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INTERACTIONS OF RADIONUCLIDES IN WATER, PARTICULATES, AND OYSTERS IN THE DISCHARGE CANAL OF A NUCLEAR POWER PLANT

Florence L. Harrison
Kai M. Wong
Robert E. Heft

August 19, 1976

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Contents

Abstract	1
Introduction	2
Site Description	4
Materials and Methods	6
Experimental Station	6
Sampling Procedures	6
Seawater	6
Particulates	6
Oysters	6
Analytical Procedures	7
Results	9
Characterization of Radionuclide Source	9
Discharge Canal Water	9
Radionuclide Concentrations during Single Releases	10
Radionuclide Concentrations Averaged over Periods of Several Releases	12
Discharge Canal Particulates	12
Suspended Particulates	14
Settled Particulates	16
Dynamics of Radionuclide Transfer Between Seawater and Particulates	21
Oysters	21
Short-Term Accumulation	21
Long-Term Accumulation	28
Concentration Factors	29
Discussion and Conclusions	29
Seawater	29
Particulates	32
Suspended Particulates	32
Settled Particulates	33
Oysters	35
Acknowledgments	39
References	40

INTERACTIONS OF RADIONUCLIDES IN WATER, PARTICULATES, AND OYSTERS IN THE DISCHARGE CANAL OF A NUCLEAR POWER PLANT

Abstract

This study was designed to provide data for dynamic modeling of radioactive pollutants in marine ecosystems adjacent to nuclear power plants.

The data are relevant to the dynamics of radionuclide transfer among seawater, suspended particulates, sediments, and biota. Gamma-emitting radionuclides (^{54}Mn , ^{60}Co , ^{65}Zn , and ^{137}Cs) were followed in the water and particulates as well as in oysters introduced into the discharge canal of the boiling water reactor of the Humboldt Bay Power Plant near Eureka, California. The liquid waste was introduced into the canal at irregular intervals and contained radionuclides at extremely low but variable concentrations. Radionuclides were determined in the oysters, water, and particulates after single releases (over about 6 hr) and over a long series of releases (18 months).

Radionuclide concentrations in the discharge canal water varied widely with time due to irregularity of release, differences in quantities of radionuclides released, and variations in hydrological conditions. Concentrations in the seawater changed

rapidly during a release, reaching peak values within 30 minutes. The released radionuclides were partitioned between the soluble and particulate phases of the water and were accumulated in the bottom sediments. The partitioning and the amount accumulated were radionuclide-dependent and varied with the biological and hydrological conditions; consistent patterns of interchange among water, suspended particulates, and bottom sediments could be discerned.

Except for radiocesium, the concentrations in the water differed from those expected from the known dilution of the source material. In general, for those radionuclides with a high affinity for particulate material, concentrations were more variable and, on the average, lower than expected during releases and higher than expected between releases. The bottom sediments in the discharge canal represent a pool of radionuclides bound to particulates. Re-suspension of these particulates due to changing hydrological conditions appears to affect the concentration

of radionuclides in the water between releases.

Oysters introduced into the discharge canal rapidly accumulated the released radionuclides. Our results indicate that suspended particulates play an important role in the accumulation of some radionuclides by oysters. Concentration of radionuclides detected in the oysters generally reflected the levels measured in the water and particulates. During a period of high biological productivity radionuclide accumulation rates were decreased considerably, particularly for ^{60}Co , by filtration of the water in which the oysters were maintained. The decrease in rate appears to be related to the quantities and compositions of the particles in the water.

For ^{54}Mn , ^{60}Co , and ^{65}Zn the concentration factors calculated for the oysters from the expected water

concentrations were higher than those predicted from the concentration factors for the corresponding stable elements. This apparent discrepancy occurs because the calculated "expected concentrations" of radionuclides in the water do not accurately represent the concentrations available to the oysters. The calculations do not take into account the binding of the radionuclides to particulate matter, which increases their residence time in the discharge canal relative to the residence time of the water. Most models of the effects of pollutants released into ecosystems have considered the transport and movement of materials in the water mass only. It is apparent from this study that for radioactive or trace-element pollutants with a high affinity for particulates, it is probably necessary to model both the water mass and the sediments.

Introduction

Power plants to generate electricity, whether from fossil or nuclear fuels, are being located on estuaries or seacoasts because of the need for cooling water. The operation of nuclear plants is attended by the release of small quantities of liquid radioactive waste into the environmental water. Such releases are

strictly controlled by the U.S. Nuclear Regulatory Commission and are kept as low as is practicable, but it is still possible that seafood animals living in the adjacent waters may accumulate radionuclide burdens that could be transferred to man. Current forecasts predict that the contributions of nuclear fuels to electrical power

production in the United States will increase significantly by the year 2000. It is important, therefore, to model the marine food chain so as to be able to assess the maximum possible individual dose as well as the total population dose to man from the projected releases.

Models of the marine pathway of radionuclides to man have had two serious limitations: in the absence of directly relevant data, it has frequently been necessary (1) to use as input parameters the concentration factors for stable elements in the marine animals, and (2) to assume that the radionuclides are dispersed into and accumulated by the organisms solely from the soluble phase of the water. (The concentration factor (CF) is the ratio of the concentration in the organism ($\mu\text{g/g}$) to the concentration in the water ($\mu\text{g/ml}$). The limitation of stable-element CF values is that they are usually representative of steady-state conditions of uptake and loss by the organism (because under natural conditions the concentrations of the element in the ecosystem may undergo little change over intervals of several months), whereas steady-state conditions are rarely if ever attained in such ecosystems for the radioactive isotopes of most elements (because the radioactive releases are usually intermittent rather than continuous). The

limitation of assuming that the radionuclides are only in the soluble phase of the water is that they frequently interact with particulates, thereby changing their rates of dispersion and their availabilities to organisms.

Since this study was initiated to provide suitable data for dynamic modeling, the data collected were those relevant to the dynamics of radionuclide transfer among water, suspended particulates, sediments, and biota. The site selected for the experiment was the discharge canal of the Humboldt Bay Power Plant operated by the Pacific Gas and Electric Company. The discharge canal offers a relatively simple model ecosystem in which the necessary equipment is readily installed and serviced and in which water, suspended particulates, and sediments can be conveniently sampled along with experimentally introduced seafood organisms. Also, radionuclide analyses of environmental components are facilitated by the relatively high concentrations in the water of the discharge canal compared to other sites in the ecosystem that receive the radioactive waste.

The short-term patterns of radionuclide transfer among components of the ecosystem were assessed by examining at short-time intervals (1/4 to 24 hr) before, during, and after single

releases, the radionuclide concentrations in the water, in the particulates, and in groups of previously unexposed oysters placed in the canal. The long-term patterns were assessed by examining at long-term intervals (1 to 4 weeks) over a series of releases, the changes in radionuclide concentrations that occurred in the water, the particulates, and the population of oysters introduced into the discharge canal and sampled serially over an 18-month period.

This study of the transfer of released radionuclides will contribute to our understanding of the interactions of radionuclides among the biotic and abiotic components of ecosystems and will provide the information needed for the dynamic modeling of the fate of radioactive releases. The data obtained are relevant also to the dispersion and accumulation of the corresponding stable isotopes when they are introduced into marine waters as pollutants in effluents from industrial sources.

Site Description

The Humboldt Bay Power Plant, adjacent to Humboldt Bay near Eureka, California, produces electricity with two 54-MW(e) fossil-fuel units and a single 65-MW(e) boiling water reactor. Cooling water from the south part of Humboldt Bay is pumped from an inlet canal through the condensers of all three generating units into a discharge canal leading back to the central part of the bay (Fig. 1).

Liquid radioactive wastes from the stack, the power building and refueling area, and the laundry operation are accumulated and processed at the plant.¹ At irregular intervals, 10,000- to 40,000-l batches of this low-level waste are released via the discharge canal into the bay. This

is done by pumping the waste from a storage tank into the stream of cooling water just before it enters the discharge canal. The concentration of radioactivity at release are in accordance with the limits prescribed by the U.S. Nuclear Regulatory Commission Administration and the State of California North Coastal Regional Water Control Board.

The discharge canal is 140 m long and 12 to 21 m wide, depending on the tide level.² The bottom and sides consist of mud with a high content of organic material. The volume of water discharged into the canal depends on the number of units operating; the usual flow rate is about 40,000 l/min (three units in operation). The

Fig. 1. Humboldt Bay area, showing the location of the Pacific Gas and Electric Humboldt Bay Power Plant and its discharge canal in which the experimental station was located.

water temperature usually is about 22°C , but it may be higher or lower depending on the number of generating units in operation; the temperature

is elevated to about 28°C when the effluent is heated as part of the operation to remove mussels from the condenser tubes.

Materials and Methods

EXPERIMENTAL STATION

The experimental station installed at the discharge canal consisted of a redwood raft and an instrument shed. The raft was held in place in the center of the canal by a system of lines and pulleys that allowed it to be pulled into shore at will. It was floated on styrofoam pontoons and had openings in its floor through which were suspended the containers for holding oysters in the water stream and the trays for collecting samples of settled particulates at the same depth as the oysters.

