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RETENTION OF FALL-OUT CONSTITUENTS IN UPPER LAYERS
OF THE PACIFIC OCEAN AS ESTIMATED FROM STUDIES
OF A TUNA POPULATION

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ABSTRACT

Repeated measurements of cobalt-60, zinc-65, manganese-54, caesium-137, silver-110m, silver-108m, and plutonium-239 in several organs of albacore tuna suggest that the upper layers of the North Pacific Ocean can retain large fractions of several species of trace elements for periods of a decade or more. For example, caesium-137 concentrations in the livers and muscle tissues of North Pacific albacore caught from 1965-1971 decreased to half in about 10 years. In comparison, the reported rate of input from fall-out during this period decreased more rapidly, closer to half in one year. This suggests a strong retention of caesium-137 in the upper water masses which are accessible to the fish.

It is of interest to note that long environmental persistences in the upper oceanic layers are also indicated for some other nuclides that are much more highly accumulated by organisms than is caesium. For example, cobalt-60 and silver-108m concentrations in albacore liver tissues fell to half during this period in 2.6 and 7.1 years respectively. Plutonium-239 concentrations in the livers decreased to half in about 3.5 years.

The attenuation rate of zinc-65 was discontinuous between 1965 and 1968. This fact, along with observation of comparatively high ratios of zinc-65 to cobalt-60 in tunas of the southern hemisphere following 1968, suggests that new large weapons were tested that gave off relatively large amounts of zinc-65.

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INTRODUCTION

The great nuclear tests of 1961-2, and earlier, ultimately brought to the world oceans many radioactive metals. Numerous and repeated measurements at Scripps of manganese-54, cobalt-60, zinc-65, silver-108m, silver-110m, caesium-137, and plutonium-239 concentrations in albacore (Thunnus alalunga) livers have been made in 1964, 1965, 1968, 1970, and 1971 to ascertain how long these metals remain available to inhabitants of the upper waters of the N. Pacific.

Livers were chosen because they concentrate many of these metals to extremely high levels relative to the concentrations suspected in the seawater in which they live. Albacore were chosen for sampling because samples from the age groups can be collected at the same time and place each year. Intensive tagging studies, by the California and U.S. fisheries services, have demonstrated that these pelagic fish perform spectacular seasonal migrations [1,2]. They arrive near southern California every July from the central Pacific, then they apparently travel north along the U.S. coast as the summer progresses. Later they migrate back to the central Pacific and subsequently into waters off the coast of Japan. Some individuals apparently return to the eastern Pacific during the following seasons. Most tuna authorities believe that the albacore of the North Pacific belong to a single population [3].

Albacore feed on a wide variety of organisms of suitable size, apparently on whatever is available. Organisms eaten include anchovies, squid, sauries, jack mackerel, juvenile rock fish, lanternfish and euphausiids. Thus they are high on the marine food chain [2]. They have been observed to depths of 300 meters [3].

The California fishery depends usually on a single age group; generally the most important size is the two-year-old fish averaging 13 pounds [2]. It is from these fish, caught in the summer months off San Diego, that the majority of the livers were collected.

ANALYTICAL METHODS

Freshly removed livers were weighed and generally dried in plastic containers (3/4 in. high and 4.5 in. in diameter) at 70°C. All samples are currently analysed in a gamma detector between two vertically opposed 6 in. x 4 in. NaI(Tl) crystals. Background reduction is accomplished by use of pure NaI light pipes and a 12 in. O.D. NaI(Tl) anticoincidence guard ring. The detector signals are fed to a 200-channel pulse-height analyser, counting in the anticoincidence mode, as well as to a two-dimensional analyser. However, some of the early zinc-65 and manganese-54 measurements were made with a single 5 in. x 4 in. NaI(Tl) crystal and the earlier cobalt-60 and silver-110m concentrations were resolved with a so-called "4 π summing" spectrometer [4].

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Radioactive silver nuclides are now routinely extracted from the wet tissue using an electroplating process employing a silver carrier [5]. Plutonium-239 has been extracted from ashed samples and determined by alpha counting [6].

PRESENTATION OF DATA

Mean trends for the six radiometals in albacore livers from 1964-1971 are tabulated in Table I. Yearly values of caesium-137 in the tail muscle are presented in Table II. Visual presentations of the data, by nuclide, are to be found in Figures 1 to 7.

FALL-OUT INPUT TRENDS FROM 1961-1971

Figure 8, which is a linear replotting of fall-out strontium-90 data of Volchok and Kleinman [7], is presented to give the reader some feel of the reported inputs of strontium-90 (as well as certain other nuclear wastes) arriving at the surface of the northern hemisphere between 1963 and 1964. Most of the fall-out occurred prior to 1965; relatively low inputs occurred thereafter. The probable behaviour of this large amount of fall-out that entered the oceans at about 1963-1964 will be the main subject of this discussion as well as can be deduced from the behaviour of tunas.

