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The Environmental Behavior of Transuranic Nuclides Released from Water Cooled Nuclear Power Plants

Prepared by V. T. Bowen

Woods Hole Oceanographic Institution

Prepared for
U.S. Nuclear Regulatory
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Prepared for
Division of Safeguards, Fuel Cycle and Environmental Research
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
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ABSTRACT

Release data are reported for three coastal water-cooled nuclear reactors: Millstone Point No. 1 and No. 2 (for the period January 1977 through April 1978), and Maine Yankee (for the period 20 June 1977 through 25 March 1978); release samples were analyzed for ^{55}Fe, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, ^{242}Cm and ^{244}Cm, but not all nuclides on every sample. Radioiron is a major component of the releases measured; the transuranium nuclides are less significant components than was expected, but levels have occasionally reached microcuries per month. Pulses of this size are adequate for tracer studies.

Environmental samples (water, sediments, and biota) have been analyzed from about the two reactor sites noted, and that of the Pilgrim No. 1 reactor. No water samples remote from reactor outflows have unequivocally shown reactor contamination. No sediment samples from near Millstone Point Point or Pilgrim 1 have shown reactor contamination; this has been clearly evident in several sediment collections from near Maine Yankee. Biota so far measured from near Millstone Point show reactor contamination only when taken from the effluent canal. From the Maine Yankee and Plymouth areas, however, biota samples frequently prove to show slight, but definite, reactor contamination. In these two areas biogeochemical studies of the fates of long-lived waste radionuclides could easily be carried out, and would be very profitable.

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Acknowledgements

We could not have proceeded so far in the present project without the support and cooperation of Mr. Philip Stohr, of NRC, whose help it is a pleasure to acknowledge. The work at Millstone Point was facilitated by assistance from Dr. William Renfro and Dr. Ralph Briscoe; that at Maine Yankee was comparably helped by Dr. David Sturniolo, Jr. of Maine Yankee and Professor C. T. Hess of Univ. of Maine. The collecting by, and analysis of samples at, WHOI were carried out by a number of colleagues, among whom special mention should be made of H. D. Livingston, J. C. Burke, D. R. Mann, A. G. Gordon, L. B. Graham and W. R. Clarke. All of this help is gratefully acknowledged.

Substantial amounts of data, reported here as essential to the understanding of this program, were obtained as parts of other projects, most prominently that funded by the Dept. of Energy under contract DE-AC02-76-EV-03563.A005 and by the Environmental Protection Agency as part of the Mussel Watch Program under contract with the University of California; we are grateful for this support and for the freedom with which these data can be used.

THE ENVIRONMENTAL BEHAVIOR OF TRANSURANIC NUCLIDES LEAKED FROM WATER-COOLED NUCLEAR POWER PLANTS

INTRODUCTION

A serious problem that faces anyone trying to predict, or to model, the behavior of transuranic radionuclides released to aquatic environments, whether in the course of planned disposals or as the result of accidents, is our present ignorance both of the geochemistry of these elements and of the extent to which this is controlled by either the chemistry of the release materials or special attributes of the local environment. A very promising approach to this problem lies in the use of each presently identifiable transuranic element release as an environmental experiment and by the comparing the differences and similarities among the behavior of the various radioelements represented, establishing those generalizations that seem to hold widely, as well as those conditions under which each generalization breaks down. It is also valuable to ascertain what similarities in geochemical behavior may appear between the transuranic elements and those better known elements whose radioisotopes are also components of each released mixture.

Pursuit of this approach has led us to study a substantial number of releases of artificial radionuclides, ranging from worldwide and close-in fallout from nuclear weapons tests, the liquid effluent releases from fuel reprocessing plants in Great Britain, France, and the USA, leakage from solid waste containers dumped at sea, to the discharges from water-cooled nuclear reactors used for electrical power production. In this last project, we have been supported partly by the U. S. Nuclear Regulatory Commission, and it is this work we are reporting here.

The cooling water stream from all nuclear power reactors contains small amounts of artificial radioactivity arising in two very different ways:

firstly, by direct neutron reactions on the components of the cooling water stream, or of the duct work through which it passes.

secondly, collected into storage tanks, as the result of a variety of technical operations, of the plant or of its laboratories, and released periodically, usually after considerable chemical cleanup, by pumping into the cool-

ing water stream.

Although the first of these processes certainly results in some transuranic radionuclides, these are produced in very small amounts, determined largely by the uranium content of the cooling water stream, and probably largely restricted to neptunium 239 and its daughter plutonium 239. There is little question that the overwhelming preponderance of plutonium, and virtually all of such heavier transuranic elements as americium and curium, result from the second class of operations. Because of this origin, in processes that are only secondary functions of the day-to-day power producing operations of the plants, the rates of release fluctuate widely in ways that, if they could be predicted, or even ascertained at the time of release, would be of considerable value in helping to establish time constants, especially of some of the biological, or sedimentological, interactions of the nuclides released.

In this report we propose to discuss the patterns of nuclide release that characterized three reactors, Millstone Point Nos. 1 and 2 and Maine Yankee, during parts of the years 1977 and 1978. We will also discuss some evidence concerning the fate of the released nuclides in their aquatic environments, including data relevant to the Pilgrim 1 reactor, even though we have no information on its release patterns.

MATERIALS

Through the good offices of Mr. Philip Stohr, NRC, we received from the operators of the two reactors at Millstone Point, Connecticut (Millstone Point No. 1 and No. 2) and of the Maine Yankee reactor at Wiscasset, Maine, samples that represented their periodic storage tank discharges for parts of the years 1977 and 1978. These were replicates of the discharge samples that the operators collect routinely for assay of major radionuclides.

In the case of the Millstone Point reactors, the samples were adjusted to represent the monthly discharge experience. The series we received began with January 1977, continued through the year (without samples for August or September), and through the first four months of 1978.

In the case of Maine Yankee, we received a separate sample to represent each discharge event, the volume representation being adjusted for those of the various tanks discharged. Discharges occurred as seldom as once or as often as eleven times a month. The period represented by samples we have so far analyzed extended from 20 June 1977 through 25 March 1978.

As discussed below, our initial expectation, from Millstone Point No. 1 data, was that the activity concentrations released would be rather large (by our environmental criteria). For that reason, we began by asking for only small volumes of sample -- 25 ml per period. Between the February and March 1977 discharges from Millstone Point No. 1, however, an additional stage of waste discharge treatment was introduced. As shown in Table 1, this resulted in a very substantial reduction, close to two orders of magnitude, in the concentrations of transuranic nuclides in the discharge, bringing them too close to our detection limits. Beginning with May 1977, then, the sample size we received was increased to one liter per discharge. This increase applied to both Millstone No. 1 and No. 2, since the No. 2 plant had proved to be significantly cleaner in its discharge than we had expected.

It would obviously be of considerable value to have been able to supplement these samples representing the tanks discharged,with a series of samples of the actual effluent stream. This would both have given NRC confirmation that the storage tanks were being correctly sampled and did represent the major source of the nuclides of concern and have provided an estimate of the nuclide concentrations that enter the environment, for clarification of the biological and sedimentological relationships being assessed. Unfortunately, we were quite unable to arrange proportional sampling of the cooling water effluent stream, so that this part of the investigation still cries out to be undertaken.

In addition to the discharge samples provided by the reactor operators, we obtained series of samples of organisms, of sediments, and of water, in the environments of these reactors, and of the reactor operated by Boston Edison Co., at Plymouth, Massachusetts (Pilgrim No. 1). Most of these samples we collected ourselves but some were provided by the environmental survey teams of the reactor operators. In the Wiscasset area we were substantially assisted, through the good offices of C. T. Hess, University of Maine, Orono, by the people and facilities of the Darling Center, of that university. A listing of these environmental samples,together with their status in analysis, was included in the Progress Report submitted October 1978 under the subject contract. As discussed there, some additional samples have been obtained; additional analyses have been performed under an extension of the subject contract, and substantial numbers, both of samples and analyses, that are highly relevant to the interpretation of the NRC-supported work, have been completed with support from other agencies.

METHODS

Our radioanalytical methods have been described in a number of publications. The following summary list of references may be useful:

^{55}Fe: Labeyrie, Livingston and Gordon, 1975. Nucl. Instr. and Meth. 128, 575-580.

^{90}Sr, ^{137}Cs and others: Wong, Noshkin and Bowen, 1970. In Reference Methods for Marine Radioactivity Studies (IAEA, Vienna), pages 119-127.

Pu, Am, Cm: Livingston, Mann and Bowen, 1975. In Analytical Methods in Oceanography (ACS, New York), pages 124-138.

We engage regularly in analytical intercomparisons organized by the International Atomic Energy Agency, the U. S. National Bureau of Standards, or the Department of Energy. Published reports of our performance in some of these may be listed as follows:

1. Fukai, Ballestra and Murray, 1973. In Radioactive Contamination of the Marine Environment (IAEA, Vienna), pages 3-27.
2. Noyce, Hutchinson, Mann and Mullen, 1976. In Proceedings of International Conference on Environmental Sensing and Assessment (Inst. Electr. Electron. Eng., N.Y.) paper 19-5.
3. Volchok and Feiner, 1979. A radioanalytical Laboratory Intercomparison Exercise, U. S. Dept. of Energy, Rept. EML-366, 43 pp.

Other performance information is either summarized or referenced in the methods papers cited above, or in the various articles reporting and analyzing our data, that are cited in the Discussion below.

RESULTS

Discharge Data:

The analytical results on the discharge samples are set out in three tables: Table 1 dealing with Millstone Point No. 1; Table 2 dealing with Millstone Point No. 2; Table 3 dealing with Maine Yankee. Data are presented for ^{55}Fe, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, ^{242}Cm, and ^{244}Cm; not all nuclides were, however, measured in each sample. For the shorter-lived nuclides, data are shown as of the date of collection of the sample. In each case, the analytical result has been multiplied by the total volume of the discharge represented to yield an estimate of the total activity of that nuclide discharged in the period represented.

Environmental Data:

For convenience, the data referring to the environmental samples, water, sediments, and biota, as well as the Figures showing respective collection locations, are inserted in the Discussion sections concerning the various reactor environments. Since most of the radionuclides that we have measured are not unique to reactor operations but are present worldwide as the result of atmospheric testing of nuclear explosions, we have included in the biota tables, comparison data referring to relatively nearby samples that we believe have experienced only fallout contamination; these data derive principally from the Mussel-Watch Program (Goldberg et al, 1978, and Bowen et al, to be published) supported by EPA.

In the order of their insertion in the text, these figures and tables are as follows:

Figure 1: Millstone Point Collecting Stations.
Table 5: Millstone Point, Conn.: Long Island Sound Surface Water.
Table 6: Millstone Point, Conn.: Sediment Cores Radiochemistry.
Table 7: Millstone Point Environmental Samples - Biota.
Figure 2: Maine Yankee Collecting Stations.
Table 8: Wiscasset, Maine: Surface Water from Montsweag Bay.