The instrument system was developed to monitor automatically water temperature, solar illuminance, and conductivity, and to collect continuous integrating water samples over preset intervals.³ A probe containing a Ge(Li) detector provided a time history of radionuclide concentrations in the water during specific releases.

SAMPLING PROCEDURES

Seawater

Water samples from the discharge canals were collected either at selected intervals by pumping directly from the flowing stream for 15-min intervals or sequentially by the integrating water sampler with preset brief sampling periods of preset frequencies over intervals of one or two weeks.³

In the first case, about 200 l of seawater was filtered through 1- μ m cartridges and then acidified with HCl to approximately pH 1. For the sequential samples, the same volume was collected without filtration directly into vessels containing enough HCl to lower the final volume to approximately pH 1; the collected water was filtered just before analysis. In both cases, stable carrier was added for each radionuclide for determination of recovery.

Particulates

Suspended particulates were collected from the water samples either on 1- μ m Nucleopore filters or on 1- μ m cartridge filters.

Settled particulates were sampled at specific intervals throughout the experimental period, both from the collection trays suspended from the raft at the same level as the oysters and from weighted collection trays placed at the bottom of the canal. At the end of the collection period, all accumulated particulate material was removed from all collection trays.

Oysters

For each experiment, several hundred 3-year-old oysters (*Crassostrea gigas*) obtained from commercial beds in the north part of Humboldt Bay were introduced into the discharge canal. The oysters survived the

transfer well and showed no adverse effects from their maintenance in the discharge canal.

Short-Term Experiment - Two releases, one in July 1973 and the other in December 1973, were studied in detail by analysis of oysters, water, and particulates before, during, and after the releases. In each case, the role of particulates in radionuclide uptake by the oysters was evaluated by maintaining half of the oysters in filtered water and the other half in unfiltered water.

The oysters in unfiltered water were placed in the discharge canal in plastic cages suspended from the raft. Those in filtered water were kept on the raft in aquaria supplied with discharge canal water pumped through 1- μ m filter cartridges at about 30 l/min during the release and more slowly at other times depending on the sampling frequency. The pump intake was in the same area as the suspended cages. Rapid turnover of aquarium water was insured by placing the inflow at the bottom of each aquarium and by keeping the total volume of contained water small to minimize dilution of the inflowing water.

The oysters were sampled at intervals of 6 hr to one day. They were prepared for analysis at the site; shortly after sampling, they were

removed from their shells, rinsed in filtered seawater, and pooled to give a composite sample. In these 3-year-old oysters, the soft tissues weighed about 50 g and the shell was about 12 cm long.

Long-Term Experiments - For this experiment, all the oysters were placed in the water stream in plastic cages suspended from the raft. They were sampled at approximately 3-week intervals from August 1971 to January 1973. For each sample, oysters were removed from the water, their shell lengths were measured, and the soft tissues were separated from the shells. The soft tissues were rinsed in filtered seawater and pooled to give a composite sample and then the sample was weighed (Table 1).

ANALYTICAL PROCEDURES

Processing of the seawater, oysters, and particulates was completed at Lawrence Livermore Laboratory. For the seawater samples, radionuclides of cesium were collected onto ammonium phosphomolybdate (AMP) crystals which were then isolated from the liquid phase by filtration onto 1- μ m cartridges. Next, radionuclides of cobalt, manganese, and zinc were coprecipitated with ferric hydroxide, and after the precipitate had settled, the supernatant was

discarded. Chemical yields of the isotopes were determined by atomic absorption spectrometry.

For the oyster samples and the particulate samples from the trays, dry weights were determined after heating for a minimum of 48 hr at 103°C in an oven. Ash weights were determined after ashing to constant weight at 450°C in a muffle furnace.

For the samples collected on cartridge filters, the cord'ng that made up the filter bed was first cut from the central core, weighed while wet, dried to constant weight, and ashed to remove the organic material.

All samples were counted on Ge(Li) detectors and the counting data were recorded on tape for computer processing.

Table 1. Long-term oyster samples.

Collection date	No. of oysters	Av shell length (cm)	Total sample wt (g)		Av wt oysters (g)	
			Body	Shell	Body	Shell
Aug. 20, 1971	5	—	291	1245	58.2	249
Sept. 20, 1971	4	—	283	1246	70.8	312
Oct. 19, 1971	6	14.8 ± 2.4	280	1412	46.7	235
Nov. 10, 1971	6	12.3 ± 2.0	252	1823	42.0	304
Dec. 28, 1971	5	11.9 ± 1.4	285	1206	57.0	241
Jan. 21, 1972	6	12.9 ± 4.1	310	—	51.7	—
Feb. 15, 1972	6	—	229	—	38.2	—
Mar. 7, 1972	7	—	367	—	52.2	—
Mar. 29, 1972	7	—	353	—	50.4	—
Apr. 18, 1972	7	11.6 ± 2.0	380	1781	54.3	254
May 16, 1972	6	12.5 ± 1.5	275	—	45.8	—
June 6, 1972	7	12.6 ± 1.1	283	1776	40.4	254
July 25, 1972	5	12.4 ± 0.7	261	1257	52.2	251
Aug. 22, 1972	6	11.1 ± 1.7	292	1872	48.7	312
Sept. 15, 1972	6	12.1 ± 1.4	312	1603	52.0	267
Oct. 10, 1972	6	12.1 ± 1.0	316	1719	52.7	286
Nov. 7, 1972	6	11.7 ± 1.9	223	1636	37.1	273
Dec. 12, 1972	5	11.1 ± 1.9	242	1328	48.6	266
Jan. 9, 1973	6	12.3 ± 2.9	245	1764	40.9	294

Results

CHARACTERIZATION OF RADIONUCLIDE SOURCE

Between April 1, 1971 and December 31, 1973, an aliquot was taken from each batch of liquid waste just *before its release into the cooling water*. Each aliquot was analyzed for its content of gamma-emitting radionuclides and tritium. The radionuclides usually detected were ^3H , ^{54}Mn , ^{60}Co , ^{65}Zn , $^{134,137}\text{Cs}$, and ^{144}Ce . The total quantities of gamma-emitting radionuclides released are summarized briefly in Table 2.

Radionuclide concentrations varied widely from one release to another. The releases were highly irregular in frequency; during the experimental period, intervals between releases ranged from one to 34 days. In addition, the releases were variable in duration, usually covering about 6 hr

Table 2. Radionuclide characterization of liquid waste, April 1, 1971 to December 31, 1973.

Nuclide	Total per release (μCi)		
	Average	Maximum	Minimum
^{54}Mn	1000	25,000	2
^{60}Co	1900	36,000	22
^{65}Zn	1500	53,000	10
^{134}Cs	6100	61,000	22
^{137}Cs	9600	151,000	66
^{144}Ce	250	10,000	1

but occasionally as short as 2 hr. Detailed information on the conditions of the releases, the concentrations of the radionuclides in the waste, and the total quantities released are given in a *compilation of the radiological data collected by Lawrence Livermore Laboratory personnel at the Humboldt Bay site*.⁴

DISCHARGE CANAL WATER

Radionuclide concentrations in the discharge canal water could be expected to vary widely with time for several reasons: the waste itself was variable in composition; the hydrological conditions differed from one release to another; the intervals between releases were not constant. Therefore, to document the expected variability, ranges in concentration were determined both during time intervals when only a single release was occurring and during time intervals when several releases were occurring. *Concentration ranges were determined for the former by collecting samples at specific intervals before, during, and after releases and for the latter by collecting integrated samples over periods of one to two weeks.* This documentation is important for model development, since it is useful to be able to relate the radionuclide concentrations

in liquid waste before release to those expected for the water in which the food organisms reside.

Radionuclide Concentrations During Single Releases

Radionuclide concentrations in the discharge canal water varied during releases not only with those in the liquid waste but also with the rate of pumping from the waste retention tank and with the rate of flow through the discharge canal. A specific

dilution factor for each release was used to calculate an "expected concentration" (soluble plus particulate) for each radionuclide as it entered the discharge canal. (The dilution factor is the volume of cooling water pumped per hour divided by the volume of waste pumped into the cooling water per hour.) Hereafter, the term "expected concentration" refers to that calculated from the dilution factor. For the three releases of concern, the quantities of each

Table 3. Time and radionuclide composition of monitored releases.

	⁵⁴ Mn	⁶⁰ Co	⁶⁵ Zn	¹³⁴ Cs	¹³⁷ Cs	¹⁴⁴ Ce
April 4, 1973 (0700 to 1300)						
Liquid waste:						
Total release (μCi)	130	300	510	12,200	22,100	110
Concentration (pCi/liter)	4,260	8,870	16,900	403,000	732,000	3,500
Discharge canal water:						
Expected concentration (pCi/liter)	1.11	2.32	4.42	105.	191.	0.91
July 31, 1973 (0900 to 1100)						
Liquid waste:						
Total release (μCi)	200	530	375	10,700	17,500	90
Concentration (pCi/liter)	18,800	49,800	35,100	1,000,000	1,640,000	8,000
Discharge canal water:						
Expected concentration (pCi/liter)	4.85	12.8	9.05	258.	423.	2.06
December 4, 1973 (0930-1230)						
Liquid waste:						
Total release (μCi)	520	780	247	29,100	38,900	210
Concentration (pCi/liter)	32,100	47,600	15,200	1,790,000	2,390,000	12,700
Discharge canal water:						
Expected concentration (pCi/liter)	6.28	9.31	2.97	349.	467.	2.50

radionuclide released and the expected concentrations indicate that the concentration of each specific radionuclide was independent of all others and that the amounts released were different for each event (Table 3).