CAESIUM-137 TRENDS

Vertical and horizontal movements of the radioisotope caesium-137 in the North Pacific have been and are currently being studied in detail by use of hundreds of seawater samples [8]. However, resolving, from trends of water sample measurements, a simple summary of the movements of fall-out in surface waters and deeper waters of the North Pacific is a horrendous task. One is tempted to employ averaging processes that are available in the biosphere.

Both radioactive and stable caesium have been shown to accumulate only moderately in albacore muscle (about 100-fold over seawater) [9]. It is evident from measurements presented in Figure 1 that caesium concentrates to a somewhat lesser degree in the liver. However, it should be noted that there is a parallel rate of decrease in caesium-137 concentrations between 1965 and 1971 in both the flesh and liver to half value in about 10 years.

A reasonable interpretation of our data is that the albacore, who swims enroute to the California fishery approximately 17 nautical miles a day (500 miles a month) accumulates traces of fall-out in some proportion to average concentrations in his environment. This average includes subsurface layers also since these far ranging fish may seek out food in water anywhere between about 160 and 320 feet deep [3] and this food may in turn depend on conditions at still greater depths.

Sharp decreases in the case of caesium-137 concentrations are noted from 1964 to 1965. On the other hand, upward changes are noted for all of the other metals suggesting that caesium is somehow unique. This may be a consequence of its ionic nature and the low build-up factor over water that characterizes caesium. It is possible, however, to speculate several other reasons for this special behavior of caesium. For example, it may be caused by persistence of caesium-137 from much earlier (premoratorium) weapons tests.

SILVER-108m AND SILVER-110m TRENDS

Usefulness of silver-108m ($t_{1/2} = 127$ yr) as a tag for ocean study has been stressed elsewhere [10,11]. Figure 2 shows trends that suggest that

silver may either be supported by the biosphere or exist as an otherwise suspended species in the ocean for many years. The apparent half-time of residence ($A_{1/2}$) is about 7 years.

After 1965 there is a decrease in silver-110m approximating its physical decay rate. If there were no new inputs this is what one would expect knowing that the $A_{1/2}$ for silver-108m is about 7 years. Thus the significant upward departure noted in 1970 and 1971 strongly suggests new inputs of silver-110m, for example, by one or more of the nuclear tests at Lop Nor during 1967-1969. New inputs can also be inferred by extrapolating the silver-110m/silver-108m ratio back in time. This ratio for the 1965 livers extrapolates to 199 on August 1962, a value consistent with the activation of silver on that date [12]. However, the 1970 ratio extrapolated to the same date is about 4300, much too large.

The 1965 peak for silver-108m and silver-110m implies a delayed response of 1 or 2 years in the biosphere to new inputs of silver. This may stem from exceedingly high concentration factors, approaching 500,000, for silver in the albacore livers [12,13].

COBALT-60 TRENDS

Cobalt concentration trends from 1964 to 1965, like silver, suggest a 1-2 year delayed response in the biosphere to new inputs (see Figure 4). After the 1965 maximum, cobalt-60 concentrations fall off at a rate giving an apparent half-time of residence of about 2.5 years. Again the 1970 fish reflect the possibility of influence from the Lop Nor testing.

Of the radioisotopes having long decay half-lives, this biologically important element has the shortest apparent half-time of residence in the upper layers of the ocean.

MANGANESE-54 TRENDS

Another biologically important element, manganese-54, has a radioactive decay rate somewhat like that of silver-110m. Thus it also would be expected to be sensitive to new fall-out. The 1970 fish concentrations show evidence for new inputs of manganese-54. The apparent half-time of residence in the sea of 320 days is essentially equal to the radioactive decay half-life of this nuclide.

An extensive study of albacore livers has been reported by Percy and Osterberg [14] from 1962-1966 relating to zinc-65 and manganese-54. Their fish were caught off three general Pacific coastal positions including Baja California. They have suggested that their manganese-54 concentrations were of fall-out origin, and therefore we have included some of their values in Figure 5 for comparison. Assuming a 1.0% ash to wet weight ratio such as we have commonly found for liver at Scripps, their absolute manganese-54 activities agree remarkably well with ours and contribute data for manganese-54 one year earlier than ours.

ZINC-65 TRENDS

The trends of zinc-65 provide strong evidence that the Lop Nor tests contributed substantial amounts of zinc-65 to world-wide fallout as has been suggested by Kolb [15]. By extrapolating the zinc-65 concentration from 1964-5 forward along its physical decay (as shown by the dashed lines in Figure 6) the suggestion is supported that large amounts of zinc-65 were put into the ocean after 1967. Subsequent smaller rises also are suggested in the 1970 and 1971 livers.

