ocean?
2. To observe the effect on ^{55}Fe specific activity in ocean organisms of a high local input of stable iron from the Columbia River.
3. To evaluate the temporal changes in total ^{55}Fe in the North

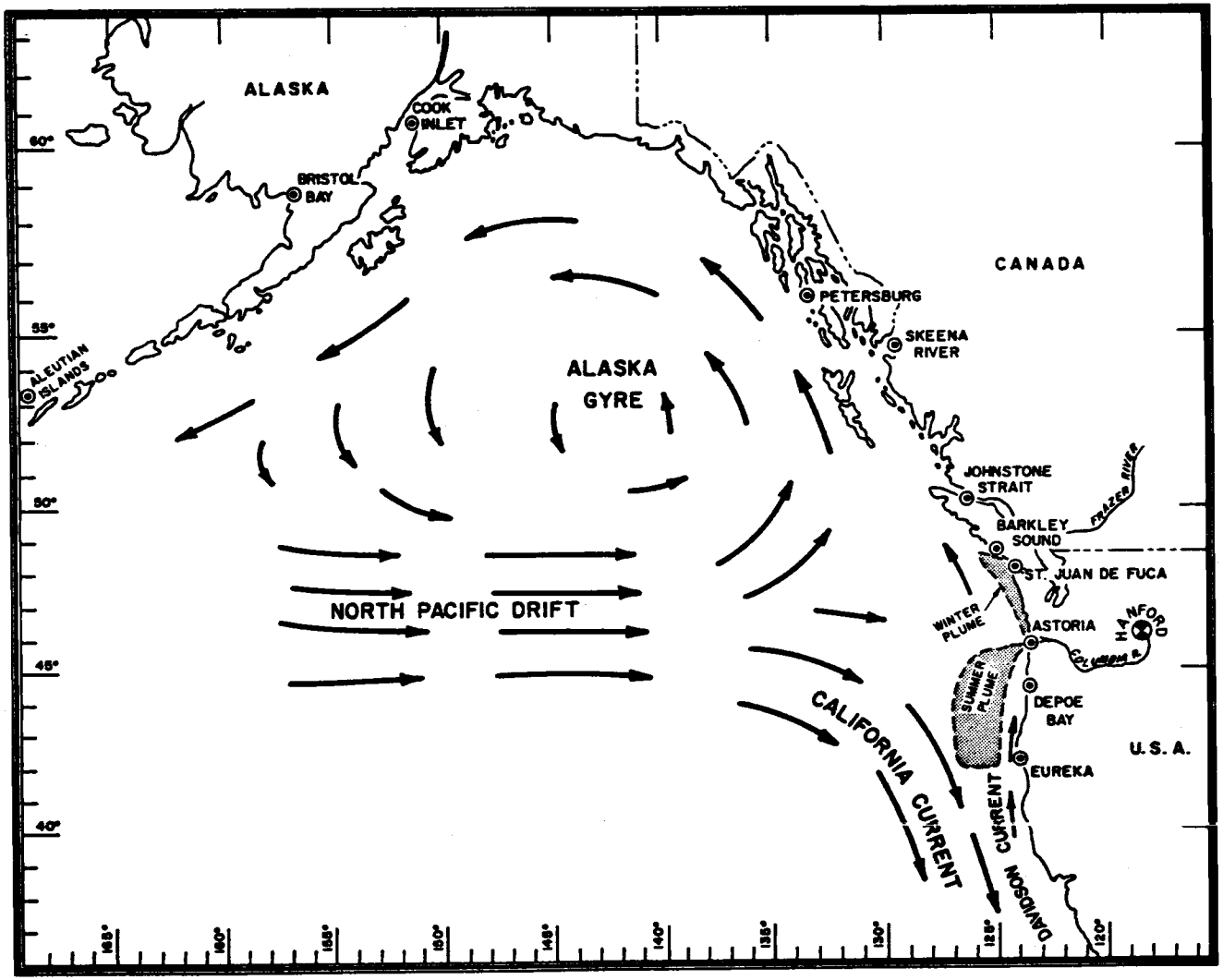


Figure 2. Northeast Pacific Ocean and collection sites of salmon samples (after Kujala, 1966).

Pacific Ocean from 1964 to the present time and to relate this to the total input of ^{55}Fe into the environment.

4. To unearth problems of distribution of ^{55}Fe which may be suitable for further study.

Specific Activity

The term specific activity refers to the ratio of isotopes of an element and may be defined for any radionuclide. The following definition for specific activity of ^{55}Fe serves as an example:

$$\text{Specific activity of } ^{55}\text{Fe } \left(\frac{\mu\text{Ci}}{\text{g}}\right) = \frac{\text{Activity of } ^{55}\text{Fe } (\mu\text{Ci}) \text{ in sample}}{\text{Total Fe (g) in sample}}$$

Specific activity is particularly important in considering the maximum allowable amount of a radionuclide present in the environment. The reasoning behind this approach is that an organism will ingest all isotopes of an element in the same proportion as they occur in the food or water source of that element, that is, isotopic fractionation will not occur. This assumes that all isotopes are in the same chemical and physical forms.

With this approach the problem of determining maximum permissible concentrations of a radionuclide in the ocean can be reduced and simplified, and stated as follows:

> If the specific activities of the elements of the sea in the region of growth, development and habitation of marine food organisms can be maintained below the allowable specific activities of these elements in man and his seafood, the allowable radiation for any individual cannot be exceeded as a result of the consumption of marine products. . . . It should be clearly noted that this approach is not primarily affected by proclivities of most marine organisms of concentrating radioisotopes, for if the organism concentrates a radioisotope, it also concentrates the stable element. Thus it does not alter the specific activity of the material taken from the environment (NAS-NRC, 1962, p. 22).

Specific activities among individual organisms have been found to vary less than either activity per unit weight or total element per unit weight (Renfro, 1968) and would seem to more consistently reflect the uptake of the radionuclide by the organism.

For these reasons, the specific activity rather than activity per unit weight of ^{55}Fe is emphasized in this study.

Iron in the Marine Environment

Iron-55 in the marine environment should be discussed relative to the physical and chemical forms of iron in the sea and relative to the forms in which it can be utilized by marine plants and animals. A background discussion of these topics at this point sets the stage for later interpretation of ^{55}Fe results.

Cooper (1948) classified the forms of iron in sea water as follows:

A. Uniformly dispersed iron
   1. Ionic iron
   2. Dissolved inorganic complexes
   3. Dissolved crystalloidal or dispersed colloidal organic compounds
   4. Colloidal inorganic compounds

B. Aggregated or particulate iron
   1. Discrete flocculent particles of ferric hydroxide or phosphate
   2. A unimicellar film of ferric hydroxide covering a considerable part of the sea surface
   3. Ferric hydroxide or phosphate adsorbed upon the surface of organic or inorganic detritus or upon living phytoplankton
   4. Iron in fecal pellets
   5. Organically bound iron in the cells and tissues of living and dead plants and animals
   6. Terrigenous iron in clay and suspended mineral matter derived from the land.

Only the main divisions, uniformly dispersed iron and aggregated or particulate iron, can be easily evaluated experimentally. The distinction between these two types is arbitrarily dependent upon the porosity of the filter utilized for the separation (Lewis and Goldberg, 1954).

Early measurements of iron in sea water (Cooper, 1935; Harvey, 1937a) showed dissolved iron in very low concentration, less than 2 ppb, their limit of detectabilities. On the other hand, particulate iron, although widely variable, was found to be as high as 100 ppb.

Cooper (1937) calculated the equilibrium amount of iron in true solution as 4×10^{-7} ppb at a pH of 8.0 although he himself stated that equilibrium is but slowly attained.

> In an azoic world, given sufficient time, thermodynamic equilibrium would no doubt in the end be reached, whereas in temperate surface waters of the sea, the speed of life cycle is likely never to allow equilibrium conditions to be even remotely approached (Cooper, 1948, p. 315).

In a more recent study of iron in the sea, Lewis and Goldberg (1954) identified particulate iron as that which is retained on a Millipore Type HA filter (0.45μ) while they designate iron passing through the filter as soluble iron. With this definition, soluble iron is found at a concentration of about 3.5 ppb at all depths, only slightly less than particulate iron at 4.5 ppb in waters deeper than 500 m. Nearer the surface particulate iron is often higher and shows a large amount of scatter. The soluble iron concentration is far greater than the allowable ionic iron at equilibrium with the constituents of sea water. The explanation of the discrepancy between observed and calculated soluble iron given by Lewis and Goldberg (1954) is that soluble iron found in the ocean likely consists of dissolved or colloidal organic compounds which arise from the marine biosphere and colloidal inorganic compounds such as ferric hydroxide and ferry phosphate with sizes less than 0.5μ. This is not surprising since iron is a transition element and exhibits a strong tendency to form complexes with organic material (Lowman, 1963a).

Iron is an important nutrient and apparently a low supply results in a reduced growth rate of phytoplankton (Harvey, 1963). The physiological function of iron in plants (Cooper, 1935) and animals (Lowman, 1963a) is carried out only when iron is in the ionic form. Ionic iron, as previously pointed out, is in very short supply in the sea. In fact, Harvey (1937a) has calculated that there is little more ionic iron in a cubic meter of sea water than is found in a single diatom of moderate size. He further states that the amount of iron taken up in normal diatom growth is over 10^4 times the amount which they could obtain from ionic iron.

Two questions about uptake and utilization of iron by marine organisms, then, are (1) what forms of iron can be taken up, and (2) how are these converted into a useable form?

In answering the first question, Goldberg (1952) found that organic iron complexes were not available as a growth nutrient to marine diatoms, whereas particulate and/or colloidal forms of iron were taken up by marine diatoms. Apparently, diatoms can dissolve and reduce particulate and colloidal iron to a useable form.

One proposed mechanism (Harvey, 1937b) for utilization of particulate iron is that the particles are adsorbed to the surfaces of phytoplankters which have many small pores in the skeleton, exposing protoplasm. The cell sap is acidic so that solution of the particles and their passage into the diatom is possible.

In summary, iron is metabolically effective only in ionic form but is taken up from marine waters in colloidal or particulate form and then dissolved and reduced to the effective ionic form. It is possible that animals, as well as plants, can utilize particulate iron directly from sea water, although there is no evidence to support this.

Terrestrial waters have a much higher iron content than marine waters, and increases in iron concentration in sea water near river mouths have been measured. Both particulate and soluble iron were seen to decrease seaward from the Fraser River in British Columbia (Williams and Chan, 1966) and particulate iron was found to be high in the Columbia River plume (Joyner, 1964). The effect of this high local input of iron upon ^{55}Fe uptake will be discussed later.

PROCEDURE

Since ^{55}Fe decays by electron capture and is detected by its 5.9 keV X-ray, counting a sample in a matrix of biological material--such as is often done in gamma-ray analysis--is impossible because of self-absorption of the X-ray. To reduce self-absorption the iron must be separated from the biological sample and distributed in a thin, uniform layer on a planchet for counting. In short, this is accomplished by dissolving the biological sample in 6 M hydrochloric acid, extracting the iron from the sample with 10% v/v Alamine-336 in xylene, and then electroplating the iron on an 11.5 cm^2 copper disk.

In order to obtain ^{55}Fe specific activity (^{55}Fe activity/weight of total Fe), the samples were also analyzed colorimetrically for stable iron using 1,10 phenanthroline.

Specific procedures for carrying out each of the steps of analysis are discussed below. All chemicals used in these procedures were of reagent grade.