TABLE 1

MILLSTONE POINT REACTOR #1 MONTHLY DISCHARGE ACTIVITY

Month and Year	Stored Volume (liters)	^{55}Fe mCi (DOC) [1]	^{60}Co mCi	^{134}Cs mCi (DOC) [1]	^{137}Cs mCi	^{238}Pu nCi	$^{239,240}Pu$ nCi	^{241}Am nCi	^{242}Cm nCi (DOC) [1]	^{244}Cm nCi
Jan. 1977	1.07×10^6		5.56±0.12	4.06±0.11	8.26±0.14	690±150	140±10	<210	3,660±360	<210
Feb.	282,118			0.30±0.05	0.70±0.03	430±70	280±50	260±50	3,190±170	520±60
March	543,418			12.1±0.3	28.5±0.2	insufficient sample				
April	216,141			0.041±0.002	0.11±0.01	insufficient sample				
May	108,638			68.0±0.1	106.0±0.1	14±2	9±1	10±2		
June	8,630					1.2±0.2	0.9±0.2	1.0±0.2		
July	9,671	4.94±0.17	14.0±0.2	21.5±0.2	50.4±0.3	11.3±0.5	7.4±0.4	8.0±0.4	49.6±5.3	9.3±0.8
August	No sample									
Sept.	No sample									
Oct.	9,463	3.05±0.06	1.50±0.02	1.17±0.02	3.00±0.02	10.2±0.5	6.1±0.3	6.2±0.1	30.9±5.7	7.6±1.1
Nov.	11,000		0.71±0.01	0.46±0.01	1.30±0.01	6.3±0.3	4.5±0.3			
Dec.	429,000		0.46±0.03	0.14±0.03	0.32±0.03	29±4	21±4			
Jan. 1978	73,662		1.90±0.04	0.86±0.04	1.56±0.01	234±8	75±4			
Feb.	18,813	30.6±0.1	61.2±0.2	0.42±0.07	1.66±0.09	40±1	28±1	25.0±1.9	97.6±5.6	26.7±1.9
March	611,447		0.75±0.06	0.35±0.10	0.83±0.07	73.7±18.4	36.8±6.1			
April	231,111		21.5±0.1	0.35±0.04	0.97±0.04	8,400±200	3,900±100			

[1] DOC = data decay corrected to date of collection of sample

TABLE 2

MILLSTONE POINT, REACTOR #2 MONTHLY DISCHARGE ACTIVITY

Month and Year	Volume Stored (liters)	^{55}Fe mCi (DOC) [1]	^{60}Co mCi	^{134}Cs mCi (DOC) [1]	^{137}Cs mCi	^{238}Pu nCi	$^{238,239}Pu$ nCi	^{241}Am nCi	^{242}Cm nCi (DOC) [1]	^{244}Cm nCi
Jan. 1977	640,422			0.30±0.05	0.61±0.09	<40	60±80	<130	<65	
Feb. 1977	333,943			0.22±0.03	0.57±0.03	40±30	110±60	20±27	40±20	<35
March 1977	333,693			0.25±0.04	0.61±0.03	Insufficient sample				
April 1977	793,820			1.41±0.13	3.52±0.06	<50	<80			
May 1977	547,735			0.85±0.11	2.36±0.05	<1	<2			
June 1977	701,376					<1	<1			
July 1977	402,769	0.40±0.01	0.11±0.02	0.03±0.02	0.053±0.035	2.4±0.8	0.4±0.4	1.6±1.2		
Aug. 1977	323,000			0.09±0.03	0.26±0.03	<0.7	<0.7	<9	<9	<9
Sept. 1977	629,000			0.25±0.02	0.52±0.05	2±2	4±3			
Oct. 1977	961,000	0.93±0.01	0.69±0.11	0.27±0.09	0.61±0.01	4±2	6±4	<8		
Nov. 1977	907,000			9.9±0.4	11.4±0.3	1±1	2±1			
Dec. 1977	536,020			7.6±0.2	13.6±0.2	<0	0.2±0.1			
Jan. 1978	485,192			3.48±0.14	6.58±0.15	<0.5	<0.5			
Feb. 1978	227,448	1.36±0.01	5.07±0.05	0.23±0.04	0.62±0.03	0.2±0.1	1.6±0.7	<2	<2	<2
March 1978	640,028		2.84±0.10	0.87±0.08	1.17±0.09	0.3±1.1	<0.9			
April 1978	730,217		1.00±0.03	0.57±0.03	1.07±0.02	<1.5	<1.5			

[1] DOC = data decay corrected to date of collection of sample

TABLE 3

MAINE YANKEE, PERIODIC DISCHARGE ACTIVITY

Date - 1977	Time	Volume (liters)	^{55}Fe mCi (DOC)[1]	^{134}Cs mCi (DOC)[1]	^{137}Cs mCi	^{238}Pu nCi	$^{239,240}Pu$ nCi	^{241}Am nCi
20 June	1300	46,934	0.033±0.001	0.007±0.003	0.014±0.004	0.85 ±0.24	0.47±0.19	2.5±1.4
19 July	1000	45,526						
	1400	40,300						
16 August	1200	35,200		0.108±0.008	0.51 ±0.01	0.11 ±0.14	3.7 ±0.5	
	1752	38,016						
25 August	1100	46,934						
26 August	1100	46,366		0.022±0.002	0.078±0.002	0.47 ±0.23	4.7 ±0.6	
27 August	1100	46,366						
30 August	1500	46,934		0.021±0.004	0.089±0.005	<0.09	0.19±0.19	
2 October	1400	40,363	0.056±0.001	3.87 ±0.01	14.4 ±0.1	<0.02	<0.02	0.6±0.8
6 October	1500	6,101						
10 October	1200	14,080		1.28 ±0.01	4.73±0.01	0.071±0.057	0.49±0.11	
24 October	1500	36,139						
8 November	1900	40,363		0.48 ±0.01	1.83 ±0.01	0.08 ±0.12	0.45±0.28	
24 November	1400	38,486						
2 December	1230	26,745		0.22 ±0.01	0.90 ±0.01	<0.05	0.05±0.10	
8 December	1300	46,934						
	1700	46,934						
12 December	1600	45,514	1.02 ±0.01	0.23±0.01	0.91 ±0.01	2.1 ±1.0	1.5 ±0.8	
23 December	1500	39,883						

...continued...

TABLE 3, Continued

MAINE YANKEE, PERIODIC DISCHARGE ACTIVITY

Date - 1978	Time	Volume (liters)	^{55}Fe mCi (DOC) [1]	^{134}Cs mCi (DOC) [1]	^{137}Cs mCi	^{238}Pu nCi	$^{238,239}Pu$ nCi	^{241}Am nCi
3 January	1500	42,229		0.099±0.004	0.39 ±0.01	0.08±0.13	0.15±0.17	
10 January	1700	36,130						
11 January	1800	16,892		0.21 ±0.01	0.85±0.01	<0.003	0.24±0.13	
12 January	1300	46,922						
	1500	24,399						
13 January	2000	46,922						
14 January	2400	42,229						
15 January	1400	42,229						
	2300	42,229		0.003±0.001	0.010±0.002	<0.08	<0.08	0.29±0.04
17 January	1700	42,229						
18 January	1400	46,922						
19 January	1500	45,983						
18 January	2000	42,229						
3 February	1800	39,883						
14 February	1400	42,105						
	2300	46,922						
16 February	1600	44,106		0.004±0.003	0.013±0.004	<0.08	<0.08	-0.02±0.04
	2300	46,922						
20 February	1600	38,017						
23 February	1700	43,164						
25 February	0800	33,792						
	2300	42,241						

...continued...

TABLE 3, Continued

MAINE YANKEE, PERIODIC DISCHARGE ACTIVITY

Date - 1978	Time	Volume (liters)	^{55}Fe mCi (DC) [1]	^{134}Cs mCi (DOC) [1]	^{137}Cs mCi	^{238}Pu nCi	$^{238,239}Pu$ nCi	^{241}Am nCi
27 February	1500	41,771						
28 February	2000	42,241						
1 March	2300	42,241		0.005±0.003	0.009±0.003	<0.08	0.21±0.13	-0.3±0.2

[1] DOC = data decay corrected to date of collection of sample

DISCUSSION

NUCLIDES STUDIED:

It may be well to preface this discussion with a few comments on the nuclides measured:

^{55}Fe: At the time we prepared this proposal, it was the prevailing wisdom at NRC that "reactors produce ^{59}Fe but not ^{55}Fe", and we were accordingly directed to omit ^{55}Fe from our program. The briefest examination of any chart of nuclides, however (as well as the most superficial background in radionuclide technology), convinces us that it is impossible by neutron reactions on natural iron to produce ^{59}Fe without simultaneously producing ^{55}Fe; the target nucleus for the latter reaction is of 5.8% abundance, compared to 0.31% for the former, and the thermal neutron cross section for the latter reaction is about twice that for the former. Accordingly, since our interest is in the environmental behavior of nuclides that are long-lived enough to persist through years of biogeochemical cycling, and to reveal local build-up in concentrations whenever they occur, we looked at ^{55}Fe in the reactor output samples (these analyses were supported separately by our Dept. of Energy contracts, and we throw them into this report for free). It should be noted that substantial amounts of ^{55}Fe are being released from each of these reactors, the ^{55}Fe to ^{60}Co ratio ranging from 0.27 to 3.64 for the six samples reported, with a mean of 1.36, or of 0.51 on an activity-adjusted basis. Clearly ^{55}Fe is a significant product of these reactors, and one that offers valuable tracer data for anyone studying the long-term environmental impact of the reactor waste streams. Some relevant biogeochemical information appears below in connection with discussions of the Maine Yankee and Pilgrim 1 reactor environments.

^{134}Cs:^{137}Cs: Both ^{134}Cs and ^{137}Cs are produced, and released, in considerable amounts in reactor operations. The very different half-lives of the two nuclides ($T_{½}$ ^{134}Cs = 2.1y; $T_{½}$ ^{137}Cs = 30.1y) provide a very useful guide to the time characteristics of tracer Cs in environ-

ments, so long as the release ratio is reasonably uniform. From the data in Table 1, the mean ratio, month-by-month, from Millstone No. 1 was 0.43, and weighted by the activities released 0.54; from Table 2, the ratios from Millstone No. 2 were, respectively, 0.50 and 0.60. Since Millstone No. 1 during this interval, put out about four times as much total radio-Cs as did Millstone No. 2, and the ratios, ^{134}Cs to ^{137}Cs, are so close, it would be possible to use the change in this ratio, in reasonably well-isolated samples, as a dating tool.

Much the same conclusion is supported, for Maine Yankee, by the data in Table 3, although the amounts of radio-Cs discharged are much lower, so that its use as a tracer may be expected to be somewhat more difficult.

^{238}Pu:$^{239,240}Pu$: The amounts of Pu that have been delivered to the higher latitudes of the northern hemisphere have been large compared to those released from any power reactors we know of: about 2 mCi Pu per km^2 from global fallout, versus the nannocuries per month released from the Millstone or Maine Yankee reactors. Tracing of the reactor effluent Pu in this situation is aided by the very substantial difference in its characteristic ratio of ^{238}Pu to $^{239,240}Pu$: ranging from about 1 to as much as 5, compared to the fallout mean of about 0.035, which rose only to about 0.10 even at the height of the SNAP 9A contribution in the northern hemisphere. Even quite small increments of reactor-released Pu can thus be identified in environmental samples by the effect they have on the ^{238}Pu:$^{239,240}Pu$ ratio.