Pearcy and Osterberg's mean zinc-65 activity in Baja California albacore livers is included in Figure 6. They had concluded that these fish samples in contrast to their albacore samples off Oregon and southern Washington, were not influenced by zinc-65 outputs of the Columbia River even in these early years when zinc-65 concentrations in the Columbia River were relatively high.

It is not impossible to conceive that some radionuclides from the Columbia might have been transferred by the California Current to southern California prior to 1964. Yet our recent analyses of livers of albacore and other large fish collected from distant areas of the North and South Pacific Oceans and from the Atlantic almost all show increased concentrations of zinc-65, substantiating new origins of world-wide fall-out (Table III). Moreover, Kolb [15,16], has narrowed the zinc-65 main new source to a probable location at the 6th Chinese test in June 1967.

It should also be pointed out that air filters collected at Pt. Barrow by Battelle N.W. Laboratory record increased zinc-65 activity following 1967 [17]. Zinc-65 in the livers of southern Pacific specimens may well have come from recent French tests [18] as well as the Chinese tests.

PLUTONIUM-239 TRENDS

Very little data has been available concerning plutonium-239 concentrations in oceanic fish [19,20]. Plutonium has a build-up factor in albacore livers somewhat like caesium. Unfortunately, low activities of about 0.1 pCi/wet kilogram necessitate very large samples. However, the long decay half-life (24,360 years) makes plutonium an intriguing ocean tag. It is of some importance to establish contemporary levels of plutonium for reference with the much larger amounts that will be anticipated with the increasing use of this nuclear fuel.

A few determinations of plutonium-239 in the San Diego albacore livers are found in Figure 7. The tentative apparent half-time of residence is three and a half years. Studies of distributions of this nuclide are continuing at Scripps.

RELATIVE HALF-TIMES OF RESIDENCE OF THE NUCLIDES

Table IV illustrates the range of half-times of residence attributable not to their radioactive decay rate but to some property of the upper layers of the ocean itself--including its organisms.

CONCLUSIONS

It is difficult to avoid oversimplification of data such as ours. Nevertheless, the overriding suggestion is an oceanic model in which trace elements of several types, both biologically active ones like cobalt and silver, and apparently unneeded elements like caesium, may be retained for a matter of decades in the upper water masses in which most marine organisms live.

Secondly, there is a body of oceanic evidence that global fallout was renewed after 1967 including large amounts of zinc-65.

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Table I. Activities of ^{54}Mn , ^{60}Co , ^{65}Zn , $^{108\text{m}}\text{Ag}$, $^{110\text{m}}\text{Ag}$, ^{137}Cs , and ^{239}Pu in livers of San Diego albacore (Thunnus alalunga) collected in July and August.

Nuclide	Year	No. of fish	pCi/wet Kg		
			Mean	S.D. of mean	Range
Mn-54	1964	8	1045	36	950-1140
	1965	10	587	83	360-1060
	1968	44	41	1	30-60
	1970	43	15	1	10-20
	1971	29	6	0.1	5.6-6.4
Co-60	1964	20	145	13	102-250
	1965	28	174	9	80-280
	1968	44	63	3	40-110
	1970	43	45	1	40-51
	1971	39	33	1	26-48
Zn-65	1964	8	1255	25	1190-1320
	1965	10	384	49	200-610
	1968	44	665	25	400-1000
	1970	43	223	4	190-270
	1971	39	122	4	73-155
Ag-108m	1964	26	4.6	0.3	2.8-7.9
	1965	26	7.5	0.4	2.7-11.0
	1968	39	5.9	0.1	5.4-8.2
	1970	59	4.6	0.05	3.8-4.8
	1971	76	4.2	0.06	3.6-6.5
Ag-110m	1964	8	81	0.4	80-82
	1965	26	100 ^a	-	-
	1968	21	8.7 ^b	-	-
	1970	59	5.8	0.08	4.4-5.8
	1971	76	4.9	0.09	3.5-7.6
Cs-137	1964	16	83	3	67-106
	1965	19	40	0.2	38-41
	1968	18	35.8	0.2	35-37
	1970	27	30.9	0.3	29-32
	1971	29	28.5	0.5	25-33
Pu-239	1964	7	0.062 ^c	-	-
	1965	7	0.29 ^c	-	-
	1968	7	0.223 ^c	-	-
	1970	7	0.098 ^c	-	-
	1971	19	0.091 ^d	-	-

a: determined from the average Ag-108m value of 7.5 pCi/wet Kg; b: one cyanide extraction of 21 livers; c: one determination; d: average of two independent determinations.