Sample Preparation

The biological samples studied in this project were collected by various means and were handled in slightly different ways. Plankton and nekton (except salmon) samples were taken from

Isaac-Kidd midwater trawl hauls and benthic organisms were collected with an otter trawl. Salmon viscera samples were graciously provided by commercial fishermen who fish the coastal waters from Eureka, California, in the south to Bristol Bay, Alaska, in the north. Addition of formalin preserved the samples until the time of laboratory analysis.

In the laboratory, samples were dried in a vacuum furnace at 125°C to constant weight and were then ashed in a muffle furnace at 600°C for at least 24 hours. Wet weight (after blotting the formalin-preserved sample), dry weight, and ash weight records were kept for each sample.

A convenient portion of the ashed sample, usually 2 g if available, was then dissolved in 6 M hydrochloric acid in a 100 ml volumetric flask. A 25 ml aliquot of the sample was pipetted into a 50 ml volumetric flask and retained for stable iron analysis and the remaining 75 ml of sample was retained for ^{55}Fe analysis.

Throughout the processing of the sample it was often necessary to transfer the sample from one container to another, for example, from beaker to volumetric flask and from volumetric flask to separatory funnel. In each of these transfers, and in the filtration step, care was taken to rinse several times with 6 M hydrochloric acid to guard against loss of sample. It is also worth noting that the aliquot for stable iron analysis was not taken until after the sample

was dissolved, so that any loss of iron in dissolution should equally affect stable iron and ^{55}Fe leaving the specific activity unaffected.

Iron Extraction

Solvent extraction was chosen to separate the iron from interfering ions. The solvent extracts the iron from the aqueous phase, selecting against calcium and phosphate ions which occur in rather large quantities in biological samples and which interfere with electrodeposition of iron (Maletskos and Irvine, 1956). A long-chain tertiary amine, Alamine-336, was chosen as the agent for extracting the iron. Use of Alamine-336 in xylene for extracting iron (III) from a 6 M hydrochloric acid solution of biological material has been previously reported (Palmer and Beasley, 1965). Coleman (1963) showed that the maximum extraction coefficient of iron (III) from hydrochloric acid by Alamine-336 occurs as a broad maximum in the 3 M to 9 M hydrochloric acid range with a slightly higher peak at 6 M. The value for the extraction coefficient in this range is about 1000 for iron (III).

Percent extraction is given by

$$\% \text{ extraction} = \frac{D}{D + (Va/Vo)} \times 100$$

where D is the extraction coefficient and Va and Vo are the volumes of the aqueous and organic phases respectively (Morrison

and Freiser, 1957). From this equation the percent extraction of iron (III) from 6 M hydrochloric acid by Alamine-336 in xylene was calculated to fall between 99.0 and 99.9 percent depending on the Va/Vo ratio.

Iron was extracted from the dissolved biological sample with 15 ml of 10% v/v Alamine-336 in xylene in a 125 ml separatory funnel. The aqueous phase was discarded and the organic phase was scrubbed with 6 M hydrochloric acid to remove traces of calcium and phosphate. Next the iron was stripped from the organic phase by contacting with an equal volume portion of 1 M perchloric acid. The aqueous phase was saved in a 100 ml test tube along with a second portion of 1 M perchloric acid used to scrub the organic phase.

The iron was precipitated as ferric hydroxide by addition of concentrated ammonium hydroxide. If it was judged by the amount of precipitate present that the samples contained less than 4-5 mg of iron, 5 mg of iron carrier were added. The sample was then centrifuged and the supernatant drawn off by suction. Finally, the sample was prepared for electroplating by washing the precipitate with water, centrifuging and again drawing off the supernatant.

Electrodeposition of Samples

Quantitative electrodeposition of iron from samples which contain only small amounts of iron (3-20 mg) is possible using

techniques developed by Maletskos and Irvine (1956). Their methods, with some modification of the apparatus, were employed in this study.

The electroplating cell (Figure 3) consisted of a stainless steel cell base notched to hold a copper disk which served as the cathode, the site of iron deposition. A plexiglass tower, held in place on the cell base with a brass nut and sealed to the copper disk with a neoprene 0-ring, contained the sample solution.

Platinum wire (~0.4 mm), bent in an L-shape and connected to a small electric motor, served both as stirring rod and anode. Stirring speed varied throughout the plating process, so each motor was separately controlled with an autotransformer to prevent spillage from rapid stirring.

Electric connection to the anode was made by copper brushes set into an insulating material mounted on the motor support rod. The anode was connected to the motor shaft by an insulating sleeve of teflon.

A schematic diagram of the power supply for the electroplating unit is shown in Figure 4.

To make the iron adhere properly, the copper disk was washed with acetone to remove grease, soaked in 2 M nitric acid to remove oxide coating, and rinsed with distilled water. After drying, the disk was weighed and positioned in the cell in preparation for sample electrodeposition.

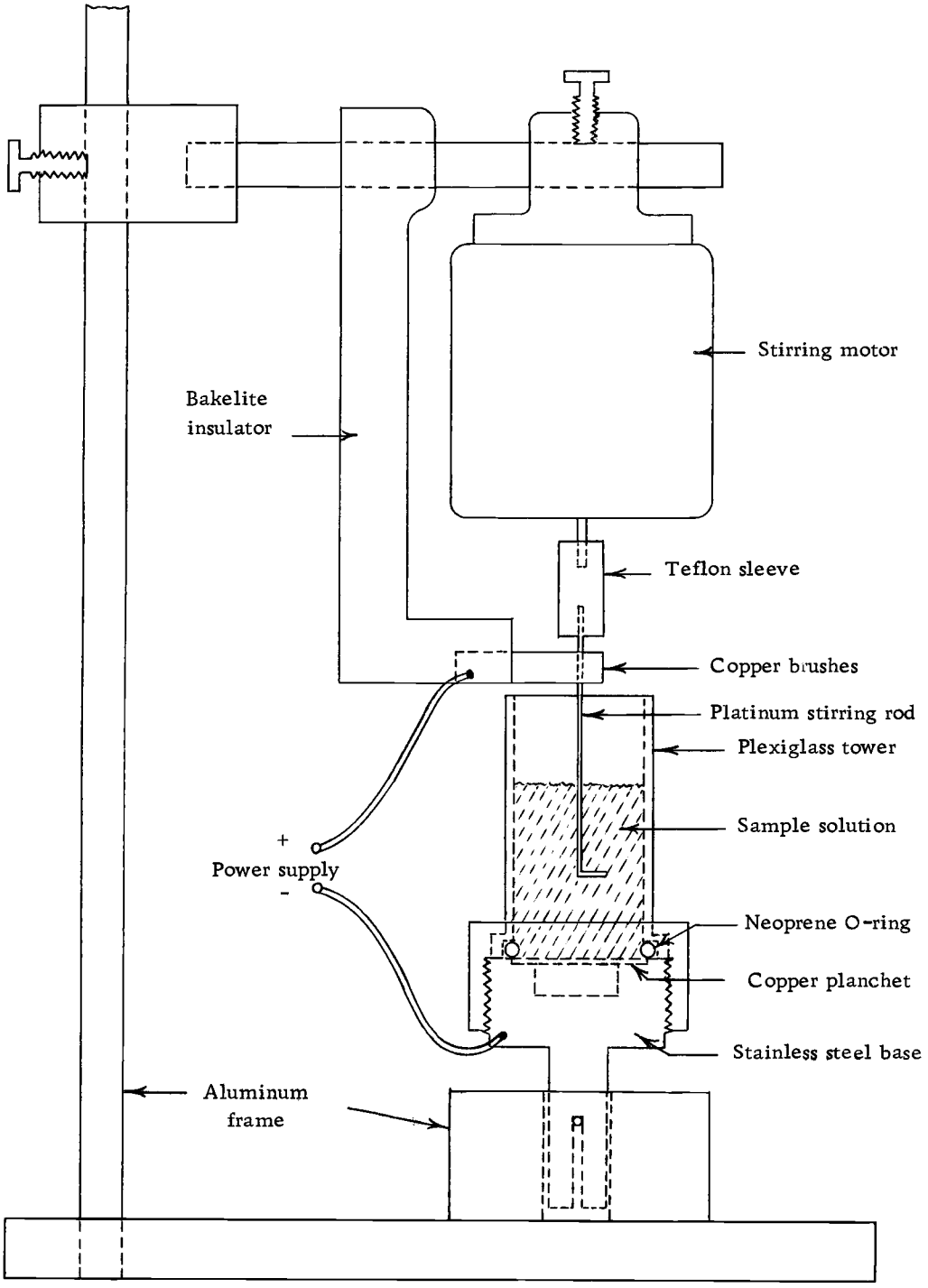
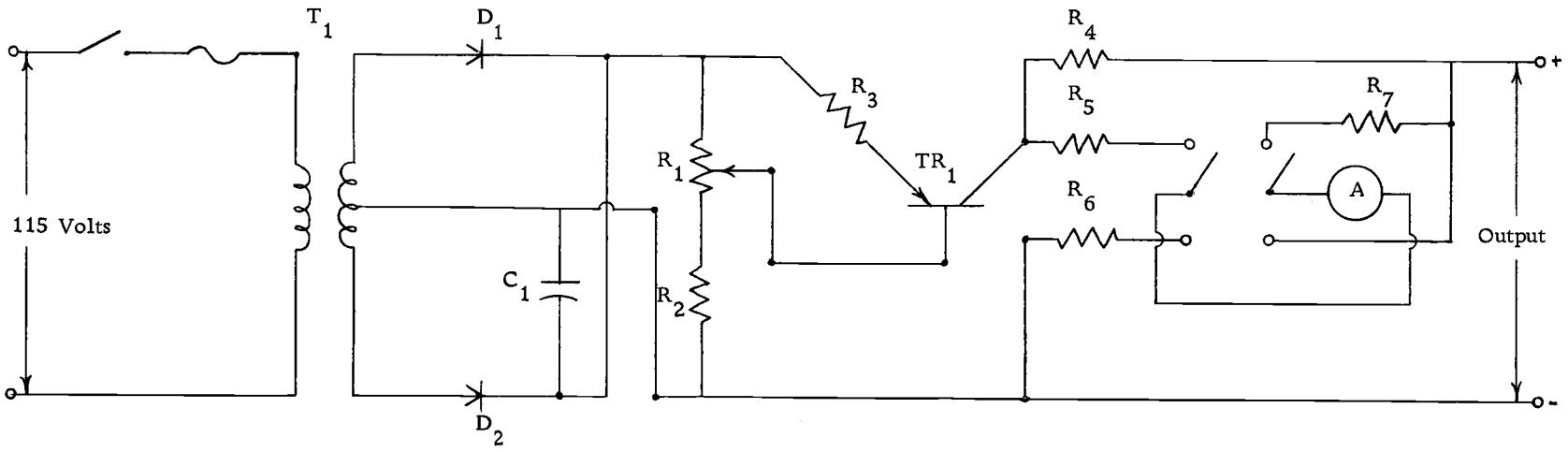


Figure 3. Cross-section of electroplating cell.



A - 0-20 microammeter

R_1 - 15 KΩPotientiometer 2 watt

R_2 - 100 Ω 1/2 watt

R_3 - 2 Ω #18 Nichrome Handwound

R_4 - 1Ω #18 Nichrome Handwound

R_5 - 50 KΩ 1/4 watt

R_6 - 470 KΩ 1/4 watt

R_7 - 50Ω potientiometer 2 watt

T_1 - Transformer 110/240 primary, 16/32 secondary 50/60 cycles, 500 volt ampere

D_1, D_2 - 1N 1185

TR_1 - 2N 174

C_1 - 4000 mf 50 V

Meter calibrated to read 0-2 amperes and 0-10 volts.