^{241}Am: This nuclide in fallout has originated almost entirely from decay of its parent ^{241}Pu. At the ratios so far observed of ^{241}Pu to $^{239,240}Pu$ in test debris, the ingrowth of ^{241}Am is not predicted to produce ratios above 0.30 ^{241}Am to $^{239,240}Pu$ until about 1980. Of course in geochemical settings where Am is separated from Pu, this ratio changes, and we have seen it as high as 1 in sediments from special situations. In general, however, reactor waste ^{241}Am can be at least partly differentiated from fallout ^{241}Am by its ratio to $^{239,240}Pu$.

^{242}Cm, ^{244}Cm: The curium nuclides were not measurable components of most nuclear test debris. A major reason for our undertaking this project was the hope, based on measurement of Cm in shellfish contaminated by the Millstone No. 1 waste stream, that it would offer us new, and generalisable, situations in which the biogeochemistry of Cm could be studied. Unfortunately, as shown in Tables 1 and 2, the new waste treatment process at Millstone wiped out this hope. In the Maine Yankee effluent we have never been able to see any Cm. These data should be reassuring to NRC, even though they have been a serious disappointment to us.

RELEASE PATTERNS

Somewhat unexpectedly to us, there appears to be no correlation between the release patterns of the various radioelements. As noted above, where two nuclides of the same element are being released (as in the cases of Cs or Pu) there is reasonable consistency in the release ratios. But Cs release rates are wholly unreliable predictions of Pu or Cm releases on the one hand, or of ^{60}Co or ^{55}Fe on the other. This conclusion is supported by Tables 1, 2 and 3. In the cases of Cs and Pu it is illustrated even more graphically in Table 4, where the monthly release figures from Millstone Nos. 1 and 2 have been converted into daily discharges. This tabulation shows the extent to which discharges from each reactor were uncoupled in respect to ^{137}Cs vs Pu, but also that, in general, the two reactors were not coupled to each other; the exception to this last statement is the reduction of Pu discharges after February 1977 which appears to have affected both plants comparably. But, in contrast, the steadily increasing Pu release rate from Millstone No. 1 from Nov. 1977 to April 1978 was not reflected at all by Millstone No. 2.

ENVIRONMENTAL SAMPLES

At least partly supported by NRC under the subject contract, we have performed radiochemical analyses on environmental samples in the areas about three coastal-sited nuclear power stations: Millstone Point Reactors 1 and 2, Maine Yankee Reactor, and Pilgrim No. 1 Reactor. Only from the

TABLE 4

MILLSTONE POINT DAILY DISCHARGE ACTIVITY

Month and Year	Reactor #1		Reactor #2	
	^{137}Cs μCi/day	$^{239,240}Pu$ pCi/day	^{137}Cs μCi/day	$^{239,240}Pu$ pCi/day
Jan. 1977	275	4,667±2,000	20.3	2,000±2,700
Feb. 1977	25.0	10,370±1,700	20.4	3,667±2,000
March 1977	950	-- [1]	19.7	-- [1]
Apr. 1977	3.7	-- [1]	117	<2,700
May 1977	3,533	300	78.7	< 67
June 1977	-- [2]	30.0	-- [1]	< 33
July 1977	1,680	287	1.8	13.3
Aug. 1977	-- [3]	-- [3]	8.7	< 23
Sept. 1977	-- [3]	-- [3]	17.3	133±100
Oct. 1977	100	203	20.3	200±133
Nov. 1977	43.3	150	380	67±33
Dec. 1977	10.7	700	453	6.7±3.8
Jan. 1978	52.0	2,500	219	< 17
Feb. 1978	55.3	1,037	23.0	59
March 1978	27.7	1,227	39.0	< 30
Apr. 1978	32.3	130,000	35.7	< 50

[1] Sample too small for analysis at these concentrations.

[2] No data.

[3] No Sample supplied.

first two of these reactor operations (Millstone Point No. 1 and 2, and Maine Yankee) do we have relevant data for the radionuclide discharges, as set out above in Tables 1, 2, and 3.

Millstone Point, Conn. Reactors No. 1 and 2:

In late October, 1977, we obtained a small series of water samples representing the reactor coolant outflow stream (identified by its temperature anomaly), the source term in Niantic Bay for the coolant intake (Station 2), and two Long Island Sound points that might have been expected to receive reactor outflow water after some circulation and dilution. In two parts of the outflow plume, the inner (hottest) plume, and the outer (mixed) plume envelope, large volumes of water were passed through 1 μm filter cartridges and the particles so collected were analyzed separately. Data from these samples are set out in Table 5 and can be compared to the October 1977 data shown for discharge in Tables 1 and 2.

Although the October 1977 discharges were characterized by ratios as follows:

^{55}Fe to ^{137}Cs	about	1
^{134}Cs to ^{137}Cs	"	0.39
^{238}Pu to $^{239,240}Pu$	"	1.7
^{244}Cm to ^{241}Am	"	1.2,

there is little evidence of this in the outflow data. Comparing water sample 5A to water sample 2, we see that the outflow has been enriched by 48% in ^{137}Cs, very slightly depleted in ^{90}Sr, shows no evidence of enrichment by ^{134}Cs, ^{238}Pu or $^{239,240}Pu$, and has been enriched by 290% in ^{55}Fe. A larger volume sample would have been needed for measurement of ^{134}Cs at the concentration indicated. It is of interest that the ^{55}Fe was essentially 100% associated with particles collected by the 1 μm filters; this has been reported for other corrosion products in reactor effluents but we had expected that a substantial fraction of the radio-Fe, arising from "plant operations", would still be in true solution. Only about 33% of the Pu in the outflow stream was particle-associated; this contrasts with the rapid particle uptake reported for Windscale-effluent Pu (Hetherington, 1976; Nelson and Lovett, 1979). What is most evident, however, is that the pulsing of the discharge pattern must be sufficiently sporadic so that water sampling

in the outflow should be guided by information that was not available to us. It is quite possible that the two Long Island Sound samples (#3 and #4) both show some ^{55}Fe from Millstone and that No. 3 shows also some ^{137}Cs, but the signals are very small, so that a large number of comparison samples (or large enough volume samples to give good counting statistics for ^{134}Cs or ^{238}Pu) would be needed to support this sort of conclusion. It would be more economical to be able to undertake detailed programs of sampling water at short intervals after known releases, preferably those expected to represent substantial pulses of the nuclides to be measured.

In Table 6 we have summarized the data for two sediment cores, collected with our 21-cm diameter, tripod-mounted, gravity corer at the same time the water samples (above) were collected. Core No. 3 was collected at about the center of Niantic Bay, and Core No. 2 offshore in Long Island Sound, at the same location as water sample No. 3 (Table 5). In comparing these two cores, attention should be given not only to their locations but also to the substantial difference in the grain size distribution of the sediments: the Niantic Bay core is much coarser grained, only 30% less than 62 μ, compared to over 66% less than 62 μ for core No. 2. The radionuclide concentrations of the sediments also differ substantially. The Niantic Bay core is much the richer in ^{55}Fe, slightly poorer in ^{137}Cs, and much poorer in $^{239,240}Pu$. Neither core showed, even in the top section, any measurable ^{134}Cs, nor any strong elevation of the ^{238}Pu to $^{239,240}Pu$ ratio, as would result from accumulation of such reactor effluents as are summarized in Tables 1 and 2. Core No. 2 showed no measurable Cm radioactivity, either. On the other hand, the inventories of radionuclides in core No. 2 -- core No. 3 was too short to permit usable inventory estimates to be made -- are unusually high. Livingston and Bowen (1979) reported ^{137}Cs and Pu analyses of a considerable series of shallow water sediment cores. Of the 12 comparable cores summarized in their report, none even approached the ^{137}Cs inventory of core No. 2, over 45.5 mCi/km^2 (note that nuclide concentrations of core #2 are still large even in the 29-33 cm bottom section, indicating these inventory estimates as minima.); the range reported by Livingston and

Bowen (1979) was 4 to 15 mCi per km^2 of fallout ^{137}Cs. The inventory of Pu in core No. 2, over 12.1 mCi/km^2, appears at least as much elevated as that of ^{137}Cs, whereas the ^{55}Fe inventory is not too far removed from that one would expect in relatively oxidizing fine-grained sediments.

Without both the carefully examined data (Table 6) for ^{134}Cs and for ^{238}Pu, and a set of comparison cores like those reported by Livingston and Bowen (1979), it would be very difficult to avoid concluding that the high ^{55}Fe of core No. 3, and the high ^{137}Cs and Pu of core No. 2 were indications of the accumulation, in sediments nearby, of long-lived radionuclides released in the Millstone Point reactor effluent stream. Examination of this whole data base, however, forces us to conclude that each of these Millstone Point cores represents a basin of sediment accumulation from a large neighboring area (as we feel Krishnaswami et al (1973) showed for the Santa Barbara Basin). Only nuclides of world-wide fallout origin are observable, and what is most interesting is the selection of ^{55}Fe by the coarse and of Pu by the fine grained sediments. This is not, of course, to say that the long-lived waste radionuclides from Millstone Point effluents do not accumulate somewhere in sedimentary deposits; we just have not so far found the place.

The Millstone Point biota data collected in Table 7 we take as confirming this point. The oyster sample, taken in 1975 from the effluent stream in the quarry, was the source of our original interest in this project; these organisms showed substantial and unequivocal evidence of reactor effluent in their concentrations of ^{137}Cs, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and Cm; oysters collected at the same time at Black Point (see Fig. 1) may have been slightly above fallout levels in respect to ^{137}Cs and $^{239,240}Pu$, but were certainly not unequivocally so. The same statement applies to the mussel sample from area 2 (Fig. 1) collected in 1977: its soft parts were above the range, for that year, of ^{137}Cs concentrations but its Pu and Am concentrations were comfortably within the range. The fallout contaminated mussel data shown at the bottom of Table 7 confirm an important point: there are localities along the U. S. coast line that have consistently yielded mussels with ^{137}Cs or Pu concentrations at or near the top of the ranges that apply, but that we have no reason to believe

have experienced any non-fallout source of artificial radionuclides. Furthermore, there is not a strong covariance of ^{137}Cs with Pu in these correlations (Bowen et al to be published); New Haven, Conn., is a good example of this, consistently on the high end of Pu concentrations but not comparably high in ^{137}Cs. Without data of this sort it might have been very difficult to explain away the Millstone Point mussel samples collected in 1977 from either area 2 or the "Mussel-Watch" station. The comparisons available, however, convince us that so far the only clearly Millstone-contaminated biota samples we have analyzed were those from the effluent stream in the quarry.

This point appears to us one that cries out for follow-up, both by analysis of a larger number of the biota samples that have been collected (see listing in our Progress Report under the subject contract, submitted October 1977), and by further sampling especially at periods of high release rate (as for instance in April, 1978 of Pu, or Jan.-Feb., 1978, of ^{137}Cs).