The validity of predicting concentrations in the water from the dilution of the waste was assessed by comparing expected and measured concentrations of γ -emitting radionuclides in the discharge canal water (soluble plus particulate) for two releases (Table 4). Measured values

(expressed as percentages of expected values) varied not only from one radionuclide to another but also from time to time during the release for each radionuclide. Radiocesium data are given here and subsequently only for ^{137}Cs because it and ^{134}Cs behaved in essentially the same fashion. No data are presented for ^{144}Ce since it was not generally detected in the discharge canal water.

The agreement between measured and expected values was good for ^{137}Cs compared to that for ^{54}Mn , ^{60}Co , or ^{65}Zn . During the release of July 31, 1973, measured values for ^{137}Cs were lower than expected at 0900 and 1100, probably because these two samples were collected during the beginning and end of the release period.

Continuous monitoring of the water with a Ge(Li) detector probe showed that $^{134,137}\text{Cs}$ did not reach maximum concentration until about 30 min after the release had started.⁵ The inflowing liquid waste appears to mix with the water already in the discharge canal rather than to replace it immediately. However, homogeneity probably was achieved by the time the inflowing water mass reached the sampling station, which was in the downstream half of the discharge canal. Concentrations were essentially the same in water samples collected at the surface and 0.3 m from the bottom of the canal.

Table 4. Measured radionuclide concentrations (soluble plus particulate) as percentages of expected concentrations in the water of the discharge canal during two releases.

	^{54}Mn	^{60}Co	^{65}Zn	^{137}Cs
April 4, 1973 ^a				
0800	65	43	65	92
0900	119	66	156	98
1000	63	22	133	104
1200	67	38	87	108
Mean	78	42	110	100
July 31, 1973 ^b				
0900	40	52	36	56
1000	41	80	73	102
1100	71	69	50	57
Mean	51	67	53	72

^aRelease began at 0700 and ceased at 1300.

^bRelease began at 0900 and ceased at 1100.

Radionuclide Concentrations Averaged Over Periods Of Several Releases

The long-term variability of radionuclide concentrations in the discharge canal water is more complex than variability during single releases because it encompasses not only variations within single releases but also the variations in their frequency and duration. To see this long-term variability it is necessary to compare concentrations averaged over different intervals each of which includes several releases. Average concentrations in water can be calculated for a specific radionuclide over specific intervals if it is assumed that the residence time of the radionuclide in the discharge canal is the same as that of the body of water into which it is diluted. The equation is:

$$C = \frac{\sum_{i=1}^{n+1} (W_i)t_i/D_i + (W_2)t_2/D_2 + \dots (W_1)t_1/D_1}{T} \quad (1)$$

where C = average concentration of radionuclide in water ($\rho\text{Ci/l}$) over total time T (hr),

W = radionuclide concentration in liquid waste ($\rho\text{Ci/l}$),

D = dilution factor,

t = duration of release (hr).

Average radionuclide concentrations in water were calculated with Eq. (1) for the period June 1972 to January 1973 during which one-week or two-

week integrated water samples were being collected (Table 5). Radio-cesium was quantitated in all samples; ^{54}Mn , ^{60}Co , and ^{65}Zn were usually below the detection limits in the samples collected from June through September and were not analyzed in December and January. Better agreement between the measured and expected concentrations was observed for ^{137}Cs than for other radionuclides. Percentage values are available for all four radionuclides only for those samples collected during intervals from September 29 to December 5. The average percentages over this period were about 140 for ^{137}Cs , 9500 for ^{54}Mn , 2000 for ^{60}Co , and 1400 for ^{65}Zn . The differences between measured and expected values for the four nuclides were in the order $^{54}\text{Mn} > ^{60}\text{Co} > ^{65}\text{Zn} > ^{137}\text{Cs}$. Since this order is the same as that of the affinities of the radionuclides for particulates,^{6,7,8} experiments were performed to obtain information on the partitioning of the radionuclides among the soluble phase of the water, the suspended particulates, and the settled particulates.

DISCHARGE-CANAL PARTICULATES

During the experimental period, radionuclide concentrations were determined both in suspended particulates separated from water samples

Table 5. Measured radionuclide concentrations as percent of expected concentration in the discharge canal water. Seawater collected by the sequential sampler from June 1972 to January 1973.

Sampling interval	^{137}Cs	^{54}Mn	^{60}Co	^{65}Zn
June 13-20	180	ND ^a	ND	ND
20-27(NR ^b)	1200	"	"	"
27-July 3(NR)	200	"	"	"
July 3-11(NR)	110	"	"	"
11-17	60	"	"	"
17-25	80	"	"	"
25-Aug. 1(NR)	170	"	"	"
Aug. 4-11	120	"	"	"
11-18	180	"	200	"
18-25(NR)	190	"	ND	"
25-Sept. 1	80	"	180	"
Sept. 1-8	80	"	24	"
8-15(NR)	370	"	ND	"
15-22	300	"	60	"
22-27(NR)	340	"	ND	"
29-Oct. 13	90	2,500	2,200	910
Oct. 13-27	100	7,300	2,100	1,200
27-Nov. 10	250	9,600	4,200	260
Nov. 10-22	170	28,000	1,400	4,700
22-Dec. 5	100	180	160	45
Dec. 5-15	340	NA	NA	NA
19-23	240	"	"	"
23-30	210	"	"	"
30-Jan. 2(NR)	280	"	"	"
Jan. 2-5(NR)	92	"	"	"
5-9(NR)	62	"	"	"
9-12	63	"	"	"

^aND, not detected; NA, not analyzed.

^bNR, no releases during sampling interval.

and in settled particulates collected at the level of the oyster containers and at the bottom of the canal. Concentrations were determined both for short-term (1/4- to 6-hr or daily) and for long-term (near weekly) collection periods. To permit direct comparisons among the different radionuclides for any one release or from one release to another, the measured concentrations are expressed as percent of the expected concentrations (pCi/l).

Suspended Particulates

The percentage of the total expected concentration found in the suspended particulate varied during single releases and differed greatly from one release to another (Tables 6 and 7). They were highest in the April 4, 1973 release and lowest in the December 4, 1973 release. Continuous monitoring of the particulate fraction after the December release showed periodic increases and decreases in radionuclide recoveries (Table 7).

The variations observed in the radionuclide concentration in suspended particulates, expressed on the basis of unit volume, could have resulted either from the same quantity of particles (mg/l^3) with different affinities for radionuclides or from different quantities of particles with the same affinity for radionuclides. The particulate load in the discharge

canal water at the sampling site differed from release to release and with time during a single release (cf. Tables 6 and 7). Examination of the percent data indicates that the differences in concentration between releases were greater than that which can be accounted for by differences in particle load. For ^{54}Mn , the highest value during July was 85 percent and the highest value during December was 35 percent but the largest particle load during July was 28 mg/l and the largest load during December was 15 mg/l . These data suggest that the radionuclide affinity of the particles was not the same in December as it was in July. These differences in affinity may reflect seasonal changes in the physical and chemical characteristics of the particles.

Radionuclide concentrations in the particulates were calculated on a per-unit-weight basis as well as on a per-unit-volume basis. Concentrations recovered in particulates (pCi/mg) per unit of radioactivity released were highest in April and lowest in December.

One attempt was made to look at the effect of particle size on radionuclide partitioning between water and particulates. During the July 31, 1973 release, the water samples were filtered consecutively through 10-, 5-, and 1- μm filters. For all

Table 6. Measured radionuclide concentrations expressed as percent of expected concentration for the particulate and soluble fractions in discharge canal water before, during, and after a release in April 1973 and another in July 1973. The April release began on April 4 at 0700 and ceased at 1300. The July release began on July 31 at 0900 and ceased at 1100.

Collection ^a	Particulate dry weight (mg/L)	Percent				
		Fraction ^b	⁵⁴ Mn	⁶⁰ Co	⁶⁵ Zn	¹³⁷ Cs
April 3, 1973						
1630	8.3	P	<4	<2	5 ^c	<0.02
		S	15 ^c	5 ^c	<4	0.12
April 4, 1973						
0630	7.4	P	<4	<2	<4	<0.02
		S	NA	NA	NA	0.24
0800	7.9	P	54	29	25	0.8
		S	11 ^c	14 ^c	40	91
0815	7.0	P	37	26	22	0.8
		S	NA	NA	NA	NA
0900	9.3	P	74	20	43	0.9
		S	45	38	113	97
1000	12.8	P	22	22	15	1.0
		S	41 ^c	<2	118	103
1100	8.5	P	22	23	21	0.9
		S	NA	NA	NA	102
1200	8.1	P	33	22	25	1.9
		S	34	16	62	106
1400	3.6	P	14 ^c	12	11 ^c	0.3
		S	NA	NA	NA	55
1600	4.5	P	9 ^c	5 ^c	<4	<0.02
		S	<20	<13	<10	0.5
1800	4.1	P	<4	3 ^c	<4	0.06 ^c
		S	NA	NA	NA	0.1
July 31, 1973						
0810	22	P	<1	<0.3	<2	<0.02
		S	<0.6	<0.2	<1	0.1
0900	25	P	29	22	15	1
		S	11	20	21	55
1000	28	P	20	14	20	0.9
		S	21	66	53	101
1105	19	P	58	31	16	0.1
		S	13	38	34	57
1200	18	T	NA	NA	NA	3

Table 6. (continued)

Collection ^a	Particulate dry weight (mg/l)	Percent				
		Fraction ^b	⁵⁴ Mn	⁶⁰ Co	⁶⁵ Zn	¹³⁷ Cs
1430	16	T	NA	NA	NA	2
2000	13	T	NA	NA	NA	0.2
August 1, 1973						
0230	9	T	NA	NA	NA	0.3
0830	18	T	NA	NA	NA	0.2

^aTime of initiation of collection; sample collected for 15 min.