Figure 4. Schematic diagram of power supply used for electrodeposition of iron.

Each sample, precipitated as ferric hydroxide as described previously, was dissolved in 0.4 ml of 6 M sulfuric acid and 5 ml of saturated ammonium oxalate and transferred to the plating cell. The sample test tube was rinsed with three 10 ml portions of saturated ammonium oxalate, also added to the plating cell. Electrodeposition took place at 0.85 amp and 8-10 V for about 1.5 hours (Table 1). After about 0.75 hour the plating solution, having become basic during the plating process, was adjusted to a pH of 7 by dropwise addition of 6 M sulfuric acid.

Table 1. Procedures for electroplating (Maletskos and Irvine, 1956).

Solution composition	$Fe(OH)_3$ from water or biological sample 0.4 ml 6 M H_2SO_4 35 ml saturated $(NH_4)_2 C_2O_4$
Current (amperes)	0.85
Voltage (volts)	8-10
Plating time (hours)	1.5
Initial pH	4
Final pH	7
Initial solution color	Yellowish-green
Final solution color	Colorless

At 1.5 hours the sample was tested for completeness of deposition (Table 2). If less than 1 μg of iron remained, the disk

was quickly rinsed with distilled water, then dried and weighed. Amount of iron deposited, given by the difference between final and original weight of the disk, allowed a self-absorption correction to be made for the 5.9 keV X-ray. If more than 1 μg remained, the sample plating was continued for additional 15 minute intervals until less than 1 μg of iron remained in the sample.

Table 2. Test for completeness of electroplating (Maletskos and Irvine, 1956).

Solution composition	1 ml plating solution
(in small test tube)	2 drops 1 M $NaHSO_3$
	2 drops 10 g/l α, α' -dipyridyl (in 0.5 M HCl)
	Heat to boiling
	1 drop 1.25 M Na_2S
	Shake well
Color in 1-10 μg Fe Range	Light pink to red
Color when < 1 μg Fe	Colorless

Counting of ^{55}Fe

Prior to counting of the 5.9 keV X-rays of ^{55}Fe by the method of Palmer and Beasley (1967b), the samples, deposited on copper planchets, were covered with plastic wrap to protect them from

oxidation and to prevent ^{55}Fe contamination of the counter. Wrapped samples were counted by positioning them on the thin (0.0038 mm) aluminized Mylar entrance window of an argon-methane gas flow proportional counter. To reduce the background counting rate, the counter was placed between two 4 in. x 9 3/8 in. (10 cm x 24.4 cm) NaI (Tl) scintillation crystals which acted as anticoincidence shields (see Figure 5). The background of the counting system was one count per minute under the 5.9 keV photopeak. Similar background rates were found when plastic scintillators were substituted for the NaI (Tl) crystals as anticoincidence shields. The minimum amount of ^{55}Fe detectable with this system is 0.008 nCi at 99% confidence in a one-hour count.

Total Iron Analysis

Analysis of total iron in each sample was carried out colorimetrically by an adaptation to biological samples of the 1,10 phenanthroline method for measuring iron in natural water (APHS, 1960).

Determination of the amount of iron in solution by absorption spectrometry is based on the characteristic absorption of ultraviolet and visible light by the iron chelate in solution. When a monochromatic light beam is passed through a cell containing a solution of the complexed iron, the absorbance is proportional to the number

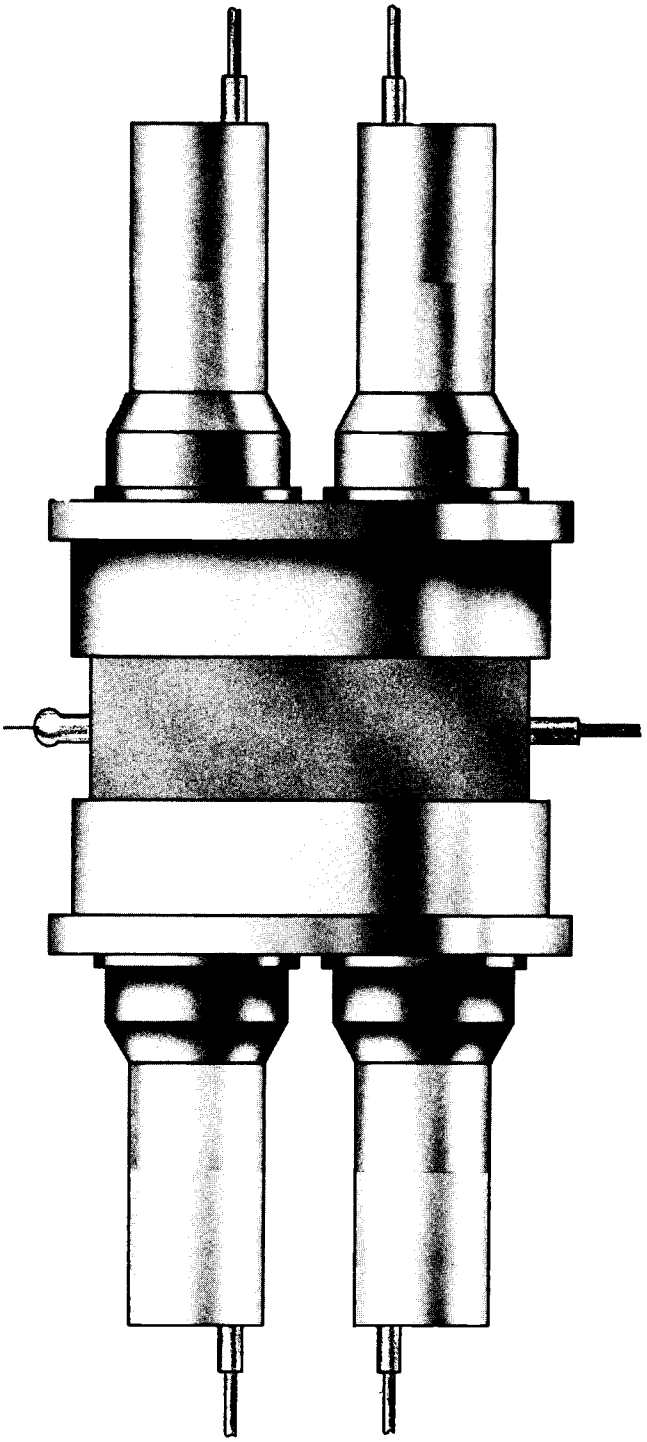


Figure 5. Iron-55 counting arrangement (after Palmer and Beasley, 1967 b).

of molecules in the path length (Skoog and West, 1963).

As previously mentioned, 25 ml of the original 100 ml sample were retained for total iron analysis so that specific activity could be determined. The sample was diluted to 50 ml in a volumetric flask with distilled, deionized water and then 5 ml of this dilution were placed into a 100 ml volumetric flask for the total iron analysis so that only 1/40 of the initial sample was utilized in total iron analysis. Dilution was necessary to insure that the concentration would fall in the 0.5-4 mg/l range which could be accurately determined by this technique. Since the samples were diluted considerably for the 1,10 phenanthroline technique, all glassware used in this analysis was washed in 6 M hydrochloric acid to remove traces of iron.

To the 100 ml volumetric flask containing 5 ml of the sample to be analyzed, were added 2 ml of 20% hydroxylamine hydrochloride and about 50 ml of distilled, deionized water. Boiling this solution for about 30 minutes reduced the iron from the ferric to the ferrous state which formed complexes with the 1,10 phenanthroline. After boiling, the color was developed by adding 10 ml of ammonium acetate-acetic acid buffer (pH 4.2) and 2 ml of 0.5% 1,10 phenanthroline. The sample was then diluted to 100 ml with distilled, deionized water and allowed to stand for at least 30 minutes for the color to develop completely before reading on a Beckman model DU

spectrophotometer at 508 mμ.

Initially a calibration curve was run by adding 0.00, 0.05, 0.10, 0.20 and 0.50 mg of iron, respectively, to five equal portions of a dissolved salmon sample. A plot of absorbance on the ordinate versus quantity of iron added on the abcissa yielded a straight line Beers Law plot. Three standards, without biological material added, were run with each set of samples, and were always found to fall on the initial calibration curve. This curve was used to relate the absorbance of the samples to iron concentration. An appropriate factor, usually 40, was then used to correct the concentration back to total iron per sample.

Iron Recovery

Some of the steps involved in sample treatment have either a theoretical or experimental check on quantitative iron recovery. For example, electrodeposition, which has been reported to be quantitative (Maletskos and Irvine, 1956), has a colorimetric check performed near the end of the electrodeposition of each sample to assure that almost none (< 1 μg) of the iron remains in the plating solution. Also, previous calculations (see Iron Extraction) showed that 99.0 to 99.9 percent of the iron is retained in the extraction step. Each of these checks supports the contention that iron recovery is quantitative in this procedure. However, there is considerable

handling of the samples and although care was taken in each solution transfer, the possibility of some iron loss prompted an iron recovery experiment to check the entire procedure.

The experiment was also designed to check the maximum amount of iron which could be extracted with 10 ml of the 10% Alamine-336 in xylene used for these samples. This was accomplished by adding increasing amounts of iron carrier to successive samples.

The procedure followed in this experiment conformed to the techniques used in actual sample analysis and thus gave total recovery of iron throughout the process.

From a well-mixed sample of ashed biological material eight 0.5 g samples were weighed, placed into separate beakers and treated with ^{59}Fe tracer and stable iron carrier as shown in Table 3. The eight samples were carried through the normal procedure using 10 ml instead of the normal 15 ml of 10% Alamine-336 in xylene in the extraction step. The ^{59}Fe in the samples was counted in 13 ml counting tubes before the electrodeposition step and only samples showing quantitative recovery (one through four) were electroplated. An ^{59}Fe standard was also prepared to use for a reference counting rate and was not carried through the sample treatment process.

The results of this experiment (Table 3) show that the procedure

Table 3. Iron recovery experiment.

Sample	Biological matrix (g ash)	Iron carrier (mg)	^{59}Fe Tracer (relative units)	% Recovery before plating	% Recovery after plating
^{59}Fe Spike	---	8.36	1	---	---
#1	0.5	8.36	1	101.6 (8.49 mg)	100.9
#2	0.5	16.72	1	106.6 (17.8 mg)	102.3
#3	0.5	33.44	1	106.1 (35.5 mg)	96.4
#4	0.5	50.16	1	99.1 (50.1 mg)	99.8
#5	0.5	66.88	1	82.7 (55.3 mg)	---
#6	0.5	83.60	1	69.5 (58.1 mg)	---
#7	0.5	100.32	1	56.9 (57.1 mg)	---
#8	0.5	117.04	1	45.4 (53.1 mg)	---

is quantitative for 50 mg of iron or less. Above 50 mg the percent recovery decreases, with approximately 50 mg being recovered regardless of the excess amount of iron present over 50 mg. This loss of percent recovery is due to loading of the Alamine-336 in the extraction step. This could be observed during the experiment by the yellow color of the ferric chloride complex which remained in the aqueous phase of samples four through eight, indicating that not all the iron was extracted.