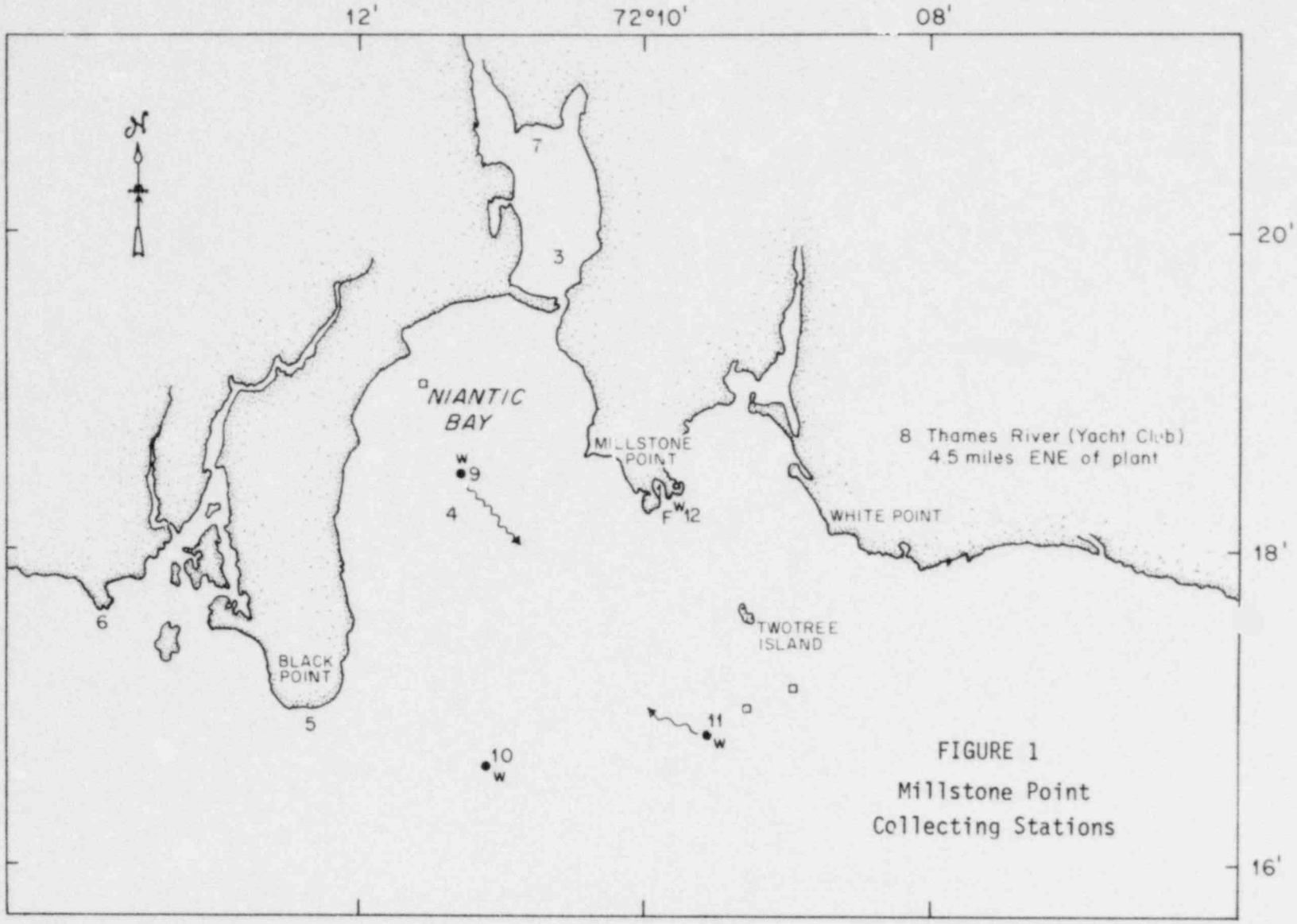

FIGURE 1

Millstone Point

Collecting Stations

TABLE 5

Millstone Point, Conn.

Long Island Sound Surface Water*

(Radionuclides in disintegrations per minute per 100 kg water)

Station	^{134}Cs	^{137}Cs	^{90}Sr	^{238}Pu	$^{239,240}Pu$	^{55}Fe
#2	3±3	29.6±3.6	28.6±0.2	0.019±0.006	0.101±0.015	7.4±1
#3	2±2	31.8±4.0	25.5±0.3	< 0.003	0.108±0.017	11±2
#4	3.6±2.5	27.0±3.1	30.4±0.2	< 0.010	0.090±0.020	11±1
#5A(outflow inner plume)	0.5±0.6	43.8±4.3	26.0±0.6	< 0.010	0.090±0.030	29±2
#5A(particulates >1μ, from outflow, inner plume)	---	---	---	0.002±0.001	0.032±0.001	26.7±0.2
#5B(particulates >1μ, from outflow, outer plume)	---	---	---	0.0008±0.0005	0.034±0.003	12.7±0.2

*For station locations see Figure 1; samples collected 20 October 1979.

TABLE 6

Millstone Point, Conn.

Sediment Cores[1] Radiochemistry

(Radionuclides in disintegrations per minute per kg dry sediment)

A. Station 2, Core #3

Section cm	Wet/Dry	^{55}Fe [8]	^{137}Cs	^{238}Pu	$^{239,240}Pu$	^{241}Am
0-1	1.53 [2]	1588±41	273±2	0.79±0.04	21.9±0.7	
1-2						
2-3						
3-4	1.40	1065±26	411±2	1.22±0.11	38.6±1.2	
4-5						
5-6						
6-7	1.34	1088±59	336±2	0.92±0.09	32.4±0.9	

B. Station 3, Core #2

0-1	2.03 [3]	325±46	600±4 [7]	3.27±0.19	129±3	32.3±1.5 [6]
1-2	1.65	274±39	487±3	2.58±0.16	102±2	
2-3	1.89	244±49	603±4	2.87±0.18	121±3	
3-5						
5-7	1.95	245±34	641±4	2.47±0.14	112±7	31.9±1.6 [6]
7-9						
9-11	1.83	388±26	498±3	3.37±0.21	130±3	
11-13						
13-15						
15-17	1.96	625±66	723±3	6.48±0.15	215±5	35.5±1.6 [6]
17-19						
19-21	1.93	---	483±3	4.98±0.12	148±4	---
21-25						
25-29						
29-33	1.77	121±32	281±2	3.34±0.22	92±3	36.1±1.9 [6]
				[4]		[5]
Inventories, mCi/km^2		30.5	45.5		12.0 [7]	
% estim. delivery		128%	38.6%		610%	

Notes:
1 = For locations see Figure 1; samples collected 20 October 1977.
2 = 70% >62μ, 30% <62μ from wet sieving.
3 = 34% >62μ, 66% <62μ from wet sieving.
4 = mean ratio ^{238}Pu:^{239}Pu = 0.028.
5 = mean ratio ^{241}Am:^{239}Pu = 0.287.
6 = No Curium detectable.
7 = ^{134}Cs less than 1/60 of ^{134} + ^{137}Cs.
8 = ^{55}Fe data decay corrected to 1 January 1975.

TABLE 7

Millstone Point Environmental Samples - Biota
(Radionuclides in disintegrations per minute per kg wet weight)

Collection Sites*	Date	Organism	Tissue	^{137}Cs	^{238}Pu	$^{239,240}Pu$	^{241}Am	^{242}Cm	^{244}Cm
Millstone Beach	Nov. 1970	M. edulis	Shell		---	0.98±0.08			
			Soft parts		0.08±0.02	0.97±0.11			
Millstone Point	Apr. 1971	M. edulis	Shell		---	1.05±0.15			
			Soft parts		0.03±0.01	0.64±0.16			
Millstone Point - Quarry	Dec. 1975	Oyster	Shell	4626±8	2.1 ±0.2	3.5 ±0.25	0.9 ±0.25	9.2±0.9	1.5±0.3
			Soft parts	1778±4	0.5 ±0.07	1.3 ±0.1	0.4 ±0.05	2.6±0.2	0.3±0.04
Black Point	Dec. 1975	Oyster	Shell	Lost	bdl	0.9 ±0.2	0.15 ±0.03	---	---
			Soft parts	20±0.3	0.02±0.01	0.3 ±0.02	ND	---	---
Millstone Point Area 2	Aug. 1977	M. edulis	Soft parts	6.1±0.3	0.005±0.003	0.3 ±0.02	0.07 ±0.01		
Fallout Contaminated Samples, for Comparison:									
Millstone Point	1976	M. edulis	Soft parts	4.1±0.2	<0.004	0.2 ±0.02	0.02 ±0.01	---	---
0.2 mi. outflow	1977	M. edulis	Soft parts	3.1±0.2	0.005±0.003	0.27±0.02	0.09 ±0.03	---	---
Herod Point, L.I.	1976	M. edulis	Soft parts	2.4±0.4	0.008±0.003	0.09±0.01	0.014±0.006	---	---
38 mi. SW	1977	M. edulis	Soft parts	3.1±0.25	0.002±0.002	0.120±0.012	<0.01	---	---
New Haven, Conn.	1976	M. edulis	Soft parts	Lost	0.011±0.004	0.24 ±0.02	0.042±0.013	---	---
42 mi. W	1977	M. edulis	Soft parts	2±0.6	0.028±0.011	0.39 ±0.04	0.07 ±0.02	---	---
	1978	M. edulis	Soft parts	3.3±0.2	0.016±0.006	0.32 ±0.03	0.042±0.008	---	---
Block Island, R.I.	1976	M. edulis	Soft parts	5.1±0.3	0.008±0.005	0.19 ±0.02	0.037±0.015	---	---
30 mi. SE	1977	M. edulis	Soft parts	2.3±0.3	0.008±0.004	0.18 ±0.02	0.04 ±0.02	---	---
	1978	M. edulis	Soft parts	3.3±0.3	<0.003	0.13 ±0.02	0.016±0.013	---	---
Sakonnet Pt., R.I.	1976	M. edulis	Soft parts	2.8±0.2	<0.005	0.15 ±0.02	0.013±0.006	---	---
	1977	M. edulis	Soft parts	2±0.3	0.011±0.004	0.22 ±0.02	0.03 ±0.01	---	---

Notes: * = See Figure 1 for collection locations.
bdl = Below detection limists.
ND = Not determined.

Wiscasset, Maine, Maine Yankee Reactor:

The setting of the Maine Yankee reactor contrasts strongly in respect to its hydrography and sedimentology, to those of Millstone Point or of Pilgrim 1. Furthermore, interpretation of our environmental data regarding the Maine Yankee reactor is affected by the transfer of the reactor effluent outflow from Bailey's Cove, west of the plant, to the diffuser in Montsweag Bay, east of the plant, a transfer that coincided with the opening of the Montsweag Bay Channel north of the plant to change the main flushing flow so it now runs northward up Montsweag Bay to Clough Point, and then southward out the Sheepscot River. Figure 2 will clarify the situation. The extreme complexity of the setting is well revealed, in respect to the channels of water circulation; it can be inferred how this affects the regimes of sediment deposition. As we discuss below, the geochemistry of the waste radionuclides is further affected by the salinity variations due to river contributions to this set of narrow passages.