^bP, particulate (>1 µm); S, soluble (<1 µm).

^cFSD >0.25 (FSD, fractional standard deviation, is the standard deviation divided by the mean).

^dNA, not analyzed.

the radionuclides, the largest fraction of the activity was associated with the largest particulates, those retained on the 10-µm filters (Table 8). The largest changes in percent with time were observed for ⁵⁴Mn on the 10-µm filter. In general, percents in the samples were in the order ⁵⁴Mn > ⁶⁰Co > ⁶⁵Zn > ¹³⁷Cs. The concentration ratios differed not only with time but in some cases from one filter to another at a given time.

Settled Particulates

The kind of information that can be derived about the behavior of particulates in the discharge canal obviously depends on the frequency of sampling and the overall period of observation. Daily or more frequent sampling before, during, and after a single release would give an indica-

tion of the dynamics of short-term reactions. With less frequent sampling over longer periods seasonal changes can be evaluated as well as the effect of differences in the interval between releases.

Short-Term Behavior - Variations in radionuclide concentration in the suspended particulate fraction of the discharge canal water during and after releases could result from changes in the amounts resuspended from the bottom sediments and/or the influx of radioactive particles recirculating from the Bay. The experiment of April 1, 1973 was set up to determine the possible role of the bottom sediments in this variation. During the release, settled particulates were sampled serially over the same sampling intervals both from the trays

Table 7. Measured radionuclide concentrations expressed as percent of expected concentration for the particulates in discharge canal water preceding, during and after a release that began on December 4 at 0930 and ceased at 1240.

Collection time	Particulate dry weight (mg/l)	Percent			
		⁵⁴ Mn	⁶⁰ Co	⁶⁵ Zn	¹³⁷ Cs
December 3, 1973					
1535-1740		0.25	0.11	2.0	0.01 ^a
1740-2200		0.53	0.62	0.67	0.02
December 4, 1973					
2200-800		2.2	1.9	1.8	0.014
0830-0930		0.2	0.15	0.64 ^a	0.05
0930-1030		2.8	4.0	4.1	0.35
1030-1130	14.8	3.5	4.5	8.4	0.60
1130-1230		3.0	4.4	6.8	0.62
1230-1330		0.25 ^a	0.24	1.1 ^a	0.031
1330-1545	15.6	0.64	0.80	1.9	0.024
1545-2000		0.14	0.10	0.74 ^a	0.003
December 5, 1973					
2000-0830		0.06 ^a	0.10	≤0.03	0.006
0839-1345		2.4	3.0	1.3	0.049
1345-1610		0.018 ^a	0.40	≤0.03	0.018
1610-1930		0.4	0.50	≤0.03	0.016
December 6, 1973					
1930-0830		0.27	0.50	0.67 ^a	0.014
0830-1230		0.02 ^a	0.01 ^a	≤0.06	0.001 ^a
1230-1730		1.1	1.2	1.5 ^a	0.027
1730-2300		0.05 ^a	0.08 ^a	0.34 ^a	0.003
December 7, 1974					
2300-0830		0.65	0.90	1.4 ^a	0.063

^aFractional standard deviation >0.25.

placed on the canal bottom and from the trays suspended at the same depth as the oysters.

Collection trays were placed on the canal bottom at three different

sites, the first about 20 ft from the point of entry of liquid waste into the canal, the second in the vicinity of the raft (about midway between the point of entry of the liquid waste

Table 8. The measured radionuclide concentrations as percent of expected concentrations and the concentration ratios in particulates during the July 31, 1973 release.

Collection time	Filter (µm)	Percent				Concentration ratios		
		^{54}Mn	^{60}Co	^{65}Zn	^{137}Cs	$^{60}\text{Co}/^{54}\text{Mn}$	$^{65}\text{Zn}/^{54}\text{Mn}$	$^{137}\text{Cs}/^{54}\text{Mn}$
0900-0915	10	22.0	17.0	10.0	0.45	0.77	0.45	0.020
	5	3.5	2.5	3.1	0.25	0.71	0.89	0.071
	1	3.7	2.3	2.0	0.26	0.67	0.54	0.070
	Total	29.2	21.8	15.1	0.96			
	Mean ratio					0.70	0.66	0.054
1000-1015	10	12.0	8.1	11.0	0.44	0.68	0.91	0.037
	5	5.6	3.7	4.9	0.26	0.66	0.88	0.046
	1	2.7	2.9	4.1	0.20	0.71	1.50	0.070
	Total	20.3	13.7	20.0	0.90			
	Mean ratio					0.68	1.10	0.051
1105-1120	10	47.0	24.8	13.0	0.10	0.53	0.28	0.0021
	5	7.8	4.3	2.4	0.02	0.55	0.31	0.0026
	1	2.7	1.6	1.1	0.02	0.59	0.41	0.0074
	Total	57.5	30.7	16.5	0.14			
	Mean ratio					0.56	0.33	0.0040

and the outlet into the central bay), and the third about 20 ft upstream from the outlet. Radionuclide concentrations detected in particulates from trays located nearest the inlet were about the same or higher than those near the midway point; those closest to the outlet were the lowest (Fig. 2). The concentrations detected in particulates from the midway point varied least with time. The observed consistency in the concentrations may have been due to the fact that the area was least subject to turbulence due to changes in flow rate and least

affected by influx or back-flux of unlabeled sediments from the Bay.

The settled particulates from the suspended trays were similar in radionuclide concentration to those from the bottom trays during the release period but lower and more variable with time in subsequent samples (Fig. 3). When the concentrations of ^{60}Co , ^{65}Zn , and ^{137}Cs in the particulates from bottom trays and suspended trays are plotted against the concentrations of ^{54}Mn , the values are distributed around a straight line (Fig. 4), as they would be if

Fig. 2. Radionuclide concentrations (pCi/g dry wt) in serial samples of bottom particulates from three sites in the discharge canal during the April 1973 experiment. Circles, 20 m downstream from the beginning of the canal; squares, near the midway point; triangles, 20 m upstream from the outlet. Closed symbols, FSD < 0.2; open symbols FSD > 0.2.

the particulates from suspended and bottom trays were derived from the same source.

Long-Term Behavior — Radionuclide concentrations in the particulates collected on suspended trays varied

Fig. 3. Radionuclide concentrations (pCi/g dry wt) in serial samples of particulates from the discharge canal during the April 1973 experiment. Squares, particulates collected on trays placed on the floor of the canal in the vicinity of the raft; circles, particulates collected on trays suspended from the raft at the same level as the oysters. The duration of the sample collection is indicated by the bracketed lines on the square symbol on the ^{54}Mn graph. Closed symbols, FSD < 0.2; open symbols, FSD > 0.2.

widely during the 16-month interval of the study. In general, the variations reflected changes in the

quantities of radionuclides released into the discharge canal water.

For ^{54}Mn , the concentrations from March through July ranged from about 10^3 to 10^4 pCi/kg dry weight (Fig. 5). They increased by more than an order of magnitude during the fall of 1972 when larger quantities of ^{54}Mn were

Fig. 4. Concentrations of ^{60}Co , ^{65}Zn , and ^{137}Cs related to those of ^{54}Mn in bottom particulate samples collected during the April 1973 experiment. All concentrations are expressed on a dry weight basis. The samples are those for which data are shown in Fig. 2. Solid triangles, bottom trays; open triangles, suspended trays. Solid line, regression of ^{60}Co , ^{65}Zn , or ^{137}Cs concentration on ^{54}Mn concentration for the sample. Dashed lines indicate ± 1 S.D. from regression for the sample.

released. The highest concentration measured (3.1×10^6 pCi/kg) was in the sample collected after the October 6 release. Even in those samples collected in periods when no releases were made, the concentrations never fell below 1.5×10^3 pCi/kg.

For ^{60}Co , the concentrations were usually higher than for ^{54}Mn (Fig. 4), partly because more ^{60}Co was released (on the average about twice as much). Concentrations usually were between 5×10^3 and 5×10^4 pCi/kg dry weight; the highest was 4.0×10^6 and the lowest was 3×10^3 pCi/kg.

For ^{65}Zn , the concentrations and the patterns of variation were similar to those for ^{54}Mn . The highest concentration measured was 1×10^6 and the lowest was 7×10^2 pCi/kg dry weight (Fig. 5).