RESULTS

Five species of Pacific salmon constitute the largest segment of samples analyzed in this study. Salmon undergo wide migrations and thus their ^{55}Fe specific activities represent a biological integration of radioactivity from the waters in which they have lived. Also, the ^{55}Fe in sea cucumber samples collected off the Oregon coast is indicative of the ^{55}Fe reaching the bottom, and that in surface plankton represents the ^{55}Fe present *in situ* off the Oregon coast. In the same way, the ^{55}Fe in Alaskan lichen samples gives a measure of the total ^{55}Fe present in the North Pacific system from airborne fallout.

Salmon

Salmon samples analyzed in this study consisted of salmon viscera collected by commercial fishermen in the coastal waters and high seas of the Pacific Ocean. Collection sites are shown in Figure 4. Specific activity was measured either in the entire viscera or in separate organs. Data to be presented later will show that ^{55}Fe specific activity is relatively constant throughout the fish so that the results are similar whether the entire viscera or separate organs are analyzed.

Sockeye

Sockeye salmon, O. nerka, present quite a different picture of ^{55}Fe specific activities from chinook samples (Table 5). With the exception of Bristol Bay samples, which show specific activities nearly equal to those in chinook, the sockeye samples have specific activities at least an order of magnitude higher than those in chinook at the same station.

Table 5. Specific activity of ^{55}Fe (μ Ci/g Fe) in sockeye salmon.

Location	No. individuals	1964	No. individuals	1965	No. individuals	1967
Bristol Bay, Alaska	19	11.4	3	4.86	8	0.510
Cook Inlet, Alaska	3	12.3	2	9.30		----
Petersburg, Alaska	2	15.4	3	8.05		----
Skeena River, Canada	5	12.8		----		----
Barkley Sound, Canada	1	19.4		----		----
Str. Juan de Fuca, Canada	2	28.7		----		----
Astoria, Oregon		----		----	5	1.17
Japan high seas		----	3	5.15		----

The north-south trend of specific activities is not well defined in sockeye but appears to be the reverse of that seen in chinook, namely an increase in specific activity is seen while moving from north to south.

In 1965, one sockeye collected by Japanese fishermen on the

high seas was analyzed and its specific activity appeared little different from American sockeye samples.

Coho

Coho salmon, O. kisutch, like chinook, showed a progressive decrease in specific activity of ^{55}Fe in 1964 from Cook Inlet south to the Strait of Juan de Fuca with an increase at Depoe Bay and Eureka (Table 6). In general, however, the values of the ^{55}Fe specific activity are much higher in coho than in chinook. Since coho were not caught north of the Aleutian Chain at Bristol Bay it was not possible to compare the initial large drop in specific activity noted in chinook samples with coho samples.

Table 6. Specific activity of ^{55}Fe (μCi/g Fe) in coho salmon.

Location	No. individuals	1964	No. individuals	1965	No. individuals	1967
Cook Inlet, Alaska	2	16.1	2	7.10		----
Petersburg, Alaska	6	11.8		----		----
Skeena River, Canada	1	8.14		----		----
Str. Juan de Fuca, Canada	2	1.57		----		----
Astoria, Oregon		----		----	5	0.290
Depoe Bay, Oregon	6	5.72		----		----
Eureka, California	1	5.83	11	1.63		----
Japan high seas		----	1	4.88		----

As was the case with sockeye, Japanese high seas samples of coho did not appear to have specific activities significantly different from those collected in American waters.

Pink and Chum

Only a few samples of chum salmon, O. keta, and pink salmon, O. gorbuscha, were analyzed for ^{55}Fe specific activity (Table 7). Pink salmon appear to have ^{55}Fe specific activities intermediate between the high specific activity sockeye and the low specific activity chinook. While there are too few chum samples to speak with confidence, it appears from the two stations sampled that their specific activity may approach that of the high activity sockeye.

Table 7. Specific activity of ^{55}Fe (μCi/g Fe) in pink and chum salmon.

	Pink				Chum			
Location	No. individuals	1964	No. individuals	1965	No. individuals	1964	No. individuals	1965
Cook Inlet, Alaska	1	7.90	2	5.75		----		----
Petersburg, Alaska	2	7.73	2	4.91	1	15.1		----
Japan high seas		----		----		----	8	7.02

Comparison of Several Species at Single Stations

Owing to differences in migration routes and spawning times, rarely was a wide range of species collected at a single station. Petersburg, Alaska, yielded the widest range of species and data

from these samples, presented in Table 8, show a relatively complete picture of species differences.

Table 8. Specific activity of ^{55}Fe ($\mu Ci/g$ Fe) in five species of salmon at Petersburg, Alaska.

Species	Number individuals	Specific activity 1964
Chinook	4	1.16
Pink	2	7.73
Coho	6	11.8
Chum	1	15.1
Sockeye	2	15.4

The most notable feature of this comparison is the very low specific activity of chinook. Sockeye exhibit the highest ^{55}Fe specific activity but are not widely different from coho or chum.

As was previously mentioned, the changes in ^{55}Fe specific activity observed while moving in a north-south direction were not the same for all species. For example, chinook displayed a decrease in ^{55}Fe specific activity from north to south while sockeye showed an increase in the same direction. The result of this is that the relationship of ^{55}Fe specific activities measured in two species varied with different stations. At Bristol Bay, chinook and sockeye were about equal, but at Petersburg sockeye were over an order of magnitude higher than chinook.

Three species at the Strait of Juan de Fuca (Table 9) further emphasize this change. Here sockeye were much higher than both chinook and coho.

Table 9. Specific activity of ^{55}Fe ($\mu Ci/g$ Fe) in three species of salmon at Str. Juan de Fuca, Canada.

Species	Number individuals	Specific activity 1964
Chinook	2	0.745
Coho	2	1.57
Sockeye	2	28.7

Separate Organ Analyses

Specific activities of separate organs from single salmon are of interest because they give an indication of uniformity of distribution of ^{55}Fe throughout the organism. They are further of value because it is not always convenient to analyze all parts of an organism and these measurements show any fractionation of iron isotopes which occur in the organism.

Specific activities presented in Table 10 show fairly close agreement among separate organs of single salmon with the possibility of the liver containing a slightly higher ^{55}Fe specific activity than the other organs.

Table 10. Specific activity of ^{55}Fe (μ Ci/g Fe) in separate organs of Pacific salmon.

Location	Species	Organ	Specific activity of organ	Average specific activity of fish
Skeena River, Canada	sockeye	eggs	10.4	11.2
	sockeye	liver + stomach + pyloric caeca	11.9	11.2
Petersburg, Alaska	coho	gills	7.65	10.1
	coho	liver	13.1	10.1
	coho	pyloric caeca	10.2	10.1
	coho	stomach	9.38	10.1
Astoria, Oregon	chinook	eggs	2.41	2.54
	chinook	liver	3.01	2.54
	chinook	pyloric caeca	2.34	2.54
	chinook	stomach	2.41	2.54
Astoria, Oregon	coho	eggs	.312	0.295
	coho	flesh	.347	0.295
	coho	liver + pyloric caeca + stomach	.226	0.295

Sea Cucumbers

The ^{55}Fe specific activities of sea cucumbers off the Oregon coast were determined in order to measure the ^{55}Fe reaching the ocean bottom (Table 11). Samples were chosen from three depths representing continental shelf, continental slope and abyssal plain organisms. Also, the samples encompassed the three year period for which salmon samples were analyzed, giving opportunity to observe whether temporal variations of ^{55}Fe specific activity occur in benthic organisms as observed in salmon in the surface layer.

Table 11. Specific activity of ^{55}Fe (μ Ci/g Fe) in sea cucumbers collected off the coast of Oregon.

Depth (meters)	Species	No. individuals	Part of organism	Year of collection	Specific activity	Specific activity corrected to 1962
50	Stichopus californicus	6	whole	1964	0.003	0.005
150	Stichopus californicus	3	whole	1964	0.003	0.005
200	Stichopus californicus	3	whole	1964	0.028	0.047
200	Stichopus californicus	?	whole	1965	0.004	0.009
200	Stichopus californicus	1	whole minus gut	1966	0.001	0.003
200	Stichopus californicus	2	whole minus gut	1967	0.002	0.007
600	Laetmophasma fecundum	8	whole	1964	0.009	0.015
820	Laetmophasma fecundum	12	whole	1964	0.003	0.005
1000	Laetmophasma fecundum	7	whole	1964	0.008	0.013
800	Laetmophasma fecundum	11	whole	1965	0.003	0.006
800	Laetmophasma fecundum	8	whole minus gut	1965	0.001	0.002
800	Laetmophasma fecundum	8	gut	1965	0.003	0.006
2808	Paelopatides sp.	2	whole	1964	0.044	0.073
3000	Paelopatides sp.	6	whole	1965	0.004	0.009
2800	Paelopatides sp.	?	whole minus gut	1965	0.003	0.006
2800	Paelopatides sp.	?	gut	1965	0.007	0.015
2853	Paelopatides sp.	6	whole minus gut	1966	0.004	0.011
2810	Paelopatides sp.	6	whole minus gut	1967	0.005	0.018
2810	Paelopatides sp.	6	gut	1967	0.001	0.004

Tables 11 and 12 also contain ^{55}Fe specific activities decay corrected to 1962, the time of major tropospheric fallout from Russian nuclear tests.

Table 12. Specific activity of ^{55}Fe ($\mu Ci/g$ Fe) in sediments collected off the coast of Oregon.

Depth	Year of collection	Specific activity	Specific activity corrected to 1962
200 m	1964	no ^{55}Fe detectable	---
200 m	1965	no ^{55}Fe detectable	---
800 m	1964	3.89×10^{-4}	6.50×10^{-5}
600 m	1965	4.67×10^{-4}	10.1×10^{-4}
2800 m	1964	3.35×10^{-4}	5.60×10^{-4}
2800 m	1966	0.92×10^{-4}	2.57×10^{-4}

The most striking feature of these data is that sea cucumbers have specific activities 10^{-3} to 10^{-4} of those found in salmon. While there is variation of specific activities among the sea cucumbers, there does not appear to be a consistent trend with either depth or time.

Sediments

Low ^{55}Fe specific activities observed in sea cucumbers prompted determination of ^{55}Fe specific activity of selected sediments collected at the same stations as sea cucumbers (Table 12).

Sediments showed low ^{55}Fe specific activities comparing closely with sea cucumbers analyzed in this study and sea urchins analyzed by Palmer, Beasley and Folsom (1966).

Euphausids

Euphausids collected in the vicinity of the Columbia River plume were used to measure the in situ specific activity of ^{55}Fe off the Oregon coast. Since these euphausids presumably experienced various amounts of Columbia River water, they should also reflect the effect of the Columbia River plume on ^{55}Fe specific activity. The results of these analyses are presented in Table 13 and sampling stations are shown in Figure 6.

Lichen

Lichen samples collected in Alaska on a unit area basis were analyzed to determine the input of ^{55}Fe into the northern Alaska region (Table 14). Lichen is a slow growing mutualistic combination of algae and fungi which retains much of the radioactivity falling on it. Because it does not die off seasonally, radioactivity from several years' accumulation is retained by the lichen.

Iron-55 activity per square meter decreased from 1966 to 1967, but when corrected for decay back to the time of highest fallout in 1962 the activities were only slightly lower in 1967 than in 1966.

Table 13. Specific activity of ^{55}Fe (μCi/g Fe) in Euphausia pacifica.