As we noted earlier, our environmental samples from this area have been collected either by our own efforts, for us by people from the Darling Center, University of Maine, or for us by commercial collectors.

Water sample analyses are set out in Table 8. At the time these samples were collected, the releases of ^{137}Cs or of Pu were too low for detection. The change in the ratio ^{137}Cs to ^{90}Sr along the major direction of flow (from station 7 to 1 to 8) we believe to correlate chiefly with the position of the fresh-water lens. There is no convincing evidence, either of reactor contribution of ^{55}Fe.

In Table 9 are set out analyses of a series of surficial sediment samples collected by the Darling Center. These date from the time when the reactor outflow was still directed into Bailey's Cove, and (we believe) when the outflow was appreciably richer in some long-lived radionuclides than it seems to have been during our sampling period as summarized in Table 3. The data in Table 9 show that during the period August 1974 to June 1975, Bailey's Cove received a quite substantial release of radiocesium that is shown by its high content of ^{134}Cs to have certainly come from the Maine

Yankee reactor. The ratio ^{134}Cs to ^{137}Cs, 55 to 58%, is unusually high in our experience of environmental samples. Furthermore, we have not measured (see Table 3) ratios as high as this in the discharge samples from Maine Yankee, although as discussed above even higher ratios have been seen in the discharges from Millstone Point. Comparison of the inventories of Pu, and of the ratios ^{238}Pu to $^{239,240}Pu$, in the sediments of Table 8 with those reported by Livingston and Bowen (1979) suggests there was little or no Pu in the high-^{137}Cs discharges to Bailey's Cove. The assumption that the sediments (sampled by "scooping" at low tide) collected represent about the top 6 cm of the column leads to the conclusion that the Pu concentrations reported are well within the range expected from fallout alone; this is true also of the ratio ^{238}Pu to $^{239,240}Pu$ which is in marked contrast to the activity-weighted ratio about 0.33 that we measured in 14 samples of Maine Yankee discharge solutions. The data of Table 3, of course, confirm that there is no strong correlation between the amounts of radiocesium and of Pu in individual discharges; perhaps there is even less correlation in this respect at Maine Yankee than at Millstone Point No. 1 or 2, but this appearance may be an artifact of the sample-pooling that was practiced at the latter facilities.

We take the change in sediment radionuclide concentrations at station 2 from June to December 1975 to indicate that Bailey's Cove is frequently flushed, transferring labelled sediments to depositories elsewhere in the area. This is confirmed by the analyses of 1975 sediments from stations 1 and 15 which show evidence of reactor-originated contamination but with significantly lower ratios ^{134}Cs to ^{137}Cs.

The two sediment cores, collected in 1977, for which data are set out in Table 10, confirm and extend this conclusion. At location 2, although the radiocesium concentration-vs-depth curve comes close to expressing logarithmically the expected effect of mixing downward, the ratio ^{134}Cs to ^{137}Cs, constant in the upper 9 cm and diminishing by 90% from 9 cm to 11 cm, suggests this sediment column has seen only two radiocesium releases, differing greatly in age or ^{134}Cs content. The fluctuations, with depth, of the ratios of either $^{239,240}Pu$ or ^{55}Fe to ^{137}Cs, do not show the patterns that we would expect to result from resolubilization and redistribution within the sediment, as discussed for Pu:^{137}Cs ratios by Livingston and Bowen (1979). More likely, we believe,

is the explanation based on redeposition, after resuspension and mixing, of sediments deposited in various parts of the core and contaminated by separate waste release events. In this context the range indicated for the ratio ^{55}Fe to ^{137}Cs is quite interesting: nowhere less than 90%, and ranging upward to 1.25 at the surface, or to 1.40 at 10-11 cm. Evidently the three releases (Table 3) in which we measured ^{55}Fe were from the low end of the range of its concentrations, or of its ratios to ^{137}Cs. Also evident is the fact that, as was seen at Millstone Point (above), ^{55}Fe is a major long-lived component of the releases from the Maine Yankee reactor; in evaluating these data it is important to bear in mind the ^{55}Fe expected from worldwide fallout: although Joseph et al (1971) estimated that 1962-1966 fallout was characterized by a ratio ^{55}Fe to ^{90}Sr of 9.4 (= ^{55}Fe to ^{137}Cs, 6.5), the elapse of 4 ^{55}Fe half-lives by 1975 reduced this to ^{55}Fe to ^{137}Cs of 0.406 or so. Although geochemical processes will affect Fe and Cs in different ways, fallout ^{55}Fe is unlikely often to distort the reactor waste patterns. The profile of Pu in this core, however, could again perfectly well be attributed all to fallout, arguing either from its similarity to data reported by Livingston and Bowen (1979), or from its ratio ^{238}Pu to $^{239,240}Pu$. The inventory estimates (all minima, because the core was too short to include the full depth of radionuclide-contaminated sediments) show large excesses, as expected from the concentration data, of ^{55}Fe (250% of fallout) or of ^{137}Cs (~65 $mCi \cdot km^{-2}$ vs the range 4 to 15 $mCi \cdot km^{-2}$ reported by Livingston and Bowen, 1979), but a deficiency of Pu.

Core No. 3, across the Montsweag Channel from the present diffuser release point for plant effluent, presents a different picture still. Both the somewhat elevated ratio ^{238}Pu to $^{239,240}Pu$, and the concentration vs depth-in-core curve of $^{239,240}Pu$ suggest that here we have a site of relatively continuous sediment deposition, and that the radionuclide concentrations are showing significant contributions from the reactor wastes. The ratio ^{134}Cs to ^{137}Cs in this core shows unusual variability, with no significant ^{134}Cs at the top, or in the 7-9 cm section, but a substantial concentration in the 1-2 cm section. This section, in fact, is not distinguishable from the upper parts of the station 2 core in terms of its Cs isotope ratio, although clearly different in terms of Cs:Pu, and Pu isotope ratios.

Contamination of this Montsweag Channel area clearly did not occur by simple transfer of sediments from Bailey's Cove.

Comparison of the data from Core No. 3 with those from analysis of sediments from the mud flat, across the river and a little north of the diffuser (Table 12, below) raises some other questions. Although the ^{137}Cs is about 50% higher on the mud flat, assuming the top 6-cm were collected, the Pu concentration is significantly lower and shows no elevation in ^{238}Pu. In fact, on the basis of the data now in hand, the mud-flat sediment looks more like material transported from Bailey's Cove than it does like the core from across the river. It is very evident that ^{55}Fe analyses would help clarify this interpretation, as will ^{134}Cs analyses that are now in process.

The analyses of Wiscasset-Area biota collected in Table 11 show reasonably clear evidence of contamination by reactor wastes. This is true for the ^{137}Cs and Pu contents of the 1973 oyster shells, which are substantially above those we would have predicted from fallout alone; it is true, however, that the ratios ^{238}Pu to $^{239,240}Pu$ are not elevated as reactor effluent samples show (Table 3), and that the sample from location 10, at Boothbay ME, quite remote from the Maine Yankee reactor, is high in both ^{137}Cs and $^{239,240}Pu$. We did not, however, collect these samples ourselves and incline to believe this sample wrongly identified. The two pairs of algae (Fucus vesiculosus and Ascophyllum nodosum) from location 14 at the mouth of Bailey's Cove versus Boothbay, agree in showing substantial elevations of ^{137}Cs near the reactor. We have some suspicion of the Pu concentration in Ascophyllum from Boothbay; as illustrated by the two samples from location 14, and also by paired samples from the Plymouth area (below, in Table 14) our usual observation has been that, from the same location, Ascophyllum accumulates less Pu than does Fucus. The Mya arenaria sample from location 6 appears high in both ^{137}Cs and Pu, although the ^{238}Pu to ^{239,240} ratio is not elevated. The two Mytilus samples, from location 11 and (we believe) from near the diffuser of the plant, show only fallout-contamination levels of either ^{137}Cs or Pu. The ^{55}Fe in the second of these samples does look high, however; in Narragansett Bay (and Plymouth area

7) mussels we have found ratios of ^{55}Fe to $^{239,240}Pu$ in the 30 to 50 range, significantly different from the value close to 100 reported here. Too little comparison data is available concerning ^{55}Fe in nearby mussels for us to be perfectly sure of this situation. Inspection of the lower half of Table 11, as was discussed above for Table 7, clearly shows that the neighborhood near Maine Yankee reactor is not characterized by radionuclide contamination of mussel samples.

In Table 12 we have set out the data from another experiment, prompted by the reports of Noshkin et al (1971), of Fowler et al (1975), and Beasley and Fowler (1976a, 1976b) that nereid worms took up substantial portions of Pu either from water or from a variety of types of contaminated sediments. A commercial worm collector obtained for us a large sample of sandworms from the mud flat just north of the Maine Yankee effluent diffuser, and another sample from a similar mud-flat, contaminated only by fallout, near Portland, ME. At each location he also scooped a sample of mud. About half of each worm sample he preserved for us in formalin, and the other half he induced, by methods he was not willing to describe, to purge their guts of contained mud before being preserved. The worms, and the sediments, were analyzed by our usual procedures.

The Wiscasset worms contained elevated levels of ^{137}Cs and of Pu; whole worms contained ^{137}Cs at a concentration about 10% and Pu about 22.7% that (estimated) in the wet sediment from which they were collected. Cleaning reduced the ^{137}Cs in worms to 24% and the Pu to 7.7% of the uncleaned value. There was some reduction in the dry weight fraction of worm tissue during the cleaning process, so it is possible that not all the loss of radionuclides was due, simply, to evacuation of gut content, but we have little doubt this was the chief process. The whole worms from Portland contained much higher percentages of sediment tracer: 49% of ^{137}Cs, and 53% of Pu; also, of these concentrations higher fractions were in the tissues: 42% of ^{137}Cs and 19.3% of Pu. Another indication that the nuclides from the fallout-contaminated situation were somewhat different, chemically, than those from the reactor-contaminated situation came from the formalin-solubilization data shown at the bottom of Table 12: The latter situation produced worms with only 2/3 of tissue ^{137}Cs, but 18% of tissue Pu,soluble in formalin, whereas the fallout contaminated worms showed all their tissue ^{137}Cs, but only 12% of their tissue Pu soluble

with the same treatment. Without these last data we would be inclined to suppose that the Wiscasset worms, whose exposure occurred during residence in a very small area of reactor-contaminated mud, simply had not yet come to equilibrium with the higher tracer concentrations; this is a reasonable contrast to the Portland worms, all of whose lives had, by definition, been spent in environments contaminated by fallout. The solubilization data suggest strongly, however, that there are differences in metabolic reactivity of the tracers from fallout or from reactor effluent, that fallout tracers are considerably more available to nereid worms for tissue uptake, than are reactor tracers, but that the distributions within the tissues are still different. Evidently this is an area of investigation that should be of real interest to NRC as it is to us.