For ^{137}Cs , the concentrations were usually between 5×10^2 and 5×10^3 pCi/kg. The detected concentrations for ^{137}Cs were lower than those for ^{54}Mn , even though most releases contained about ten times more ^{137}Cs than ^{54}Mn (Fig. 5). The highest concentration measured was 3.7×10^4 and the lowest was 1×10^2 pCi/kg dry weight.

The settled particulates from the suspended trays varied in quantity from 1 mg/cm^2 to almost 12 mg/cm^2 per day (Fig. 6). Quantities were higher in July and August than in November and December. The deposition rate may be related to the resuspension of

bottom sediments during periods of changing hydrological conditions, but Fig. 6, which includes data on the maximum low tides, indicates no obvious relationship with the low tide level.

Dynamics of Radionuclide Transfer between Seawater and Particulates

Some indication of the dynamics of interaction between water and particulate compartments can be obtained by following the concentration changes of radionuclides in settled particulates during intervals covering large differences in the quantities released and/or frequencies of releases. For example, there were two periods during which the interval between releases was very large — between June 13 and July 17, 1972, and between October 6 and 27, 1972. These two periods are of special interest because before the first, the releases were relatively constant in frequency and quantity, whereas before the second, there was an unusually large release (October 6) which, in effect, pulse-labeled the system. During the June 13 — July 17 interval (34 days), the concentrations in the settled particulates showed no consistent decreases with time for any of the four radionuclides; the variations in concentrations appear to be related to changes in deposition rates (Table 9). However, during the October 6-27 interval (21 days), there was a

decrease in the concentrations of the four radionuclides. Particulates were collected on October 10, 17, and 24; concentrations were lower on October 24 than on October 10 by a factor of 10. The rates of change were similar for ^{54}Mn , ^{60}Co , and ^{65}Zn but higher for ^{137}Cs .

OYSTERS

Short-time radionuclide accumulation in oysters in the discharge canal was studied during two releases by serial sampling of previously unexposed oysters at 6-hr to daily intervals before, during, and after each release. In addition, long-term accumulation over a period that encompassed a number of releases was followed in oysters sampled at near monthly intervals over a period of 18 months.

Short-Term Accumulation

The two release periods examined were chosen to contrast a period of high biological productivity (July 1973) and a period of low biological productivity (December 1973). In each case, half the oysters were maintained in filtered seawater and the other half in unfiltered seawater.

In the July experiment, the oysters were placed in the canal water at 1700 on July 30. The prerelease sample was taken nearly 14 hr after placement of the oysters, at 0850 on July 31. The

Fig. 5. Concentrations of ^{54}Mn , ^{60}Co , ^{65}Zn , and ^{137}Cs in serial samples of oyster soft tissues (closed triangles) and of particulates collected on suspended trays (circles) compared to expected concentrations in the water during releases (bars). Open triangles, concentrations in oysters were below detection levels.

Fig. 5. (continued)

single postrelease sample was taken about 2 hr after the first sample, at 1100. The results are given in Table 10. In the prerelease oysters from unfiltered water, only ^{65}Zn was detected; in those from filtered water, ^{65}Zn was higher and ^{54}Mn was detected also. The higher values obtained from filtered water may have been due to leaching of radionuclides from particulates collected on the filters over the 8-hr collection period. Entrapment of particulates on the filter could

have resulted in an increase in their normal residence time in the discharge canal so that particles collected early in the sampling period could have been leached later by water containing radionuclides at lower concentrations. In the postrelease oysters, all radionuclides were detected in higher concentrations than in prerelease oysters and all concentrations were higher in the oysters from unfiltered water. Filtration of the seawater medium lowered the rate

Fig. 6. Lower curve, deposition of particulates on the suspended collection trays during the 18-month experimental period. Upper curve, low tide values (ft).

Table 9. Concentrations of radionuclides (pCi/kg dry wt) in settled particulates for two intervals without releases (June 13-July 17, 1972 and October 6-27, 1972). Also shown for each sampling interval are the deposition rates for the settled particulates and the tidal extremes.

Collection date ^a	Deposition rate (mg/cm ² /day)	Tide (ft)		⁵⁴ Mn	⁶⁰ Co	⁶⁵ Zn	¹³⁷ Cs
June 15	7.6	+7.6	-2.2	3,400	4,900	1,700 ^b	2,600
June 23	9.6	+6.3	-0.4	7,500	16,400	4,300 ^b	990
June 30	6.8	+6.6	-1.1	1,600	4,700	1,500 ^b	650
July 7	11.5	+7.1	-0.8	8,100	22,600	3,600 ^b	960
July 14	9.2	+7.5	-1.8	1,900	3,900	ND ^c	440
October 10	7.6	+6.4	+0.1	3,130,000	3,990,000	1,060,000	43,600
October 17	8.2	+6.0	+0.2	500,000	640,000	173,000	11,100
October 24	11.6	+7.7	-1.5	210,000	266,000	70,300	2,800

^aOn each collection date, all particulates were removed from each collection tray.

^bFractional standard deviation >0.25.

^cND, not detected.

Table 10. Normalized radionuclide concentrations^a in oysters maintained in unfiltered water in the discharge canal of the Humboldt Bay Power Plant during a period of high biological productivity (July 31, 1973) and a period of low biological productivity (December 4, 1973).

		¹³⁷ Cs		⁵⁴ Mn		⁶⁰ Co		⁶⁵ Zn	
		Non-filtered	Filtered	Non-filtered	Filtered	Non-filtered	Filtered	Non-filtered	Filtered
<u>High biological productivity period (July 1973)</u>									
Prerelease (pCi/kg)	ND ^b	ND	ND	9.5	ND	ND	ND	17	22
Postrelease	6.76	0.52	28	19.4	20	2.6	83	61	
Rate (pCi/hr) ^c	0.38	0.26	14	5.0	10	1.3	33	20	
<u>Low Biological productivity period (December 1973)</u>									
Prerelease (pCi/kg)	ND	ND	11	13	3.3 ^d	ND	40 ^d	22 ^d	
Postrelease (pCi/kg)	1.1	1.1	9 ^d	25	4.3 ^d	ND	67	67	
Rate (pCi/hr) ^c	0.37	0.37	—	4.0	0.3 ^d	—	9 ^d	15 ^d	

^aNormalized: measured concentration (pCi/kg) divided by expected concentration in discharge canal water (pCi/liter).

^bNot detected.

^cAmount accumulated per hour during the release.

of accumulation by 65% for ^{54}Mn , by 90% for ^{60}Co , by 40% for ^{65}Zn , and by 30% for ^{137}Cs .

In the December experiment, the oysters were placed in the discharge canal at 1230 on December 3. The prerelease oyster sample was taken 8-1/2 hr later, at 0900 on December 4, and the postrelease sample 3-1/2 hr after that, at 1230 on the same day. The results are given in Table 10. In the prerelease samples, ^{54}Mn , ^{60}Co , and ^{65}Zn were detected in oysters from unfiltered water, but only ^{54}Mn and ^{65}Zn were detected in those from filtered water. The rates of accumulation in the oyster during the release period were higher in July than in December for all the radionuclides except ^{137}Cs . For ^{137}Cs , the accumulation rates were very similar during both periods, suggesting that the higher July rates for the others reflected differences in radionuclide availability rather than different conditions in the aquaria and cages. The differences in accumulation rates between July and December were greatest in unfiltered water, again except for ^{137}Cs . The December rates were lower by 100% for ^{54}Mn and ^{60}Co and by 70% for ^{65}Zn ; they were essentially the same for ^{137}Cs .

In the December experiment, three additional postrelease samples of oysters were taken during the three days following the release. Figure 7

shows the relationship between the radionuclide concentrations in the oyster soft tissues in all five samples and the concentrations in the suspended particulates during the same period. (Note that the values plotted in the graph are not normalized and therefore not directly comparable to those normalized values in Table 10.) In the oysters the four radionuclides exhibit quite different patterns of accumulation and loss in relation to their concentrations in the particulates.

^{137}Cs

The concentrations in the oysters were essentially unaltered by filtration of their seawater medium. In both media, the concentration in the oysters rose very sharply during the release, then decreased rapidly during the next 24 hr and much more slowly during the subsequent days (Fig. 7). The concentrations in the particulates were elevated during the release but fell off immediately thereafter, except for small increases between the high-high tides and the succeeding low tides.

^{65}Zn

The patterns of accumulation and release by oysters were considerably altered by filtration of the medium (Fig. 7). The slower decreases in concentration in unfiltered water appeared to reflect continued input

Fig. 7. Radionuclide concentrations in serial samples of oysters (pCi/kg wet wt) and suspended particulates (pCi/l) collected during the December 1973 short-term experiment. Zero time, 1530 on December 3, is the time the oysters were placed in the discharge canal water. Solid triangles, oysters in filtered seawater; open triangles, oysters in unfiltered seawater. The first two sets of points are the prerelease and postrelease samples listed in Table 10. The shaded portions of the rectilinear curve (suspended particulates) indicate the release intervals.

of ^{65}Zn from the suspended particulates, where periodic increases may bear some relation to changes in the tidal cycle.