Station	Location	Species	Date of collections	Specific activity	Specific activity corrected to 1962
1	46° 03.9'N 124° 19.8'W	E. pacifica	August 1967	0.007	0.025
2	46° 10.3'N 124° 28.9'W	E. pacifica	August 1967	0.036	0.130
3	46° 38.5' N 124° 38.7'W	E. pacifica	August 1967	0.101	0.365
4	45° 38.5'N 124° 17.8'W	E. pacifica	August 1967	0.030	0.108
5	45° 41.5'N 124° 28.9'W	E. pacifica	August 1967	0.016	0.058
6	44° 55.3' N 124° 31.9'W	E. pacifica	August 1967	0.066	0.238
7	45° 05.1'N 124° 47.5'W	E. pacifica	August 1967	0.080	0.289
8	46° 06.4' N 124° 19.4'W	E. pacifica	August 1967	0.053	0.191
9	45° 58.3'N 124° 27.3' W	E. pacifica	August 1967	0.072	0.260
Average				0.051	0.185

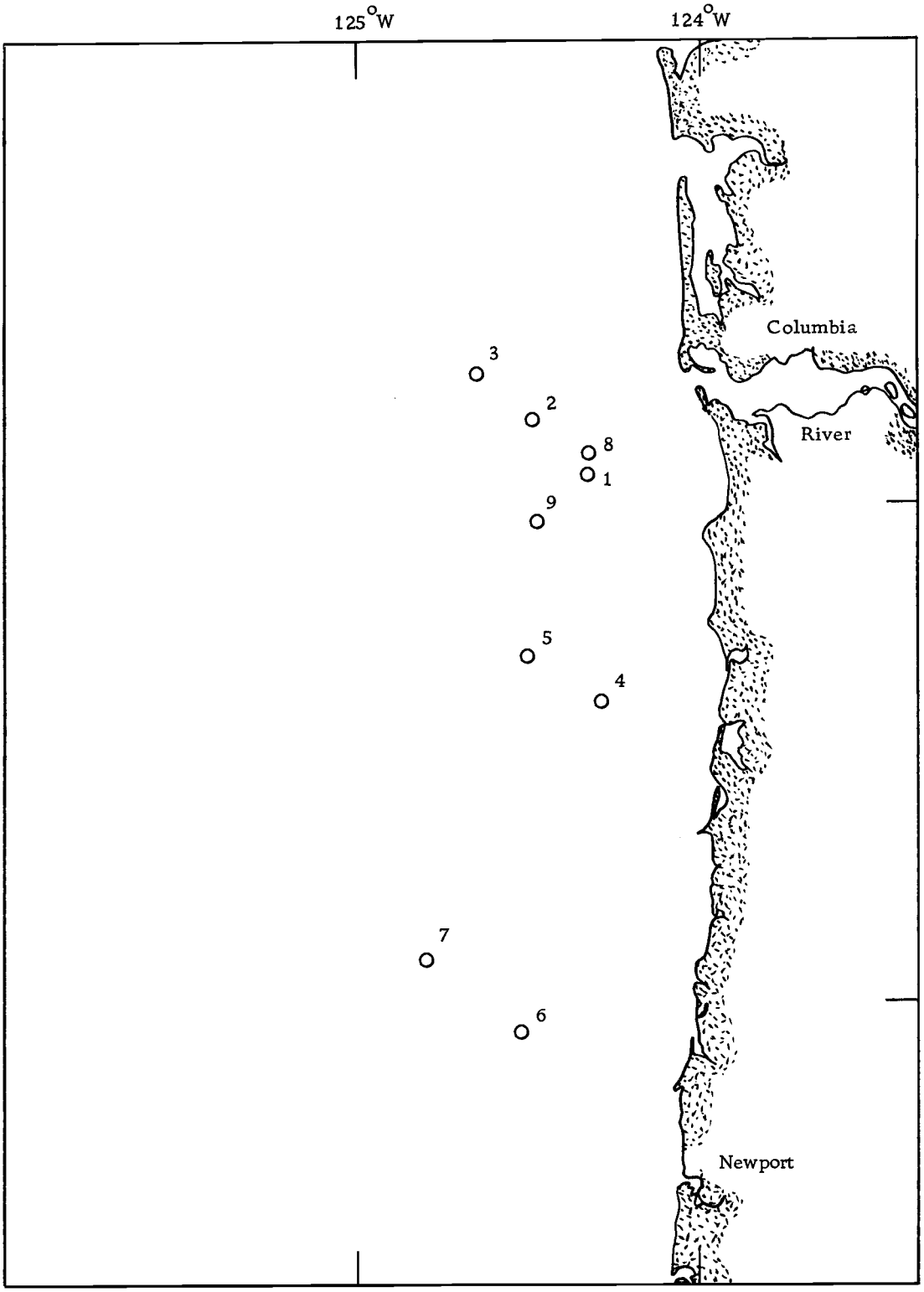


Figure 6. Sites of collection of euphausid samples. See Table 13 for data.

Apparently, the ^{55}Fe is held quite tenaciously by the lichen.

Table 14. Activity of ^{55}Fe in lichen.

Location	Species	Year collected	^{55}Fe at (nCi/m^2) collection time	^{55}Fe (nCi/m^2) corrected to 1962
Anaktuvuk Pass, Alaska	Cladonia rangiferina	1966	90.2	252
Anaktuvuk Pass, Alaska	Cladonia rangiferina	1966	68.6	192
Selawik River, Alaska	Cladonia alpestris	1966	56.4	157
Selawik River, Alaska	Cladonia alpestris	1966	116.2	324
Anaktuvuk Pass, Alaska	Cladonia alpestris	1967	47.3	171
Anaktuvuk Pass, Alaska	Cladonia alpestris	1967	51.3	185
Selawik Rive , Alaska	Cladonia alpestris	1967	55.6	201
Selawik River, Alaska	Cladonia alpestris	1967	52.5	190

Miscellaneous Samples

An assortment of other samples analyzed is presented in Table 15. Mussels collected at Bristol Bay and Petersburg contained levels of ^{55}Fe which are only a small fraction of the levels measured in salmon. A mackerel from Astoria had a specific activity similar to chinook collected during the same year. Steelhead from the Alsea River showed similarities to coho collected at Depoe Bay, not far from the mouth of the Alsea River.

Table 15. Specific activity of ^{55}Fe ($\mu Ci/g$ Fe) in miscellaneous samples.

Location	Date of collection	Sample	Number individuals	Specific activity
Bristol Bay, Alaska	1964	Mussels *Mytilus* sp.	6	0.0042
Petersburg, Alaska	1965	Mussels *Mytilus* sp.	21	0.0173
Astoria, Oregon	1964	Mackeral *Pneumatophorus japonicus*	1	0.918
Alsea, Oregon	1964	Steelhead trout *Salmo gairdnerii*	2	6.65
Dexter Dam, Oregon	1966	Chinook salmon *O. tshawytscha*	1	0.193

DISCUSSION

Before discussing the ^{55}Fe in the five species of Pacific salmon it will be valuable to review the sources of ^{55}Fe in the North Pacific Ocean and the habits of the Pacific salmon.

As stated earlier, most of the ^{55}Fe in the North Pacific probably came from tropospheric fallout from the Russian nuclear tests in 1961-1962. Such a more or less instantaneous input into the surface layer will be rapidly distributed throughout the mixed layer above the permanent thermocline which occurs at about 100 m (Wooster and Ketchum, 1957). Revelle et al. (1955) give evidence that radioactive material introduced into the surface layer will reach a 100 m deep thermocline in about 28 hours and Lowman's (1960) work near Eniwetok shows that the distribution is likely not uniform and that after 48 hours much of the radioactivity begins to collect near the thermocline. So the ^{55}Fe introduced as fallout should be available to organisms throughout the mixed layer in a short time.

In the Gulf of Alaska the surface currents move counter-clockwise in the Alaska Gyre, which may act to retain fallout occurring in this area. The center of this gyre, far from river runoff which would dilute the ^{55}Fe with stable iron, may be a site of especially high ^{55}Fe specific activity. Similarly, the Aleutian Islands present a barrier to circulation (Dodimead, Favorite and

Hirano, 1963) which could also maintain high levels of ^{55}Fe in the Bering Sea, since this is a region of high fallout due to proximity to Russian nuclear test sites.

The general picture which emerges, then, is that of ^{55}Fe introduced into the North Pacific Ocean as fallout being rapidly mixed in the surface layer, but with the thermocline forming a barrier to mixing with deeper water. Bathymetry and current systems modify the distribution of ^{55}Fe leading to patchiness and possibly leading to fairly large scale regimes of ^{55}Fe, such as in the Bering Sea and the Alaska Gyre.

In his discussion of salmon migration, Kujala (1966) points out that chinook migrate great distances, generally north of their natal streams while coho migrate shorter distances with a more random pattern. Sockeye, chum and pink salmon have a more northerly range, and their migratory patterns are more east-west than chinook or coho.

While no species of Pacific salmon feeds solely on plankton or solely on fish, the sockeye, pink and chum salmon are primarily plankton feeders while the chinook and coho salmon are more predaceous.

Since the ^{55}Fe data in this study are reported in terms of specific activity, the following list of conditions which will lead to high specific activity or low specific activity will guide the discussion.

In simple terms, the specific activity may be varied only by changing one or both of the two factors of which it is comprised, ^{55}Fe and total iron, but the conditions which occur to vary these two factors are manifested in several ways.

1. High specific activity in sea water results from high ^{55}Fe fallout or from a circulation pattern which tends to limit dispersion of radioactivity. An organism spending much of its time in this high specific activity region will likewise attain high specific activity.
2. Conversely, an organism living primarily in an environment which is high in stable iron from river runoff will develop low specific activity.
3. Foster (1959) has shown that organisms which feed at higher trophic levels have lower specific activity than lower trophic level feeders in the same environment. This occurs if an organism attains its radioactivity primarily through the food chain and is simply a result of physical decay of the radioactive isotope while the stable isotopes remain unchanged.
4. The biological half-life of an organism (the time required to replace one half of the element in the organism) also affects its specific activity but with different results under different conditions. An organism with a short biological half-life will more rapidly approximate the specific activity of its environment than

an organism with a long biological half-life which tends to reflect its previous environment for a longer time. If organisms move from a high specific activity environment to a low specific activity environment, those with short biological half-lives will soon have low specific activity while those with long biological half-lives will retain their high specific activity much longer. When moving from a low to high specific activity environment the opposite case will occur and the organisms with short biological half-lives will be found with higher specific activity. However, an organism with a long biological half-life will always tend towards low specific activity due to decay of ^{55}Fe during its slow exchange of iron with the environment. Certainly, biological half-life is not constant during an organism's lifetime and may be short in the juvenile stages, allowing rapid uptake, but long in adults so that little turnover occurs in the late stages of life.

5. In addition to the above mentioned factors it must be remembered that a migrating organism may sample several environments and thus represent a composite of the specific activities throughout its path.

Salmon

Chinook

The general trend of ^{55}Fe specific activity in chinook salmon, as shown in Table 4, exhibits a decrease from north to south. The highest specific activities of chinook were found in Bristol Bay samples collected north of the Aleutian Islands. The specific activities at this station were an order of magnitude higher than those in southeast Alaska and Canada. Although little information is available on their migration routes, this striking difference is likely due to their migration patterns since Bristol Bay chinook may spend much of their oceanic life in the Bering Sea and North Central Pacific Ocean (Kujala, 1966). This migration pattern would lead to the observed high specific activity in these chinook since the far northern waters are the site of heaviest tropospheric fallout from the Russian tests and this region is not well mixed with waters to the south. Chinook caught farther south spend much of their lifetime moving in a north-south direction along the coast where they are under the influence of high stable iron from the rivers of the west coast of North America and also spend more time away from the region of highest ^{55}Fe input. These factors combine to give the observed decrease in specific activity at the southern stations.