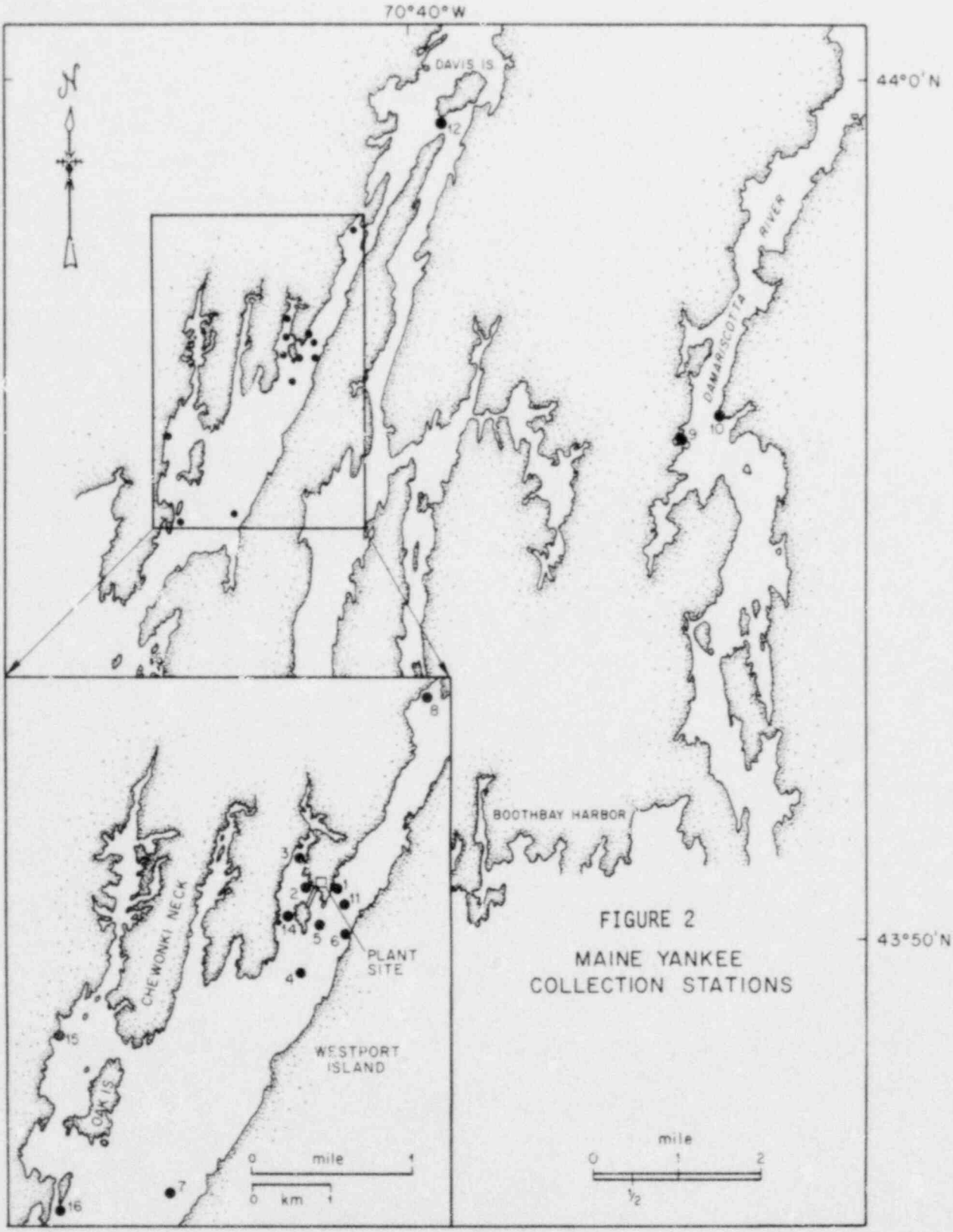

FIGURE 2

MAINE YANKEE
COLLECTION STATIONS

TABLE 8

Wiscasset, Maine, Area

Surface Water from Montsweag Bay*

(Radionuclides in disintegrations per minute per 100 kg water)

Station	^{137}Cs	^{90}Sr	^{238}Pu	$^{239,240}Pu$	^{55}Fe[1]
7	22.5±0.2	72.5±0.6	<0.005	0.21±0.02	11.8±8
8	28.8±0.3	41.0±1.0	0.004±0.004	0.21±0.02	8±1.4
1 (outflow)	26.0±0.2	42.4±1.1	<0.005	0.27±0.03	6.9±0.8

* = See Figure 2 for station locations; samples collected 5 January 1977.

1 = Decay correction to 1 January 1975.

TABLE 9

Wiscasset, Maine, Area

Surficial Sediment Radiochemistry, I

(Radionuclides in disintegrations per minute per kg dry sediment)

Sample Location[1]	Date	^{134}Cs [2]	^{137}Cs	$^{134}Cs/^{137}Cs$ [2]	^{238}Pu	$^{239,240}Pu$	$^{238}Pu/^{239,240}Pu$
2	14 Aug 1974	<20	885±30	<0.02	4.04±0.73	95±7.7	0.043
2	25 June 1975	16,000±600	27,700±300	0.58	3.03±0.27	71±2.2	0.042
2	8 Dec. 1975	3840±230	8,260±110	0.46	3.55±0.41	98±4	0.036
1	30 May 1975	340±50	1,030±40	0.33	2.17±0.23	63±2.5	0.035
3	30 June 1975	10,600±500	19,200±40	0.55	7.6±0.5	174±5	0.044
15	"	500±50	1,620±30	0.31	3.13±0.24	87±2.8	0.036
16	"	lost	lost	--	0.80±0.08	22.4±0.6	0.035

Notes: 1 - For locations see Figure 2.

2 - Decay corrected to date of collection

TABLE 10

Wiscasset, Maine, Area

Sediment Cores[1] Radiochemistry, II

(Radionuclides in disintegrations per minute per kg dry sediment)

A. Core #3 (Location 6)

Section cm	Wet/Dry	^{55}Fe [2]	$^{134,137}Cs$ [3]	$^{134}Cs/^{137}Cs$ [3]	^{238}Pu	$^{239,240}Pu$
0-1	2.26		534±5	0.09±0.10	6.4±0.4	98±3
1-2	1.72		994±8	0.350±0.016	4.2±0.4	79±3
2-3						
3-4	2.34		872±7		5.8±0.5	87±4
4-5						
5-6	2.68		713±7		3.6±0.3	68.4±2
6-7						
7-9	--		552±4	0.06±0.12	4.4±0.3	101±3
9-11						
11-13	--		lost			lost

B. Core #4 (Location 2)

0-1	2.30	8700±240	10650±12	0.35	3.1±0.2	75.2±1.7
1-2	1.88	4580±135	6765±25	0.37	1.5±0.25	39.9±0.1
2-3	1.63	3360±120	7560±20	0.36	1.1±0.2	29.8±1.2
3-4						
4-5	1.65	670±50	2050±8	0.38	0.40±0.12	14.5±1.1
5-6						
6-7	1.69	500±40	1280±6	0.35	0.33±0.11	8.0±0.4
7-8						
8-9	1.59	430±50	495±4	0.35	0.05±0.05	3.4±0.4
9-10						
10-11	1.61	335±40	240±3	0.033	0.07±0.06	4.9±0.6
11-13						
13-15	1.65	lost	lost	lost	lost	lost
15-17						
Inventories:						
mCi/km^2:		61.7	102	---	---	0.605
% estimated delivery:		250%	83%			29%

Notes: 1 = For locations see Figure 2; samples collected 5 January 1977.
2 = ^{55}Fe data decay corrected to 1 January 1975.
3 = Decay corrected to date of collection.

TABLE 11

Wiscasset, Me, Area: Environmental Samples - Biota I

(Radionuclides in disintegrations per minute per kg wet weight)

Collection Sites*	Organism	Tissue	Date	^{55}Fe	^{137}Cs	^{238}Pu	$^{239,240}Pu$	^{241}Am
1	C. virginiana	Shell	Oct. 1973		25±0.8	0.13±0.04	3.1±0.2	
2	"	"	"		18±0.5	0.09±0.03	2.6±0.2	0.7±0.1
3	"	"	"		33±1.2	lost	lost	
4	"	"	"		11±1.4	0.06±0.01	1.4±0.8	
10	"	"	"		29±0.9	0.22±0.05	5±0.25	
14 Boothbay	A. nodosum	Whole	Aug. 1976		40±0.4	0.07±0.03	1.53±0.17	
	" "	"	Sept. 1976		27.6±0.3	0.11±0.03	4.16±0.29	0.690±0.072
14 Boothbay	F. vesicularus	"	Aug. 1976		53±0.5	0.05±0.02	2.7±0.2	
"	" "	"	Sept. 1976		14±0.3	0.07±0.01	1.4±0.1	
	Green Urchin	"	" 1976		4.1±0.2	0.035±0.007	0.60±0.03	
6	M. arenaria	Soft Parts	May 1977		21±0.5	0.043±0.012	0.57±0.05	
11	M. edulis	" "	Dec. 1977		5.2±0.3	bdl	0.25±0.02	
?	" "	" "	Nov. 1977	13.1±1.2	3.7±0.5	0.006±0.004	0.14±0.02	
Fallout contaminated samples, for comparison:								
Sears Isl.	M. edulis	Soft Parts	1976		2.9±0.2	0.006±0.005	0.34±0.03	0.069±0.009
53 mi. NE.	" "	" "	1977		2.6±0.4	0.004±0.003	0.31±0.025	0.07±0.01
	" "	" "	1978		3.7±0.4	0.005±0.004	0.22±0.03	0.06±0.02
Cape Newagen	" "	" "	1976		2.2±0.3	0.011±0.006	0.25±0.03	0.09±0.02
11.5 mi. S.	" "	" "	1977		1.7±0.4	0.032±0.012	0.38±0.04	0.05±0.01
Bailey Isl.	" "	" "	1976		2.5±0.3	0.010±0.007	0.29±0.03	0.054±0.015
21.5 mi. SW.	" "	" "	1977		2.4±0.3	0.005±0.003	0.31±0.03	0.06±0.01
	" "	" "	1978		2.8±0.3	ND	0.27±0.03	0.04±0.02
Portland	" "	" "	1976		3.4±0.3	<0.003	0.21±0.02	0.06±0.01
36 mi SW	" "	" "	1977		9.1±0.2φ	0.006±0.002	0.27±0.02	0.07±0.01

Notes:
- * See Figure 2 for collection locations.
- ? Collected by Maine Yankee, site not reported but thought to have been near diffuser.
- φ Result of replicated counting.
- bdl Below detection limits.
- ND Not determined.