^{60}Co

In the oysters in unfiltered water, changes in concentration were irregular and clearly reflected changes in the concentrations in the particulate fraction (Fig. 7); ^{60}Co

was almost absent in oysters from filtered water (see Table 10). In the suspended particulates, concentrations were elevated over several of the sampling periods following the release; they were highest in those samples collected between a high-high tide and the subsequent low tide. The periodicity of the changes suggest that the variations

are related to changes in the tidal cycle. The relatively high concentrations in the prerelease samples are probably attributable to the two preceding releases (16,000 μCi on November 27 and 4000 μCi on November 30). The quantities were much higher than in the December 4 release (780 μCi); the discharge canal sediments probably contained a considerable amount of residual ^{60}Co . Consequently, the accumulation during the December experiment appears to reflect not only the December 4 release but the preceding two releases as well.

^{54}Mn

Filtration of the seawater medium affected the rate and maximum level of ^{54}Mn accumulation in the oysters, but the two curves are similar in shape, in both cases reflecting changes in the suspended particulates (Fig. 7). As with ^{60}Co , concentrations in the particulates were relatively high in the prerelease samples, probably because of the large quantities of ^{54}Mn in the two preceding releases (11,500 μCi on November 27 and 2800 μCi on November 30; only 520 μCi was released on December 4).

Long-Term Accumulation

^{54}Mn

During the first 12 months of the

experiment, concentrations of ^{54}Mn in the oysters were generally several hundred picocuries per kilogram wet weight (Fig. 5). During periods of low release rates, concentrations were below detection limits. After the series of large releases in the Fall of 1972, concentrations in the oysters reached more than 1700 pCi/kg. The particulates as well as the oysters reflected the quantities released, but concentrations in the oysters were lowered and less variable than those in the settled particulates (pCi/kg dry weight).

^{65}Zn

Concentrations of this radionuclide were about 10 times higher and were less variable than those of ^{54}Mn (Fig. 5). The differences between ^{54}Mn and ^{65}Zn were greater than expected from the average concentrations released (Table 2), suggesting that the two nuclides have different metabolic pathways. As with ^{54}Mn , concentrations in the oysters reflected changes in the quantities released; the maximum (100,000 pCi/kg) was in December 1972.

^{60}Co

Concentrations of ^{60}Co were lower and more variable than those of ^{65}Zn (Fig. 5). The average quantity released was greater for ^{60}Co than for ^{65}Zn (Table 2), but

^{60}Co concentrations in the oysters were less than 1000 pCi/kg except in the Fall of 1971 and 1972 when the releases were larger and more frequent.

^{137}Cs

Concentrations of ^{137}Cs in the oysters were generally less than 100 pCi/kg (Fig. 5), even though the average quantity released was considerably greater than for the other radionuclides (see Table 2). During the Summer of 1972, concentrations in the oysters were below detection levels.

Concentration Factors

Comparisons of the concentrations of radionuclides in the oysters to those in the water provide an indication of the ability of the oysters

to accumulate the radionuclides. Calculations of concentration factors for the discharge canal oysters were complicated by the absence of steady-state conditions in the ecosystem due to the irregularity of the releases and the changing hydrological conditions. However, concentration factors were calculated for each sampling interval over the 18-month experimental period, by dividing the concentrations in the oysters by the average expected concentrations (see Eq. (1)) of the water during the period preceding their removal. The periods between oyster samplings ranged between 21 and 49 days. During the 18-month period, the average concentration factor (CF) was about 18,000 for ^{54}Mn , about 6000 for ^{60}Co , about 120,000 for ^{65}Zn , and about 50 for ^{137}Cs .

Discussion and Conclusions

The radionuclide burdens in seafood animals living in waters adjacent to nuclear power plants are dependent on the concentrations of radionuclides in the environmental water and on their availability to food-chain organisms. The concentrations in water are dependent on the quantities released, the specific hydrodynamic conditions, and the interactions of the radionuclides with the particulates in the system. The availability is dependent

in part on the physicochemical form of the radionuclide in the water.

SEAWATER

Radionuclide concentrations in the seawater of the Humboldt Bay environment varied widely with site and with time. Some indication of the expected differences in ^{137}Cs concentration with site can be obtained from Fig. 8, where average concentration values for

^{137}Cs are plotted for the waste water and the successive sites of dilution: the discharge canal, Central Bay, and Humboldt Bay as a whole. For these calculations it was assumed that there was instantaneous dilution into each successive body of water and that the radionuclides from one release were totally washed out before the next release occurred. The largest reduc-

tion in concentration resulted from the dilution of the liquid waste by the cooling water.

The concentrations of radionuclides at each dilution site varied considerably with time due to differences in the amounts released, in the conditions of the release, and in the frequency of release. The greatest variation resulted from the differences in the

Fig. 8. Changes in ^{137}Cs concentration at successive dilution sites in its passage from the reactor to Humboldt Bay. Points, mean concentrations; bars, variations due to differences in quantity released, flow rate of cooling water, and stage of tidal cycle.

quantities released; for ^{137}Cs the minimum was 66 μCi and the maximum was 151,000 μCi (Fig. 8). The variation due to differences in flow rate through the canal was about a factor of 2; the minimum dilution factor was 2406, the maximum was 5170. The variation due to stage of tidal cycle at the time of release was about a factor of 2 for Central Bay and about 1.7 for Humboldt Bay as a whole. The variations in concentration due to differences in the frequency of release were less than those due to the quantities released but more than those due to changes in flow rate and stage of tidal cycle. The minimum interval between releases was one day, the maximum was 34 days.

Because ^{54}Mn , ^{60}Co , and ^{65}Zn interact with particulates in the sea water, their concentrations at different sites might be expected to vary more than that of ^{137}Cs or that predicted by hydrodynamic conditions and the quantities released. On the average, measured values were more variable and lower during release periods and higher between release periods. The lower concentrations during release periods may be a result of radionuclide binding to particulates on the bottom and sides of the canal upstream from the sampling station. If such depletion of radionuclides from the water occurs, the discrepancy between measured and

expected concentrations of radionuclides with a high affinity for particles might increase with distance from the source. No measurements were made to test this hypothesis.

Measured and expected radionuclide concentrations were compared during release of water from the cooling canal of the Oyster Creek nuclear power plant in Ocean County, New Jersey.⁹ For radionuclides that were retained quantitatively on filters and ion-exchange columns, the ratios of measured to predicted values were 0.6 to 1.2 with one exception. During one discharge, ratios were four to five times lower than expected. Deviations from a ratio of one were attributed to the following uncertainties in these measurements:

- dilution factors based on nominal values of waste release and canal water,
- assumption of a homogeneous mixing of waste and canal water,
- possible deposition of radionuclides from canal water prior to sampling or resuspension from the canal sediment,
- recirculation effects not measured simultaneously with discharge canal sampling, and
- low concentrations of radionuclides in liquid wastes following dilution with sea water.

In both this study and the one at Oyster Creek,⁹ the largest fraction of radionuclides in the sea water of the discharge canal during releases was associated with the particulates. In the Oyster Creek study, 60 to 100% of the ⁵¹Cr, ⁵⁴Mn, and ⁶⁰Co and large fractions of ⁹⁵Nb, ⁹⁵Zr, ¹⁴⁰Ba, ¹⁴¹Ce, and ¹⁴⁴Ce were associated with particles. Thus, in model development, it is important to consider that some elements do not behave conservatively, i.e., their concentrations are not directly proportional to the salinity of the water. Consequently, their concentrations cannot be calculated as simple dilutions of the source material. In addition, dilution parameters may vary from one site to another, and biological and geochemical factors may change with time.

PARTICULATES

The particulates in the discharge canal can be considered to be in two compartments, suspended and settled; radionuclides are constantly interchanged between the two compartments. The dynamics of interactions in the discharge canal were investigated by following the changes in radionuclide concentrations in the two compartments subsequent to scheduled releases.

Suspended Particulates

Comparisons of measured radionuclide

concentrations in the soluble and/or particulate phases of the water at any time for a given release indicated that the radionuclides differed in their affinities for the particulates. These differences in affinity are related to the chemical properties of the radionuclides and were in the same order (⁵⁴Mn > ⁶⁰Co > ⁶⁵Zn > ¹³⁷Cs) as the sequences determined by others for the distribution coefficient of the same radionuclides.^{6,7}

Radionuclide concentrations in the suspended particulates (pCi/l) varied greatly from one release to another. These differences probably were due to differences in particle size and composition, which in turn undoubtedly depended on biological processes occurring in the south part of Humboldt Bay. Since South Bay is shallow, light easily penetrates to the bottom; the mud flats support extensive beds of eelgrass. Measurements of phytoplankton productivity and eelgrass standing stock during the previous year (1972) indicated higher biological activity in July and August than in December.¹⁰ Increases in phytoplankton productivity would have resulted in direct increases in suspended particulates; increases in eelgrass standing stock would have resulted in indirect increases (suspended detritus from the decomposition of decaying plant material).

Radionuclide concentrations for a given radionuclide also were observed

to change with time in the suspended particulates subsequent to releases. These changes are probably attributable to variations in the rate of resuspension of the bottom sediments, in the amounts of radionuclides bound to these sediments, and in the quantities of unlabeled particulates in the inflowing water. Another possible source of these changes may be the influx of radioactive particles contained in water flowing into the discharge canal. There is evidence of recirculation of water from the discharge canal.^{11,12} This recirculation appears to take place under certain tidal conditions and is not a general occurrence; however, when it does occur, the inflowing water may contain both radioactive particles from the discharge canal and unlabeled particles from the Bay.