The same pattern of radionuclide distribution observed with

^{55}Fe can be seen from the ^{54}Mn data of Kujala (1966). Manganese-54, like ^{55}Fe, is a fallout product caused by neutron activation in nuclear explosions, and decreases from Bristol Bay south to Astoria, and then increases at Eureka.

The surprising increase of ^{55}Fe specific activity at Eureka, far from the Russian test site, probably arises because of the current systems in this area. Tag returns (Moore, McLeod and Reed, 1960) indicate that Eureka chinook do not undergo the far northern migration of chinook from northern stations so that ^{55}Fe specific activity of chinook caught at Eureka reflects that of water farther south. The current system of the Northeast Pacific (Figure 2) shows a division of the North Pacific Drift off Oregon into two systems, one to the north and the other to the south. This current system operates to maintain a separation between water to the north and water to the south, presenting a barrier to intermixing of these waters. The chinook caught at Eureka spend more time under the influence of the southern water than do those caught farther north. Also, the Davidson Current running north along the coast of California brings water from the south to the region in which these salmon spend much of their lives. It is possible, then, that the increase in ^{55}Fe specific activity at Eureka is due to influx of ^{55}Fe from the south, perhaps from the American tests in the Pacific. This suggestion is supported by the work of Folsom and Young (1965) who find much higher

concentrations of fallout radionuclides west of the California Current than on the east side.

Another possible cause of this increase could be addition of ^{55}Fe from stratospheric fallout which has its maximum at middle latitudes (Mauchline and Templeton, 1964). However, stratospheric fallout of ^{55}Fe is only a small fraction of that existing in the North Pacific Ocean (see location of ^{55}Fe in the ocean, below), so it is doubtful that it has a significant effect on ^{55}Fe specific activity of chinook salmon.

An additional feature seen in chinook salmon is that their ^{55}Fe specific activities are in general lowest of the five species of Pacific salmon. They are the most predaceous and have the longest life span of any salmon and this may account for their low specific activities. As previously noted, organisms feeding at high trophic levels tend toward low specific activities because of lag time allowing for decay. It has also been suggested (Palmer and Beasley, 1967b) that salmon may retain much of their iron pool once they are adults. If this is true, the long life of chinook would also contribute to the low specific activity observed.

Coho

Coho salmon showed the same general pattern of specific activity as chinook, primarily because their migration routes are similar to

those of chinook.

Although chinook were not collected at Depoe Bay, Oregon, coho samples collected there were seen to have specific activities about equal to those at Eureka. These Depoe Bay samples may represent nearly the northern limit of fallout influx from the south since Astoria samples 100 miles to the north displayed lower specific activities.

Possibly owing to the shorter life span of coho, they have ^{55}Fe specific activity levels considerably higher than those found in chinook.

Sockeye

Sockeye salmon contained the highest ^{55}Fe specific activity of any of the salmon samples analyzed, showing similar values at all stations (Table 5). In contrast to the other species of salmon, the ^{55}Fe specific activity of sockeye showed an increasing trend at southern stations, again corresponding to the trend which is evident in ^{54}Mn data (Kujala, 1966).

High values of specific activity in sockeye likely result from their feeding habits and migration routes. They are omnivorous, feeding lowest in the food chain of any of the Pacific salmon. They have 30 to 50 fairly long gill rakers used for filtering compared with 25 to 30 in pinks and 19 to 22 in chinook (Schultz, 1936). This feature

is often used as an indicator of feeding habits in salmon and indicates that sockeye feed low on the food chain. According to the work of Foster (1959) this should lead to higher specific activities in sockeye than in higher trophic level feeders.

If marine organisms gain most of their radioactivity directly from the water as Polykarpov (1966) suggests instead of through the food chain, it is curious that the specific activities of salmon seem tied so closely to feeding habits. Of course, this observation does not rule out the possibility that organisms obtain most of their radioactivity directly from the water since there could be a link between feeding habits and the amount of water filtered.

Another behavior which leads to high specific activity in sockeye salmon is their northerly range and their predominantly east-west migration route. This takes them away from the coastal influence of high river runoff and stable iron dilution and into the region of high fallout. These factors combine to produce high specific activities.

The southerly increase of ^{55}Fe specific activity is difficult to explain. However, it may be that sockeye from southern stations have migrated through water with sufficiently higher specific activity than the water through which the northern sockeye have migrated, accounting for these differences. Hartt et al. (1964) show some of the migrations to the southern stations occurring from farther west and farther from land than those migrating to some northern stations.

These actions could combine to give the slightly higher specific activity at the southern stations.

An anomalous feature of the sockeye is that their specific activity is high despite the fact that they live longer than any other salmon except chinook. The longevity of chinook was cited as a possible contributing factor to their low specific activity. If indeed long life has this effect in chinook but not in sockeye, perhaps this is due to iron having a longer biological half-life in adult chinook than in adult sockeye.

Pink and Chum

Two few pink and chum salmon were analyzed to observe trends relating their ^{55}Fe specific activities with location. The data that are available (Table 7) do not appear far different from sockeye data. The similarity in migration paths of sockeye, pink and chum (Hartt et al., 1964) likely accounts for similarity in their ^{55}Fe specific activities.

Estimate of ^{55}Fe in the North Pacific Ocean

Salmon undergo wide migrations and can thus be assumed to sample many different areas of the ocean. The specific activity of ^{55}Fe in the salmon is a composite of the specific activities of the various waters through which the salmon has migrated. This type of

integration by tuna led Rama, Koide and Goldberg (1961) to estimate the total ^{55}Fe in the ocean using the specific activity of the tuna.

Since a large input of ^{55}Fe has occurred since 1961 a calculation of the total ^{55}Fe in the North Pacific Ocean has been made for each of the years sampled in this study.

The calculation of the total ^{55}Fe in the North Pacific is made by assuming that the ^{55}Fe specific activity of the salmon is equal to the ^{55}Fe specific activity of the ocean water and that an average of the various species at the various stations sampled gives a reasonable average for the North Pacific as a whole. The 1964 ocean average was estimated by giving each species at each station equal weight regardless of the number of individuals involved in the species average. Since not all stations were sampled in subsequent years, the averages for 1965 and 1967 were normalized to the Bristol Bay specific activities which were measured in each year.

Total ^{55}Fe in the North Pacific Ocean is calculated from

$$\text{Total } ^{55}Fe = \bar{G}\, I\, A\, \bar{Z}, \text{ where}$$

$\bar{G}$ = average specific activity of salmon

I = concentration of iron in sea water = 10 ppb (Goldberg, 1963)

A = area of North Pacific Ocean - 86.5×10^6 km^2 (estimated from chart of the North Pacific Ocean)

$\bar{Z}$ = average depth of North Pacific Ocean = 4 km
(Pickard, 1963)

From this equation the total ^{55}Fe content of the North Pacific was calculated for the three years for which data were available and compared in Table 16 with the value found by Rama, Koide and Goldberg (1961) prior to the nuclear tests of 1961-1962.

Table 16. Estimate of the total ^{55}Fe in the North Pacific Ocean.

Average specific activity (nCi/mg Fe)	Total ^{55}Fe (Ci)	^{55}Fe per unit area (nCi/cm^2)	Year	^{55}Fe per unit area corrected to 1962 (nCi/cm^2)
0.037	0.0129×10^{10}	0.149	1961[a]	----
9.13	3.16×10^{10}	36.5	1964	61.0
3.37	1.17×10^{10}	13.5	1965	29.2
0.49	0.170×10^{10}	1.96	1967	7.08

[a]Values in 1961 were calculated from the data of Rama, Koide and Goldberg (1961).

Location of ^{55}Fe in the Ocean

In the introduction to this dissertation a case was developed to support the notion that much of the ^{55}Fe present in the North Pacific Ocean during this study was a result of tropospheric fallout from the Russian tests in 1961 and 1962. The input from this source can be estimated by measuring ^{55}Fe in Alaskan lichen collected on a unit area basis and assuming that the lichen retain all the ^{55}Fe which falls on them. Although this assumption is not entirely true, it gives a good approximation of total ^{55}Fe fallout, since in the region where

these samples were collected, lichen forms a dense ground cover, filtering most of the rain falling in the area. The average ^{55}Fe activity in lichen, corrected for physical decay to 1962, the time of high tropospheric fallout from Russian nuclear tests, gives a total accumulated input of 209 nCi/m^2 on central Alaska. Iron-55 added from stratospheric fallout is an insignificant fraction of this amount. This is known because in 1963 when radioactivity was near its maximum in the atmosphere (Savannah River Laboratory, 1967), the amount of ^{55}Fe in rain was found to be 0.25 nCi/m^2 in New York (Hardy and Rivera, 1966). In subsequent years the contribution of ^{55}Fe was even less. Thus, the yearly contribution of ^{55}Fe from stratospheric fallout is about 10^{-3} of the amount initially introduced through tropospheric fallout.

Organisms ingesting ^{55}Fe in the surface water (euphausids) and those ingesting ^{55}Fe reaching the bottom of the ocean (sea cucumbers) were not collected in the same location as the lichen samples. Likewise, salmon travel widely and will not represent the same region as the other marine organisms analyzed or the lichen. Consequently, comparisons among these data are not strictly valid, but nevertheless may give useful insight into ^{55}Fe location once it is introduced into the ocean.

Euphausids collected in the Columbia River plume (Table 12) can be used to calculate the ^{55}Fe now present in that area using a

method similar to the one used for estimating the total ^{55}Fe in the North Pacific Ocean. Assuming the ^{55}Fe is distributed throughout the surface layer of the ocean to a depth of only 100 m and that the concentration of stable iron is 10 ppb, the ^{55}Fe (corrected to 1962) can be calculated as 0.185 nCi/mg Fe x 10^{-2} mg/l x 10^{5} l/m^2 = 185 nCi/m^2. This number is approximately the same as the input value calculated from lichen. It is surprising that these values are so nearly the same, since the 10 ppb value for iron in the ocean is an average value and Joyner (1964) found values as high as 85 ppb for particulate iron in sea water near the mouth of the Columbia River. Silker (1964) measured iron in Columbia River water filtered by a 0.45 μ filter and found large variations with concentrations up to 100 ppb. Thus, the ^{55}Fe near the mouth of the Columbia River should be diluted by stable iron resulting in a low specific activity in euphausids collected in this area and a correspondingly low estimate of ^{55}Fe in the ocean near the Columbia River. Such a decrease of ^{55}Fe specific activity was observed in salmon collected in the Columbia River.

Since the calculation of input of ^{55}Fe into the ocean and the calculation of the amount present in the surface layer are about equal, it appears that much of the ^{55}Fe is retained in the surface layer and benthic organisms would not be expected to contain a large amount of ^{55}Fe. Sea cucumbers (Table 11) from all depths off the coast of

Oregon appear to support this as do sediment samples collected at the same locations (Table 12), since they were found to have very low ^{55}Fe specific activities.

Vertical transport of fission products in the ocean has been discussed by Osterberg, Carey and Curl (1963) who measured ^{95}Zn - ^{95}Nb in sea cucumbers at a depth of 2800 m and calculated a transport time of seven to twelve days from the surface to 2800 m. They proposed fecal pellets carrying unassimilated radionuclides as the vehicle for rapidly transporting these fission products to the bottom. However, ^{55}Fe does not appear to be rapidly transported to the bottom in this way.