TABLE 12

Wiscasset, Me ,Area:Environmental Samples - Biota II

SANDWORMS (Nereis versicolor)

(Radionuclides in disintegrations per minute per kg wet weight)

Collection Sites[1]	Sample	Date	^{137}Cs	$^{238}Pu/^{239}Pu$	$^{239,240}Pu$
Wiscasset (Area 1)	Whole Worms	1-9 Aug. 1977	50.3±0.4	0.036±0.004	4.71±0.145
	Cleaned [2]	"	12.2±0.3	0.023±0.010	0.36±0.025
	Sediment [3]	"	1524±10	0.041±0.005	62.5±1.6
	[4]		(508)		(20.8)
Portland	Whole Worms	"	18.1±0.3	0.036±0.007	1.69±0.08
	Cleaned "	"	7.6±0.3	0.045±0.021	0.325±0.029
	Sediment [3]	"	111±3	0.028±0.013	9.57±0.82
	[4]		(37)		(3.2)

Notes:
1 Wiscasset worms from mud-flat just N. of diffuser; Portland worms from area expected to represent only fallout contamination.
2 Cleaned by being allowed to evacuate gut contents.
3 Sediment data as dpm per kg dry sediment.
4 Sediment data calculated as dpm per kg wet sediment, assuming wet/dry ratio = 3.

Ratios:

Wiscasset	Tissue concentration as % of whole-worm concentration	24%	7.7%
Portland	" " "	42%	19.3%

Extractions: Fraction of each nuclide soluble after 44 days in 5% Formaldehyde sea water

Wiscasset	Whole	3%	1%
	Cleaned	63%	18%
Portland	Whole	4%	8%
	Cleaned	116%	12%

Plymouth, Mass., Pilgrim 1 Reactor:

During the period of this project, we were unable to obtain any samples of discharge solutions from the Pilgrim 1 reactor. Our attention had, however, been drawn to its environmental effects by the finding in the first series of "Mussel Watch" samples (Goldberg et al, 1978) of mussels from near Pilgrim 1 (station 7, Figure 3) that exhibited definitely high Pu levels. This stimulation, plus the fact that the Plymouth area is very easily accessible to us, either by car or boat, led to our doing a good deal of collecting of biological material for radiochemical analysis.

Failing cooperation from the Pilgrim 1 reactor operators, we did obtain (via a local fisherman) one water sample from the reactor outflow. The data from this analysis are set out in Table 13, together with a small number of relevant sediment analyses. The water sample showed no evidence of reactor output of ^{137}Cs or of Pu; its ratio ^{137}Cs to ^{90}Sr was as we would expect of a coastal seawater sample (Bowen et al, 1974). Evidently either no releases were in progress at the time of sampling, or the sample was taken at a poor place. Inspection of Figure 3 will show that the bars protecting Plymouth Harbor and Duxbury Bay form an excellent sediment trap for any materials carried northward (clockwise) from the Pilgrim 1 outfall. Our Duxbury Bay sample, like the others in the Plymouth area, shows no anomalies either in the concentrations or in the ratios of the nuclides measured. The core from near the center of Cape Cod Bay falls well within the range of radionuclide concentrations, and ratios, reported for near-shore sediments (Livingston and Bowen, 1979); this core was taken to explore the hypothesis that the gyral circulation of the bay might concentrate fine sediments from the periphery to be deposited centrally. Evidently we have no evidence that this happens.

In a way, it is odd that we have not found a site with reactor contaminated sediments since we seem to have found evidence for such contamination of biota in several places. The relevant data concerning samples from the area about Plymouth are set out in Tables 14A and 14B; comparison data from mussels, collected either in Boston Harbor or the Cape Cod Canal, are set out in Table 15.

It is worthwhile to start by pointing out that macroalgae, like Fucus, Ascophyllum, or the mixed Red Algae analyzed, are well established as showing seasonal variability in their concentrations of many trace elements; we would not, then, expect to be able to compare directly collections taken at different times. Much the same is proving to be true of mussels (Bowen et al, to be published). Examples of this phenomenon in the case of macroalgae are to be seen comparing the samples from station 1 (Table 14A) collected late October 1976 with those collected early May 1977:

	Chondrus	25.2	^{137}Cs;	1.07	^{239,240}Pu
Fall	Fucus	13.4	" ;	1.9	"
	Red Algae	47.9	" ;	2	"
	Chondrus	38.1	" ;	0.67	"
Spring	Fucus	12.7	" ;	1.33	"
	Red Algae	21.9	" ;	0.66	"

It appears that the seasonal change has led to increased ^{137}Cs in Chondrus, decreased in the Red Algae, and no change in Fucus, while each spring sample shows lower Pu.

If we compare given species collected at different sites at about the same time, however, then we expect to see differences that characterize the sites. Examples are fall-collected Fucus or Ascophyllum from site 1 (Figure 3) compared to site 2, which we now believe is mostly up-current from the Pilgrim 1 reactor outfall, or spring-collected Fucus from site 1 compared to site 3, downstream and behind the bar in Plymouth Harbor. Site 1 Fucus, Ascophyllum, and Red Algae are significantly richer in ^{137}Cs than are those from site 2, and the first two from site 1 are also richer in Pu. At site 3 Fucus collected in spring were richer by almost a factor of 3 in ^{137}Cs than at site 2, and significantly richer in Pu also. The Red Algae (a collection of mixed genera) were richer in Pu at site 2 both in fall and in spring; it is, however, possible that the genera in this mixture are not truly comparable between the two sites. It should be pointed out that none of these samples contained concentrations of any nuclide we measured that were sufficiently elevated to cause alarm. The Pu data can be compared with those summarized by Livingston and Bowen (1976); none of the samples, for instance, exceeds the range that those data predict to have been characteristic of fallout contamination in the 1970-

1971 period. The concentration factors in algae that are indicated, using as basis the data of the water sample in Table 13, range for ^{137}Cs from 20 to 150, and for Pu from 1000 to 2000. These are very like the ranges, for Pu, reported by Livingston and Bowen (1976).

Mytilus edulis, the common blue mussel, is the animal concerning which we have most information, and as we have shown earlier, most other data for comparison. In respect to ^{137}Cs and to Pu most of the mussel samples shown in Tables 14A and 14B are not clearly outside of the ranges expected in this general area (see Table 15) from fallout alone. At station 1, the sample from 29 October 1976 is unusually rich in ^{137}Cs, but elevations as great as this have been reported elsewhere (see Tables 7,11). The other sample from site 1, that from site 2, one of the two from site 3, and three of the four from site 7, all are within the "normal" range. Those mussels that appear abnormally high in some radionuclides are as follows:

Site 1: 10/29/76 ^{137}Cs
Site 1: 5/28/78 ^{55}Fe (ratio to Pu is 390, vs 30 to 50 elsewhere)
Site 2: None
Site 3: 6/15/71 ^{137}Cs, Pu (but these may be in fallout range for that year; see Livingston and Bowen, 1976, Table 3)
Site 5: 10/27/76 Pu
Site 7: 9/23/76 Pu

We have no question that the site 1 high ^{55}Fe and the two high Pu, one from site 5 and one from site 7 are real and are related to the output from the Pilgrim 1 reactor. These are factor of 2-3 (Pu) or 10 (^{55}Fe) effects, and the site 7 high Pu was accompanied by the lowest ^{241}Am we have seen in a mussel (or other environmental sample); a ratio ^{241}Am to $^{239,240}Pu$ less than 5% really establishes that the Pu-Am mixture must have been produced very recently, certainly less than 3 years and from the irradiation of uranium fuel. Such an origin is also confirmed (or at least not contraindicated) by the relatively low ratio ^{238}Pu to $^{239,240}Pu$. Unfortunately the ^{241}Am sample from site 5 still remains to be measured, having somehow gotten jostled out of its priority position.

The other animals reported in Tables 14A or B fall into two

classes: grazers like Mytilus: These are Littorina, Strongylocentrotus, Balanus, and Spisula; or predators, largely eating grazers: These are Homarus, Lunatia, Buccinum, and the Eider Duck. At the start of this program we had some data, from analysis of starfish eating analyzed mussels, suggesting that invertebrate predators by some mechanism exhibit higher actinide concentrations than do the grazers on which they feed. One of our purposes in undertaking the present project was to explore this question and especially to establish whether in an area of sporadic higher contamination of grazers with transuranic nuclide wastes, tissue concentrations in predators might systematically rise to levels high enough to cause concern. Comparison among the data points reported here, and with analyses reported by Livingston and Bowen (1976), leads us to the following conclusions:

1. Strongylocentrotus and Littorina from site 2, and Balanus from site 3, probably show no evidence of Pilgrim 1 waste radionuclides.
2. Spisula from site 8 exhibits probably about double the Pu concentration expected from fallout alone.
3. Neither lobster nor eider-duck show evidence of concentrations of ^{137}Cs or Pu in their tissues that are higher than, or even as high as, those inferred for their prey.
4. Buccinum and Lunatia probably agree with our earlier observations on starfish in showing tissue Pu two to three times higher than in their prey, but there is no evidence from comparison of site 3 data with that either from site 2, or reported by Livingston and Bowen (1976), that these concentrations escalate systematically in organisms from an area that sometimes receives the reactor coolant stream.

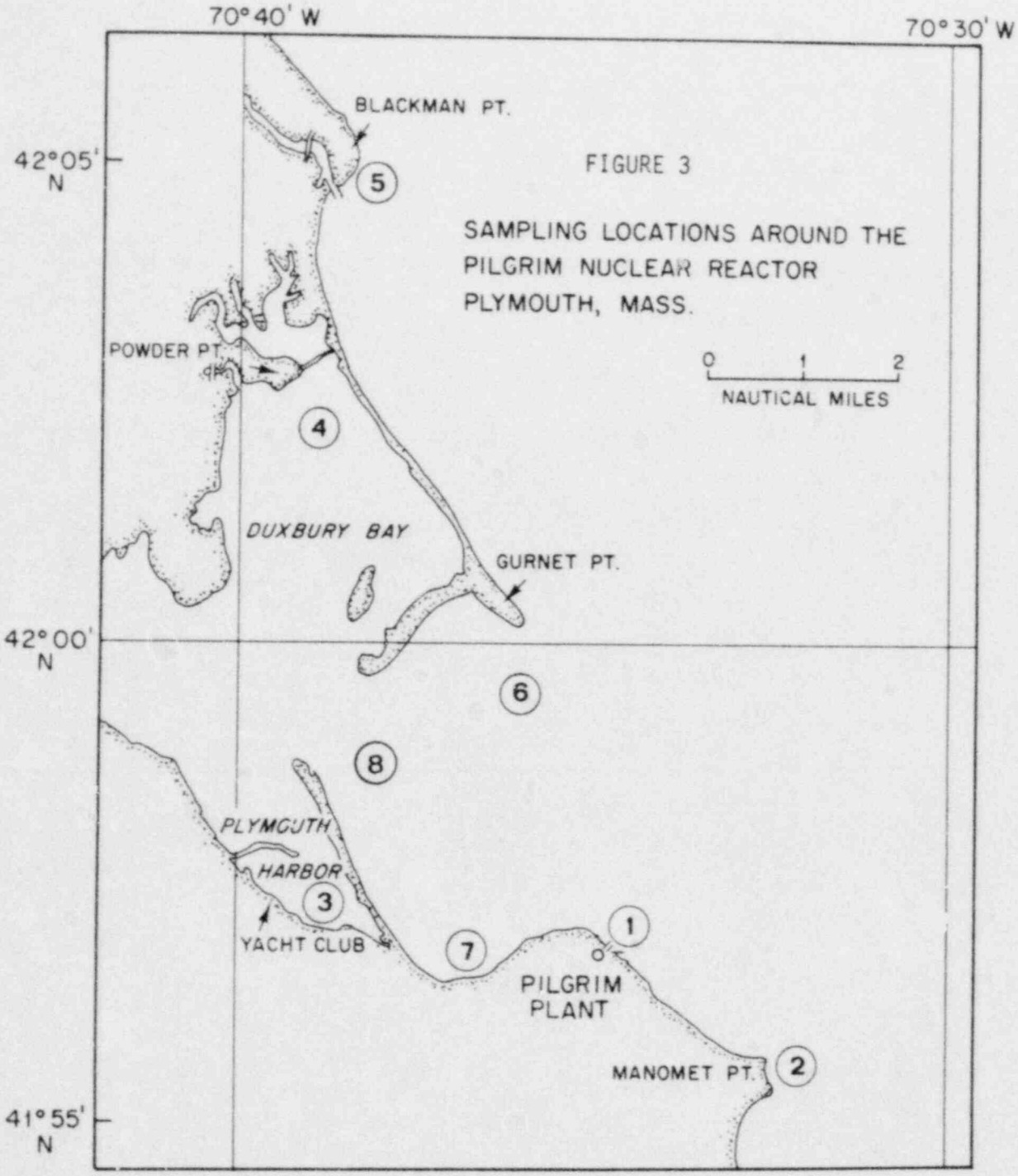

FIGURE 3

SAMPLING LOCATIONS AROUND THE PILGRIM NUCLEAR REACTOR PLYMOUTH, MASS.