Table II. Activities of ^{137}Cs in muscle tissue
of San Diego albacore (Thunnus alalunga).^a

<u>Year</u>	<u>No. of Fish</u>	<u>pCi/wet Kg</u>		
		<u>Mean</u>	<u>S.D. of mean</u>	<u>Range</u>
1964	1	160	-	-
1965	18	90	4	61-135
1966	6	88	4	74-99
1967	6	81	5	67-95
1968	6	58	3	45-70
1969	6	78	4	68-94
1970	6	64	2	57-70
1971	6	58	2	49-65

a: collected in July and August, round weights of approximately 20 lb (3 yr old).

Table III. Comparisons of activities in livers of several species of tuna from widely separated ocean regions.

Species	Collection Date	Position	Fish Wt. (lb)	Sample Wet wt. (g)	pCi/wet Kg $\pm$ one counting standard deviation					$\frac{\text{Zn-65}}{\text{Co-60}}$
					Zn-65	Co-60	Ag-110m	Ag-108m	K-40	
<u>Hawaiian Islands</u>										
Big Eye	12/ 9/70a	17°50'N 156°30'W	76	246	193 $\pm$ 9	51.3 $\pm$ 2.1	0.45 $\pm$ 2.27	0.86 $\pm$ 0.57	1844 $\pm$ 74	3.8
Big Eye	12/ 9/70b	"	86	224	139 $\pm$ 9	35.3 $\pm$ 1.4	0.51 $\pm$ 2.21	0.99 $\pm$ 0.61	2126 $\pm$ 67	3.9
Yellowfin	12/ 9/70a	"	210	201	742 $\pm$ 22	67.7 $\pm$ 2.4	178 $\pm$ 5	175 $\pm$ 6	2256 $\pm$ 89	11
Yellowfin	12/ 9/70b	"	186	216	639 $\pm$ 18	41.2 $\pm$ 1.6	67.1 $\pm$ 2.9	49.5 $\pm$ 1.9	2042 $\pm$ 73	16
Yellowfin	4/15/71a	450 mi S. of Oahu	82	184	145 $\pm$ 8	24.8 $\pm$ 1.2	0.33 $\pm$ 2.20	0.9 $\pm$ 1.2	1976 $\pm$ 63	5.9
<u>Marquesas Islands</u>										
Albacore	1/ 5/71a	14°S 130°W	74	133	264 $\pm$ 18	6.2 $\pm$ 1.4	0 $\pm$ 3.0	1.1 $\pm$ 1.4	2350 $\pm$ 120	43
Albacore	1/ 5/71b	"	79	127	858 $\pm$ 17	15.3 $\pm$ 1.5	0.7 $\pm$ 3.4	1.6 $\pm$ 1.4	2597 $\pm$ 96	56
<u>N.W. Africa</u>										
Yellowfin	11/15/70a	off coast of Sierra Leone	80	205	90 $\pm$ 8	1.22 $\pm$ 0.55	4.6 $\pm$ 2.2	0.88 $\pm$ 1.07	2133 $\pm$ 78	74
Yellowfin	11/15/70b	"	80	183	98 $\pm$ 8	3.1 $\pm$ 0.6	4.9 $\pm$ 2.4	1.4 $\pm$ 0.9	2178 $\pm$ 76	32
<u>N.W.-S. America</u>										
Yellowfin	2/ 7/71a	200 mi off Ecuador	80	170	60 $\pm$ 9	12.3 $\pm$ 1.0	0 $\pm$ 2.1	0 $\pm$ 1.1	1900 $\pm$ 77	4.9
Yellowfin	2/ 7/71b	"	80	183	68 $\pm$ 9	12.0 $\pm$ 0.9	0.66 $\pm$ 2.30	0.93 $\pm$ 0.78	2013 $\pm$ 83	5.7

Table IV. Half-times of residence (corrected for radioactive decay) for ^{60}Co , ^{108}Ag , ^{137}Cs , and ^{239}Pu in upper layers of the North Pacific.

<u>Nuclide</u>	<u>Corrected half-time (years)</u>
Co-60	5.0
Ag-108m	7.4
Cs-137	17
Pu-239	3.5

Figure 1. Concentrations of ^{137}Cs in muscle and liver tissues of albacore tuna caught off San Diego from 1964-1971.

Figure 2. Concentrations of ^{108m}Ag in liver tissues of albacore caught off San Diego from 1964-1971.

Figure 3. Concentrations of ^{110m}Ag in liver tissues of albacore caught off San Diego from 1964 - 1971.

Figure 4. Concentrations of ^{60}Co in liver tissues of albacore caught off San Diego from 1964-1971.

Figure 5. Concentrations of ^{54}Mn in liver tissues of albacore caught off San Diego from 1964-1971.

Figure 6. Concentrations of ^{65}Zn in liver tissues of albacore caught off San Diego from 1964 - 1971.

Figure 7. Concentrations of ^{239}Pu in liver tissues of albacore caught off San Diego from 1964-1971.

Figure 8. Fall-out input trends of ^{90}Sr deposition in the northern hemisphere from 1958-1971.