Two possible reasons why this mechanism does not result in high ^{55}Fe specific activities in sea cucumbers and in sediments can be suggested. Firstly, iron, unlike zirconium and niobium, is an important nutrient which is in short supply in the ocean and is much less likely to pass through the gut unassimilated compared to ^{95}Zn - ^{95}Nb. Secondly, stable iron is relatively abundant in sediments so that any ^{55}Fe reaching the bottom will be diluted by stable iron, resulting in a low specific activity.

Any valid description of the location of ^{55}Fe in the ocean must take into account the ^{55}Fe in salmon. In the preceding section, ^{55}Fe in the North Pacific Ocean (decay corrected to 1962) was calculated from salmon data to be 6.1×10^{5} nCi/m^{2} in 1964, decreasing to

0.71×10^5 nCi/m^2 in 1967. These values are 1000 times the values calculated from euphausid and lichen data. One assumption which may lead to error in calculating ^{55}Fe in the ocean from salmon data could be the assumption that salmon obtain their ^{55}Fe from the entire 4000 m water column. If instead, it is assumed that only the top 100 m contain most of the ^{55}Fe, the ^{55}Fe content of the ocean is calculated to be 1.5×10^4 nCi/m^2 in 1964 and 0.18×10^4 nCi/m^2 in 1967. Even these values are at least an order of magnitude higher than the input numbers obtained from lichen analyses. The most straightforward explanation of this difference is simply that more ^{55}Fe was deposited on the North Pacific Ocean than was deposited on land in Alaska. Bowen and Sugihara (1963) have calculated that fallout is greater on the oceans than on an equal area of land at comparable latitudes. They suggest that the efficiency of precipitation over land in scavenging fallout from the atmosphere may be different from that over the sea. Since the Pacific Ocean lies between the Russian test sites and Alaska, it is also probable that more ^{55}Fe was deposited on the Pacific Ocean than on Alaska simply because of proximity to the test sites. Thus, it does not seem unreasonable that at least 10 times as much ^{55}Fe per unit area was deposited on the North Pacific Ocean than on Alaska following the Russian tests, due to a combination of scavenging efficiency by rain and proximity to the test site.

The salmon data further indicate (Table 15) that there has been

a decrease in the amount of ^{55}Fe available to salmon each year since 1964. Decay alone does not account for the decrease in ^{55}Fe available to them, an eightfold decrease occurring between 1964 and 1967 after decay corrections have been applied.

It would be interesting to know how long ^{55}Fe introduced from fallout remains available to biological organisms residing in the surface layer of the ocean. A calculation giving at least an approximation of the length of time ^{55}Fe remains available in the surface layer can be made if two assumptions are made. The first assumption is that most of the ^{55}Fe was introduced into the ocean in 1962 following the Russian nuclear tests. The second assumption is that the removal of ^{55}Fe from the surface layer of the ocean occurs by a first order reaction. Fallout data supports the first assumption, although recent Chinese tests have caused some additional periods of fallout, but not nearly as high as those following the Russian tests (Savannah River Laboratory, 1967). A semilog plot of ^{55}Fe in the ocean vs. year of collection (Figure 7) results in a straight line and appears to support the second assumption.

The equation for this line in Figure 7 is derived from

$$-\frac{dA}{dt} = k_1 A \qquad (1)$$

giving

$$\log A = \frac{-k_1}{2.303} t + \log A_0 \qquad (2)$$

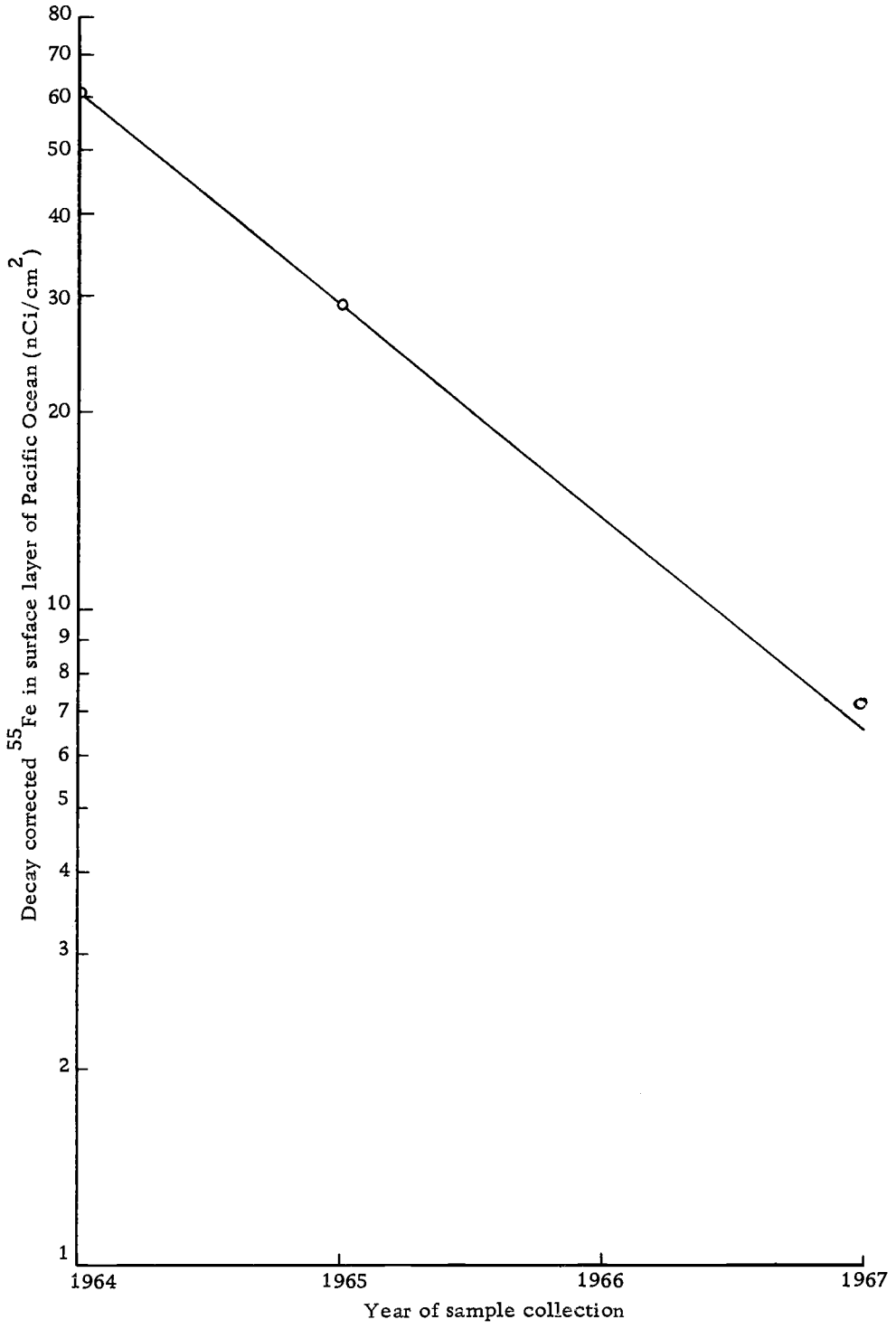


Figure 7. Elimination of ^{55}Fe from the surface layer of the North Pacific Ocean from 1964 to 1967.

where A = activity of ^{55}Fe in the ocean at t

A_0 = activity of ^{55}Fe in the ocean at t = 0

k_1 = rate constant for removal of ^{55}Fe from the ocean.

From the slope of the line described by equation 2 above, k_1 was calculated to be 0.0603 month^{-1} and the half-time for the removal of ^{55}Fe from the surface layer of the ocean to be 11.5 months.

The most likely cause of the decrease of ^{55}Fe in the surface layer of the ocean is a settling out of ^{55}Fe on the bottom. This would seem to conflict with the earlier comparison of ^{55}Fe in euphausids and lichen as well as with the low specific activities measured in sea cucumbers. However, it has since been pointed out that fallout is higher over the ocean than over land at the same latitude so that the close comparison between euphausids collected in 1967 and lichen implies only that the ^{55}Fe in the ocean has now decreased to approximately the same level as was initially deposited on land in Alaska. The location of the ^{55}Fe leaving the surface layer has not been established, but the low specific activity of sea cucumbers and sediments does not conclusively rule out deposition of ^{55}Fe on the bottom, since high stable iron in sediments acts to dilute any ^{55}Fe laid down.

It is also possible that some of the decrease of ^{55}Fe in the surface layer is due to biological retention. Iron retention for long periods of time is known in some organisms and has been suggested

for salmon (Palmer and Beasley, 1967b). However, it is difficult to conceive of a large percent of the ^{55}Fe being bound in this way.

In summary, the contribution from fallout of ^{55}Fe to the North Pacific Ocean appears to be far greater than that falling on land to the east of the Pacific Ocean. About 80 percent of the ^{55}Fe available to salmon in the surface layer was lost between 1964 and 1967. It is proposed that this loss occurs by a combination of biological retention in the surface layer and by transport out of the surface layer. The ^{55}Fe lost from the surface layer does not result in high ^{55}Fe specific activity in bottom samples, possibly because high stable iron in sediments dilutes ^{55}Fe in benthic organisms diminishing the effect of ^{55}Fe reaching the bottom.

Harm to Man

Recommendations of the allowable concentration of artificial radionuclides in sea water have been formulated by a working group of the National Academy of Sciences Committee on Oceanography (NAS-NRC, 1962). The specific activity approach discussed earlier in this paper was the guideline used for these recommendations. In order to make sure man is adequately protected, ^{55}Fe has a safety factor of 100 included in the calculation of its maximum permissible specific activity in sea water.

Since the spleen is the critical organ for ^{55}Fe concentration in

man, the maximum permissible specific activity of ^{55}Fe in the spleen was calculated to be 250 μCi/g. When the safety factor, and a decay factor taking into account both biological half-life and physical half-life were included, the maximum specific activity of sea water was calculated to be 250 μCi/mg x 1.5 x 10^{-2} = 3.8 μCi/g, where 1.5 is the decay factor and 10^{-2} is the safety factor.

If the ^{55}Fe specific activity of sea water is not allowed to exceed this specific activity there is no way for organisms living in sea water to attain an ^{55}Fe specific activity greater than the recommended level, assuming ^{55}Fe and stable iron exist in the same form. In turn, man is assured of not receiving more than an allowable radiation dose by eating seafood from this environment.

Sockeye salmon (Table 5), which represent the highest values of ^{55}Fe specific activity found in this study, are seen to exceed the maximum permissible ^{55}Fe specific activity of sea water both in 1964 and 1965, but not in 1967. The highest value, at the Strait of Juan de Fuca in 1964, was 7.5 times the maximum permissible ^{55}Fe specific activity of sea water. Since organisms living in sea water exceeded the recommended ^{55}Fe specific activity, it follows that the sea water from which they derived their ^{55}Fe specific activity must have also exceeded the recommended value.