The largest fraction of the radioactivity in the suspended particulates was collected on the 10- μ m filters. These larger particles would tend to settle out more rapidly than the smaller ones and thus would be more likely to be present in greater quantity in the settled particulates in the discharge canal and its vicinity than in the settled particulates in the open Bay.

Settled Particulates

The bottom sediments in the discharge canal represent a pool of

particulate-bound radionuclides that appears to control the concentrations of radionuclides in the water between releases. Resuspension of these bottom sediments is affected by the flow rate of the cooling water into the canal and by the tidal level and wave action of the water in the bay. In the interval between releases time-related decreases with time in the radionuclide concentrations of the particulates that settled on the bottom were probably due to dilution with nonradioactive particles present in water flowing in from the Bay as well as to desorption of radionuclides. Measurements of the particulate loads in the intake canal water indicated that the unlabeled particulates in the intake canal water were sufficient in quantity to reduce the radionuclide concentrations in the bottom sediments when they were mixed with them.

Bottom sediments can be considered to consist of a historical (unmixed) layer and a mixed layer.¹³ In the discharge canal the homogeneity of the mixed layer depends not only on physical and chemical processes but also on the frequency of the radioactive releases and the magnitude of the variations in quantity discharged from one release to another. If the mixed layer is heterogeneous and the depth of scouring varies with tidal cycle and flow rate, one would expect

variations in radionuclide concentration like those observed here in the particles deposited on the suspended trays and separated on our filter cartridges.

During the 34-day interval after June 13, 1972 when no releases occurred, the radionuclide concentrations changed very little in the settled particulates collected on the suspended trays; they were not significantly lower than at other times. In this case, the absence of a large decrease may be related to the rates of turnover of radionuclides in particulates. Let us consider briefly radionuclide turnover in particulates in the discharge canal. Uptake and loss of radionuclides by particles involves both relatively rapid reactions and slow reactions.¹⁴ The more rapid include ion exchange reactions and nonspecific absorption reactions. The slow reactions include penetrations into the interstices of crystal matrices and into the broken ends of crystal lattices, and/or penetration through the thin layer of organic material covering the particles.⁷ With each new release in the discharge canal, the bottom particulates were exposed to elevated radionuclide concentrations, and the overall result was periodic increases and decreases in the radionuclide concentrations in these particulates. However, if the intervals between releases were about

the same, and if the quantities released differed by less than an order of magnitude, then the concentrations of the interstitial water of the sediments probably remained high enough for sufficiently long periods to permit relatively complete labeling of those particulate compartments with slow turnover times. Such a series of releases occurred before the 34-day interval without a release that followed the release of June 13, 1972. Consequently, concentration changes over most of this interval probably were due primarily to loss of radionuclides from compartments with slow turnover rates. Thus, one would expect only small changes with time.

Another long interval without a release occurred after the release of October 6, 1972. Large decreases in radionuclide concentrations were observed in the settled particulates in the 21-day interval following this release. These changes probably resulted from the desorption of radionuclides from the compartments with rapid turnover times; the latter had been labeled by the exceptionally large amounts of radionuclides in the release. However, the uptake of radioactivity by particulates appeared to be considerably more rapid than its loss. The seawater maintained its high radioactivity levels only during the six hours of the release, but the particulates remained higher than in

the prerelease period for the entire 21-day interval until the next release. These data indicate that the half-time of radionuclides is considerably longer in the particulate compartment than in the water.

The relationships among the soluble phase in the water, suspended particulates, and settled particulates can be summarized as follows. Suspended particulates in the discharge canal water appear to be derived from two sources: inflowing water from the Bay and bottom sediments. During release periods, the radionuclide concentrations would in general be higher in the inflowing water than in the bottom sediments, whereas during interrelease periods the reverse would be expected. If over a given interval of time, whether during or between releases, there is a constant rate of influx of particles from the Bay and a constant rate of resuspension of bottom sediments, changes in radionuclide concentrations in serial samples of the suspended particulates can be attributed to adsorption and desorption reactions. However, the contribution from the two sources may differ because of changing hydrological conditions in the Bay and/or in the discharge canal. The relationships among the abiotic compartments in the discharge canal are summarized in Fig. 9. The interactions shown take place for all the radionuclides, but

the fractionation among the compartments differs from one radionuclide to another and from one time to another. These differences are due to the differing affinities of the radionuclides for particulates and to seasonal changes in the composition of the particulates.

OYSTERS

The availability of radionuclides to filter-feeding aquatic organisms is determined in part by the amount of particulates in the water -- living microorganisms, organic detritus, inorganic material, or any combination of these. The quantities of suspended particulates in the water column can vary both in time and in space.

The suspended material available to oysters in the discharge canal came from two sources: particulates resuspended from the material deposited onto the bottom and particulates carried in with the inflowing water. Resuspension of bottom sediments appears to be related to changing hydrological conditions in the bay and in the discharge canal. The particulate load in the inflowing water is probably dependent on biological and physical processes occurring in South Humboldt Bay.

The differences in accumulation between July and December were probably due to differences in both the

Fig. 9. Dynamics of interactions of radionuclides among the soluble and particulate phases in the discharge canal of the Humboldt Bay reactor. The soluble and particulate compartments are represented as equal in size for simplicity's sake, but in reality the distribution between them differs from one radionuclide to another. For example, the soluble fraction may contain more than 95% of the total ^{137}Cs , but only about 50% of the total ^{60}Co . The intensity of the stippling or shading approximately reflects the difference in concentration between one compartment and another. The major direction of movement from one compartment to another is indicated by the heavier arrows.

quantity and composition of the particulates in the water. Both these parameters are related to changes in production in phytoplankton and eel grass in the Bay. The differences between July and December may be related also to the levels of stable elements in the water. This factor would be important for any element whose concentration in the oysters is under metabolic regulation. For such elements, the rate of radionuclide accumulation would be expected to change with changes in the proportion of stable nuclide in the water: the accumulation per unit quantity of radionuclide released would decrease with increased concentrations of the stable nuclide in the water. For elements whose concentration in the oyster is not under homeostatic control (no regulation), but is dependent on the amount present in the water, the accumulation per unit quantity of radionuclide released would be independent of the concentration of the stable element in the water.

Little information is available on the regulation of Mn, Co, Zn, or Cs in oysters. It has been calculated that Zn is accumulated in oysters to levels much higher than those required for metabolic functions.¹⁵ Shuster and Pringle¹⁶ report that, in general, for each doubling of the environmental level of a trace metal, the tissue level of the metal approximately

doubled in *Crassostrea virginica*.

Their results obtained from a series of experiments¹⁶⁻¹⁸ suggest that in this oyster there is little or no regulation of tissue concentrations of Cd, Cu, Cr, Pb, or Zn.

It has been established for many bivalve molluscs that the presence of food material in the water stimulates their circulation of water for feeding as well as respiration purposes.¹⁹⁻²¹ Because primary productivity in Humboldt Bay is considerably greater in July than December, it is very probable that in July more of the oysters in the discharge canal would be circulating seawater for feeding. The differences in accumulation between the July and December animals are probably related to the food material present in the water.

From these experiments, the role of particulates in radionuclide accumulation appeared to differ with the radionuclide. In oysters, ⁶⁰Co appears to be accumulated primarily from the suspended particulate fraction whereas ¹³⁷Cs appears to be accumulated primarily from the soluble fraction; ⁵⁴Mn and ⁶⁵Zn accumulation are between these two extremes. Since the particulates vary in amount and composition both temporally and spatially in an environment, the quantities accumulated by organisms would vary correspondingly for those radionuclides with high affinities for particulates. The

importance of particulates in the accumulation of ^{65}Zn by oysters is confirmed by Preston.²² He found that in the River Blackwater estuary, the concentrations of ^{65}Zn were related to the silt and were poorly related to the expected distribution of ionic zinc in the seawater.

The turnover rates of radionuclides have been measured recently in bivalve molluscs both in the field²³⁻²⁶ and in the laboratory.²⁷⁻³³ In the field, ecological half-lives of ^{65}Zn have been measured^{23,24} as well as effective and biological half-lives.²⁵ Our results indicate that the turnover rates obtained in the field should vary with the season; such variation is suggested by the turnover rates observed for ^{65}Zn in oysters.²⁵

Turnover rates determined in the laboratory would be expected to vary with the kinds and amounts of particulates available for food. Some recent reports have considered the effects of food and water on turnover rates. Romeril²⁷ attributes the differences between his observed turnover rates and concentration factors and other results obtained in the field under natural conditions to be due to the virtual absence of the ingestion pathway under his experi-

mental conditions. Kenfro *et al.*,³⁴ state that for certain animals such as shrimp, the food pathway appears to play a less important role in zinc accumulation than had been generally accepted. On the other hand, Rice and Baptist³⁵ reported that *Artemia* incorporated ^{65}Zn much faster from food than water. However, more information is needed on turnover rates under varying environmental conditions in the field and under different controlled feeding regimes in the laboratory before the proper rates can be selected for biological modeling.