This does not mean, however, that this situation resulted in harm to man. For any deleterious effect to result from high specific

activity, the ^{55}Fe must be maintained at a high level specific activity for a time sufficient to allow man to come to exceed his maximum allowable ^{55}Fe specific activity through consumption of seafood. A high specific activity for a short time followed by a decrease to below recommended levels gives no cause for alarm since the recommendations were made on the basis of an equilibrium value persisting over a lifetime. It should also be pointed out that for man to receive a harmful level of ^{55}Fe from this source, his entire iron supply would have to come from sockeye salmon, an unlikely situation indeed. However, this does account for high body burdens of ^{55}Fe in people whose diet consists largely of ocean fish (Palmer and Beasley, 1967b). In all cases, the 10^{-2} safety factor more than covers any high specific activity measured in this study.

CONCLUSIONS

In the introduction to this dissertation specific purposes related to increasing the knowledge of ^{55}Fe in the marine environment were suggested. Since little previous work had been done in this area, much of this study was exploratory in nature and only the most general conclusions can be inferred. However, some inroads into the behavior of ^{55}Fe have been made which serve to emphasize the radioecological importance of this radionuclide.

The problem of where ^{55}Fe goes once introduced into the marine environment still remains somewhat of an enigma. The ^{55}Fe specific activities of salmon suggested that ^{55}Fe is lost from the surface layer, but sea cucumbers residing on the ocean floor did not exhibit a corresponding increase from 1964 to 1967. Sediments, like sea cucumbers, were also found to have only low ^{55}Fe specific activities. This much can be said, ^{55}Fe does not remain available to organisms in the surface layer but is lost with a half-time of about one year. Where this ^{55}Fe from the surface goes, cannot be stated surely, but it seems probable that it reaches the ocean bottom where it is diluted by large amounts of stable iron in the sediments.

Higher concentrations of stable iron in Columbia River water compared to adjacent sea water present the possibility of two effects on ^{55}Fe in the ocean off the Columbia River mouth: (1) decreased

^{55}Fe specific activity due to dilution of fallout ^{55}Fe by stable iron and (2) increased ^{55}Fe specific activity due to production of ^{55}Fe by neutron activation of stable iron as a portion of the river is diverted through the reactors at Hanford for cooling. The decrease in ^{55}Fe specific activity in salmon near the mouth of the Columbia River suggested that the dominant effect of the Columbia River on ^{55}Fe in the ocean is that of dilution by stable iron input, resulting in a lowering of specific activity in this region. Specific activities of euphausids were also low in the Columbia River plume, but an endeavor to obtain comparable samples from Alaska for comparison was not successful.

The total input on the North Pacific Ocean of ^{55}Fe from fallout is considerably higher than on land at the same latitudes. The input on the land in Alaska, decay corrected to 1962, the time of high tropospheric fallout from the Russian nuclear tests, was calculated to be 209 nCi/m^2. From the ^{55}Fe specific activities of salmon, the ^{55}Fe in the top 100 m of the North Pacific Ocean was calculated to be 1520 nCi/m^2 in 1964. In subsequent years the ^{55}Fe in the top 100 m was seen to decrease with a rate constant for the removal of ^{55}Fe from the surface layer of 0.0603 $month^{-1}$, assuming a first order reaction governs the removal of ^{55}Fe. However, uptake by biological organisms indicated that the site of highest ^{55}Fe specific activity throughout the period of this study was the surface layer of the ocean.

Need for further study of ^{55}Fe in the ocean is evident from this

study. While the specific activities found were not high enough to cause concern, they were high enough to indicate that in the case of future nuclear testing, ^{55}Fe may be one of the first radionuclides to become a health hazard. Consequently, it is due much more attention than it has received in the past. Its vertical distribution in the ocean needs to be elucidated both in marine organisms and, if possible, in sea water. Study of ^{55}Fe in plankton over several years in several oceans would further illuminate the residence time of ^{55}Fe in the surface layer and would point up regions of the ocean containing high ^{55}Fe. This information may prove valuable in prohibiting harm to man if a large input of ^{55}Fe into the ocean should occur.

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APPENDIX

APPENDIX

Iron-55 and Total Iron in Marine Samples

The average ^{55}Fe per sample, the average total iron per sample and the average sample dry weights are presented in this appendix. Persons wishing to use these data should be cautioned that in some cases the total sample listed in the dry weight column may not have been completely dissolved or may have experienced some loss in ashing. These errors do not affect specific activity but may bias estimates of ^{55}Fe or total iron per unit weight.

The data are in the order in which they appear in the tables in the text of this dissertation.

Location	Sample type	No. individuals	Year of collection	dry wt. (g)	^{55}Fe per sample (nCi)	Total Fe per sample (mg)
Bristol Bay	chinook	7	1964	27.03	33.68	2.78
Bristol Bay	chinook	3	1965	38.69	19.86	5.71
Bristol Bay	chinook	7	1967	31.16	3.72	5.10
Petersburg	chinook	4	1964	101.2	15.79	12.7
Skeena River	chinook	4	1964	37.30	6.25	8.05
St. Juan de Fuca	chinook	2	1964	32.20	5.34	7.20
Astoria	chinook	4	1964	43.80	5.72	9.45
Astoria	chinook	4	1967	unknown	4.22	23.8
Eureka	chinook	2	1964	42.94	26.53	3.98
Bristol Bay	sockeye	19	1964	13.66	20.21	2.07
Bristol Bay	sockeye	3	1965	65.15	34.13	7.32

Location	Sample type	No. individuals	Year of collection	dry wt. (g)	^{55}Fe per sample (nCi)	Total Fe per sample (mg)
Bristol Bay	sockeye	8	1967	28.11	5.02	11.5
Cook Inlet	sockeye	2	1965	55.66	53.82	5.90
Petersburg	sockeye	2	1964	82.89	85.00	5.41
Petersburg	sockeye	3	1965	50.04	61.84	8.05
Skeena River	sockeye	5	1964	51.72	109.7	8.08
Barkley Sound	sockeye	1	1964	29.40	117.7	6.08
St. Juan de Fuca	sockeye	2	1964	60.76	93.32	3.28
Astoria	sockeye	5	1967	32.53	7.02	5.61
Japan High Seas	sockeye	3	1965	32.68	36.19	7.02
Cook Inlet	coho	2	1964	19.92	33.78	2.10
Cook Inlet	coho	2	1965	60.44	22.59	3.22
Petersburg	coho	6	1964	30.44	60.80	4.68
Skeena River	coho	1	1964	28.75	51.13	6.28
Str. Juan de Fuca	coho	2	1964	65.47	13.04	8.32
Astoria	coho	5	1967	unknown	1.51	6.36
Depoe Bay	coho	6	1964	25.88	18.06	3.69
Eureka	coho	1	1964	92.52	128.92	22.1
Eureka	coho	11	1965	unknown	23.58	14.8
Japan High Seas	coho	1	1965	29.80	15.81	3.24
Cook Inlet	pink	1	1964	14.39	9.48	1.20
Cook Inlet	pink	2	1965	20.42	9.38	1.66
Petersburg	pink	2	1964	64.66	87.78	13.0
Petersburg	pink	2	1965	60.04	32.50	9.16
Petersburg	chum	1	1964	22.90	70.08	4.64
Japan High Seas	chum	8	1965	37.37	57.84	8.66
Oregon (50m)	chum	6	1964	9.99	0.104	38.3
Oregon (150m)	Stichopus californicus	3	1964	11.63	0.030	8.76

Location	Sample type	No. individuals	Year of collection	dry wt. (g)	^{55}Fe per sample (nCi)	Total Fe per sample (mg)
Oregon (200m)	Stichopus californicus	3	1964	15.10	0.127	4.48
Oregon (200m)	Stichopus californicus	?	1965	3.52	0.015	3.68
Oregon (200m)	Stichopus californicus	1	1966	5.57	0.003	3.08
Oregon (200m)	Stichopus californicus	2	1967	9.62	0.008	3.32
Oregon (600m)	Laetmophasma fecundum	8	1964	4.37	0.344	36.6
Oregon (820m)	Laetmophasma fecundum	12	1964	2.54	0.115	36.6
Oregon (1000m)	Laetmophasma fecundum	7	1964	3.22	0.268	34.1
Oregon (800m)	Laetmophasma fecundum	11	1965	1.87	0.097	30.0
Oregon (800m)	Laetmophasma fecundum	8	1965	2.10	0.099	34.5
Oregon (800m)	Laetmophasma fecundum	8	1965	1.36	0.017	15.2
Oregon (2800m)	Paelopatides sp.	2	1964	0.42	0.041	0.92
Oregon (3000m)	Paelopatides sp.	6	1965	2.83	0.031	8.00
Oregon (2800m)	Paelopatides sp.	?	1965	4.85	0.049	15.6
Oregon (2800m)	Paelopatides sp.	?	1965	0.41	0.027	4.16
Oregon (2853m)	Paelopatides sp.	6	1966	7.35	0.018	4.40
Oregon (2810m)	Paelopatides sp.	6	1967	3.47	0.023	4.40
Oregon (2810m)	Paelopatides sp.	6	1967	0.40	0.003	3.40
Oregon (200m)	sediment	-	1964	unknown	none visible	26.65
Oregon (200m)	sediment	-	1965	unknown	none visible	64.15

Location	Sample type	No. individuals	Year of collection	dry wt. (g)	^{55}Fe per sample (nCi)	Total Fe per sample (mg)
Oregon (800m)	sediment	-	1964	unknown	0.0035	8.99
Oregon (600m)	sediment	-	1965	unknown	0.0084	17.97
Oregon (2800m)	sediment	-	1964	unknown	0.0079	23.61
Oregon (2800m)	sediment	-	1966	unknown	0.0044	47.87
Oregon (Sta 1)	Euphausia pacifica	many	1967	10.27	0.0106	1.48
Oregon (Sta 2)	Euphausia pacifica	many	1967	17.44	0.0189	0.530
Oregon (Sta 3)	Euphausia pacifica	many	1967	12.61	0.0302	0.300
Oregon (Sta 4)	Euphausia pacifica	many	1967	11.70	0.0371	1.24
Oregon (Sta 5)	Euphausia pacifica	many	1967	30.51	0.0317	2.16
Oregon (Sta 6)	Euphausia pacifica	many	1967	27.29	0.0329	0.540
Oregon (Sta 7)	Euphausia pacifica	many	1967	unknown	0.0385	0.480
Oregon (Sta 8)	Euphausia pacifica	many	1967	25.98	0.0553	1.12
Oregon (Sta 9)	Euphausia pacifica	many	1967	6.55	0.0143	0.200
Anaktuvuk Pass	Cladonia rangiferina	-	1966	14.54	0.440	4.08
Anaktuvuk Pass	Cladonia rangiferina	-	1966	17.60	0.400	5.88
Selawik River	Cladonia alpestris	-	1966	16.64	0.416	3.80
Selawik River	Cladonia alpestris	-	1966	18.28	0.497	4.32
Anaktuvuk Pass	Cladonia alpestris	-	1967	8.85	0.257	4.44
Anaktuvuk Pass	Cladonia alpestris	-	1967	14.14	0.376	6.36

Location	Sample type	No. individuals	Year of collection	dry wt. (g)	^{55}Fe per sample (nCi)	Total Fe per sample (mg)
Selawik River	Cladonia alpestris	-	1967	21.44	0.484	3.84
Selawik River	Cladonia alpestris	-	1967	25.15	0.509	3.88
Bristol Bay	Mytilus sp.	6	1964	2.75	0.017	3.92
Petersburg	Mytilus sp.	21	1965	22.85	0.144	8.32
Astoria	Pacific Mackeral	1	1964	34.20	5.69	6.20
Alsea	Steelhead trout	2	1964	34.30	75.50	14.56
Dexter Dam	chinook salmon	1	1966	33.70	2.23	11.56