The stable-element Cf values for this species are considerably lower than those calculated for the radionuclides. The stable Cf values are approximately 1000 for Mn, 300 for Co, 10,000 for Zn, and 15 for Cs.³⁶ The differences between the stable and radioactive isotope concentration factors are in the order $^{60}\text{Co} > ^{54}\text{Mn} > ^{65}\text{Zn} > ^{137}\text{Cs}$. These data suggest that the radionuclides available to the oysters are higher in concentration than those calculated from the dilution of the source (expected water concentrations). The greater availability is related to the resuspension of radioactive bottom sediments during interrelease periods.

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References

1. R. E. Heft, F. L. Harrison, and W. A. Phillips, "Radioecological Study of the Humboldt Bay Marine Environment," *Proc. Am. Nuclear Society Meeting on Nuclear Methods in Environmental Research*, Columbia, Mo., Aug. 23, 1971; also Lawrence Livermore Laboratory, Preprint UCRL-73168 (1971).
2. R. E. Heft, W. A. Phillips, H. R. Ralston, and W. A. Steele, "Radionuclide Transport Studies in the Humboldt Bay Marine Environment," in *Symposium on the Interaction of Radioactive Contaminants with the Constituents of the Marine Environment* (IAEA, Vienna, 1973), pp. 595-614.
3. G. Holladay, P. L. Phelps, R. E. Heft, and F. L. Harrison, "Instrumentation for a Radioecological Study of the Humboldt Bay Marine Environment," *IEEE Trans. RS-19*, 245 (1972).
4. V. E. Noshkin, F. L. Harrison, R. E. Heft, K. M. Wong, *Environmental Radiological Measurements in Humboldt Bay*, (in preparation).
5. G. Huckabay, Lawrence Livermore Laboratory, personal communication (1975).
6. E. K. Duursma and D. Nijss, "Theoretical, Experimental and Field Studies Concerning Reaction of Radioisotopes with Sediments and Suspended Particles of the Sea. C. Application to Field Studies," *Netherlands J. Sea Res.* 6, 265-324 (1973).
7. E. K. Duursma and M. G. Gross, "Marine Sediments and Radioactivity," in *Radioactivity in the Marine Environment* (National Academy of Sciences, Washington, D.C., 1971).
8. F. L. Harrison, "Biological Implications of Nuclear Debris in Aquatic Ecosystems," *Nucl. Technol.* 11, 444-458 (1971).
9. R. L. Blanchard, W. L. Brinck, H. E. Kolde, H. L. Krieger, D. M. Montgomery S. Gold, A. Martin, B. Kahn, *Radiological Surveillance Studies at the Oyster Creek BWR Nuclear Generating Station*, U.S. Environmental Protection Agency, Rept. EPA-520/5-76-003, 1976.
10. L. W. Harding, Jr. J. H. Butler, and R. E. Heft, *The Standing Stock and Production of Eelgrass (Zostera marina L.) in Humboldt Bay, California*, Lawrence Livermore Laboratory, Preprint UCRL-76987 (1975).
11. K. M. Wong, V. E. Noshkin, and G. Holladay, Lawrence Livermore Laboratory personal communication (1975).
12. D. P. Serpa, Pacific Gas and Electric Company, San Ramon, California personal communication (1975).

13. G. F. Lee, "Factors Affecting the Transfer of Materials between Water and Sediments," in *Literature Review No. 1, Eutrophication Information Program Water Resources Center, University of Wisconsin, Madison* (1970), pp. 1-35.
14. E. K. Duursma and C. J. Bosch, "Theoretical, Experimental and Field Studies Concerning Diffusion of Radioisotopes in Sediments and Suspended Particles of the Sea," *Netherlands J. Sea Res.* 4, 395-469 (1970).
15. J. E. Pequegnat, S. W. Fowler, and L. F. Small, "Estimates of the Zinc Requirements of Marine Organisms," *J. Fish. Res. Bd. Canada*, 26, 145-150 (1969).
16. C. N. Shuster and B. H. Pringle, "Trace Metal Accumulation by the American Eastern Oyster *Crassostrea virginica*," *Natl. Shellfisheries Assoc.* 59, 91-103 (1969).
17. B. H. Pringle, D. E. Hissong, E. L. Katz, and S. T. Mulawka, "Trace Metal Accumulation by Estuarine Mollusks," *J. Sanit. Eng. Div. (Proc. Am. Soc. Civil Eng.)* 94, 455-475 (1968).
18. C. N. Shuster, Jr., and B. H. Pringle, "Effects of Trace Metals on Estuarine Mollusks," *Proc. 1st Conf. on Mid-Atlantic Waste*, University of Delaware (CE-5) (1968), pp. 285-304.
19. P. S. Galtsoff, "The American Oyster *Crassostrea virginica* Gmelin," *U.S. Fish. Wildl. Serv. Fish. Bull.* 64, 1-480 (1964).
20. T. R. Rice and R. J. Smith, "Filtering Rates of the Hard Clam (*Venus mercenaria*) Determined with Radioactive Phytoplankton," *U.S. Fish. Wildl. Serv. Fish. Bull.* 58, 73-82 (1958).
21. C. Davids, "Influence of Suspensions of Microorganisms of Different Concentrations on the Pumping and Retention of Food by the Mussel (*Mytilus edulis* L.)," *Netherlands J. Sea Res.* 2, 233-249 (1964).
22. A. Preston, "The Control of Radioactive Pollution in a North Sea Oyster Fishery," *Helgol. wiss. Meeresunter.* 17, 269-279 (1968).
23. E. O. Salo and W. L. Lest, "The Concentration of ⁶⁵Zinc by Oysters Maintained in the Discharge Canal of Nuclear Power Plants," *Proc. 2nd Natl. Symp. on Radioecology*, D. J. Nelson and F. C. Evans, Eds. (National Technical Information Service, Springfield, Va., 1967), pp. 363-371.
24. D. A. Wolfe, "Levels of Stable Zn and ⁶⁵Zn in *Crassostrea virginica* from North Carolina," *J. Fish. Res. Bd. Canada* 27, 47-57 (1970).
25. A. H. Seymour and V. A. Nelson, "Biological Half-lives for Zinc and Mercury in the Pacific Oyster, *Crassostrea gigas*," in *Proc. 3rd Natl. Symp. on*

Radioecology (National Technical Information Services, Springfield, Va., 1972), pp. 849-856.

26. A. H. Seymour and V. A. Nelson, "Decline of ^{65}Zn in Marine Mussels Following the Shutdown of Hanford Reactors," in *Symp. on the Interaction of Radioactive Contaminants with the Constituents of the Marine Environment*, (IAEA, Vienna, 1973), pp. 277-286.
27. M. G. Romeril, "The Uptake and Distribution of ^{65}Zn in Oysters," *Mar. Biol.* 9, 347-354 (1971).
28. C. L. Schelske, "Fallout ^{54}Mn Accumulated by Bay Scallops *Argopecten irradians* (Lamarck) near Beaufort, North Carolina," in *Symp. on Interaction of Radioactive Contaminants with the Constituents of the Marine Environment* (IAEA, Vienna, 1972), pp. 331-346.
29. R. J. Pentreath, "The Accumulation from Water of $^{65}\text{Zinc}$, $^{54}\text{Manganese}$, $^{58}\text{Cobalt}$, and $^{59}\text{Iron}$ by the Mussel, *Mytilus edulis*," *J. Mar. Biol. Assoc. U.K.* 53, 127-143 (1973).
30. R. J. Pentreath, "The Role of Food and Water in the Accumulation of Radionuclides by Marine Teleost and Elasmobranch Fish," in *Symp. on the Interaction of Contaminants with the Constituents of the Marine Environment* (IAEA, Vienna, 1973), pp. 421-436.
31. A. W. Van Weers, "Uptake and Loss of $^{65}\text{Zinc}$ and $^{60}\text{Cobalt}$ by the Mussel *Mytilus edulis*," in *Symp. on the Interaction of Radioactive Contaminants with the Constituents of the Marine Environment* (IAEA, Vienna, 1973), pp. 385-402.
32. F. L. Harrison, "Accumulation and Loss of Cobalt and Cesium by the Marine Clam, *Mya arenaria*, under Laboratory and Field Conditions," in *Symp. on the Interaction of Radioactive Contaminants with the Constituents of the Marine Environment* (IAEA, Vienna, 1973), pp. 453-478.
33. G. Cranmore and F. L. Harrison, Loss of $^{137}\text{Cesium}$ and $^{60}\text{Cobalt}$ from the Oyster *Crassostrea gigas*," *Health phys.* 28, 319-333 (1975).
34. W. C. Renfro, W. S. Fowler, M. Heyraud, and J. LaRosa, "Relative Importance of Food and Water Pathways in the Bio-accumulation of Zinc," in *Activities of the International Laboratory of Marine Radioactivity, 1974 Report* (IAEA, Vienna, 1974), pp. 11-20.
35. T. R. Rice and J. P. Baptist, "Ecological Effects of Radioactive Emissions from Nuclear Power Plants, in *Human and Ecological Effects of Nuclear Power Plants*, L.A. Sagon, ed. (Thomas, Springfield, Ill., 1974), pp. 373-439.

36. F. L. Harrison and R. E. Heft, Lawrence Livermore Laboratory, unpublished data (1977).