TABLE 13

Plymouth, Mass., Area: Water and Sediment Radiochemistry
(Radionuclides in disintegrations per minute per 100 kg water, or per kg dry sediment)

Part A - Water Analyses:

Collection Site:*	Date 1976	Depth - cm	^{90}Sr	^{137}Cs	^{238}Pu	$^{239,240}Pu$	$^{238}Pu/^{239}Pu$
1 +	Oct.	surface	36.2±0.6	32.0±0.5	N.M.	0.15±0.06	---

Part B - Sediment Analyses:

1 x	Oct. 20	0-3	Grab	66.1±1.5	0.29±0.04	8.2±0.3	0.035
2	Oct. 19	0-4	Core	157±3	2.6 ±0.2	61.5±1.7	0.042
4	Oct. 19	0-6	from Anchor	59.7±1.3	0.68±0.07	21.3±0.6	0.032
Cape Cod Bay	Aug.	0-1	Core	187±6	2.5 ±0.3	101±3	0.034
		1-2	Core	181±3	2.8 ±0.2	75±2	0.037
		3-4	Core	176±2	2.6 ±0.2	69±2	0.038
		5-6	Core	181±3	2.7 ±0.3	62±3	0.043
		8-10	Core	174±3	2.5 ±0.3	69±3	0.036
		12-14	Core	198±3	2.4 ±0.2	69±2	0.034
		16-18	Core	107±3	1 ±0.1	30±1	0.032

Notes: * = For locations, see Figure 3.

+ = Water directly from Pilgrim I outfall.

x = Sediment from plant intake basin.

TABLE 14A

Plymouth, Mass. Area Environmental Samples - Biota

(Radionuclides in disintegrations per minute per kg fresh weight)

Collecting Site[1]	Date	Organism[3]	^{55}Fe[2]	^{137}Cs	^{238}Pu	$^{239,240}Pu$	^{241}Am
1	Mar 2 71	Chondrus crispus	N.D.	lost	N.D.	0.8±0.4	0.044±0.018
1	Oct 29 76	" "	N.D.	25.2±0.3	0.10±0.03	1.07±0.11	N.D.
1	" "	Fucus vesiculosus	N.D.	13.4±0.2	0.045±0.022	1.9±0.2	N.D.
1	" "	Ascophyllum nodosum	N.D.	14.5±0.2	0.027±0.020	0.97±0.15	N.D.
1	" "	Red Algae - mixed	N.D.	47.9±0.3	0.087±0.014	2±0.08	0.31±0.03
1	" "	Mytilus edulis	N.D.	9.1±0.2	0.017±0.004	0.30±0.02	N.D.
1	May 5 77	Chondrus crispus	N.D.	38.1±0.4	0.017±0.006	0.67±0.09	N.D.
1	" "	Fucus vesiculosus	N.D.	12.7±0.3	0.059±0.011	1.33±0.06	N.D.
1	" "	Red Algae - mixed	N.D.	21.9±0.4	0.04±0.01	0.66±0.04	N.D.
1	Aug 22 77	Homarus - flesh	N.D.	6.1±0.5	<0.002	0.006±0.003	N.D.
1	" "	" - guts	N.D.	7.4±1.0	<0.007	0.260±0.043	N.D.
1	May 28 78	Mytilus edulis	112±4	1.6±0.2	0.005±0.002	0.29±0.02	0.086±0.012
2	Aug 27 75	Lunatia heros	N.D.	20.3±0.3	0.034±0.005	0.65±0.03	N.D.
2	Aug 28 75	Strongylocentrotus	N.D.	5.9±0.4	0.05±0.02	0.37±0.06	N.D.
2	Oct 29 76	Fucus vesiculosus	N.D.	9.9±0.2	0.083±0.016	1.62±0.10	N.D.
2	" "	Ascophyllum nodosum	N.D.	7.6±0.2	0.024±0.015	0.86±0.11	0.05±0.027
2	" "	Red Algae - mixed	N.D.	19.5±0.2	0.16±0.02	3.58±0.15	0.54±0.04
2	" "	Littorina littorea	N.D.	16±0.5	0.040±0.011	0.75±0.05	N.D.
2	" "	Mytilus edulis	N.D.	3.8±0.2	0.013±0.004	0.23±0.02	N.D.
2	May 5 77	Red Algae - mixed	N.D.	17.1±0.4	0.045±0.009	1.21±0.06	N.D.
2	Oct 24 77	Eiderduck - flesh	N.D.	6.1±0.2	<0.002	0.014±0.005	N.D.
2	" "	" - bones	N.D.	3±5	0.005±0.005	0.026±0.013	N.D.
2	" "	" - livers	N.D.	4.4±1.3	<0.007	0.015±0.010	N.D.
2	" "	" - guts	N.D.	3.8±0.3	<0.001	0.026±0.005	N.D.

Notes: See Table B.

TABLE 14B

Plymouth, Mass. Area Environmental Samples - Biota

(Radionuclides in disintegrations per minute per kg fresh weight)

Collecting Site[1]	Date	Organism	^{55}Fe[2]	^{137}Cs	^{238}Pu	$^{239,240}Pu$	^{241}Am
3	June 15 71	Fucus vesiculosus	N.D.	33.2±0.9	0.26±0.09	1.6±0.1	N.D.
3	June 15 71	Mytilus edulis	N.D.	13.6±1.2	0.03±0.02	0.57±0.09	N.D.
3	Aug 8 74	Buccinum un[illegible]	N.D.	4.6±0.3	0.03±0.01	0.75±0.04	N.D.
3	" "	Lunatia	N.D.	39±3	0.05±0.01	0.69±0.04	N.D.
3	Aug 28 75	Myti[illegible]s	N.D.	3±0.2	0.02±0.01	0.35±0.05	N.D.
3	" "	Balanus [illegible]alanoides	N.D.	6±1	<0.1	0.6±0.3	N.D.
5	Oct 27 76	Mytilus edulis	N.D.	2.5±0.2	0.026±0.005	0.77±0.034	N.D.
7	Sept 23 76	Mytilus edulis	N.D.	2.0±0.2	0.02±0.02	0.67±0.09[d]	0.003±0.007
7	June 23 77	Laminaria longicruris	N.D.	12.5±0.6	0.132±0.018	2.72±0.10	0.186±0.031
7	" "	Mytilus edulis	N.D.	3.1±0.3	0.013±0.007	0.149±0.024	N.D.
7	Aug 18 77	Mytilus edulis	N.D.	2.6±0.3	0.008±0.004	0.263±0.024	0.05±0.02
7	Sept 26 78	Mytilus edulis	6.4±1.0	1.6±0.4	0.006±0.002	0.14±0.005[D]	0.032±0.004[D]
Brown's Bank (8)	Apr 6 77	Spisula solidissima	N.D.	3.2±0.4	<0.005	0.40±0.05	N.D.

Notes:
1 - See Figure 3 for location of collecting sites.
2 - Decay corrected to date of collection
3 - For common names or detailed taxonomy, see Appendix Table.
N.D. - Not measured.
d - Result of duplicate counts.
D - Mean of duplicate analyses.

TABLE 15

Biota Samples for Comparison[1] to Plymouth Area (Tables 14A + 14B)

(Radionuclides in Disintegrations per Minute per Kg Wet Weight)

Collection Site	Date	Organism[2]	^{137}Cs	^{238}Pu	$^{239,240}Pu$	^{241}Am
Boston, MA[3]	9-22-76	Mytilus edulis	1.8±0.3	0.002	0.11±0.02	0.008±0.004
	8-18-77	" "	2.1±0.2	0.004±0.002	0.16±0.01	0.02 ±0.01
Cape Cod Canal[4]	9-25-76	" "	1.9±0.3	0.012±0.006	0.13±0.02	0.020±0.008
	8-19-77	" "	3.9±0.2	0.021±0.005	0.26±0.02	0.05 ±0.01

Notes: 1 = See also fallout-contaminated mussel data on Tables 7 and 11.
2 = Soft parts only
3 = About 32.5 miles NW of Pilgrim 1 reactor site
4 = North end of canal; about 17.5 miles south of Pilgrim 1 reactor site.

SUMMARY AND CONCLUSIONS

It is reasonable to preface this summary with the statement of our strong dissatisfaction at the outcome of the project. All of our early discussions with NRC emphasized that an appreciable time would be needed to provide either enough data, or the right sampling coverage, to answer either the questions of interest to us, or those we believed certain ultimately to concern NRC. Our initial proposal stated explicitly (page 7): "The first year will be devoted to survey work...detailed studies will begin only in the second year. This is a long-term program". What we have to report here is just the bare bones of preliminary work, that was planned to provide a skeleton for the program in subsequent years; that any conclusions other than decisions what to sample and/or analyze next can be drawn from these data is pure good fortune. We should emphasize also that NRC has profited enormously in respect to the interpretation of the analyses of biota collected under this program, by the availability of data from the Mussel-Watch program (Goldberg et al, 1978) supported by EPA. It appears from where we sit that both NRC and EPA would benefit from some liason in matters of this sort.

We are familiar with the type of change of policy that led to the decision to stop support of this project after one year, and do not wish to appear to be complaining about that. The mistake that we believe we see on NRC's part was the complete avoidance of any communication with us. An occasional discussion of the ways in which the interests of NRC, or of the Division of Reactor Safety Research, were being redefined would have permitted us to adapt to some of these changes, and at least to argue against others. The result of this complete absence of any interactive communication between the agency and the researchers it supported has been that we complete this report with a strong feeling that we have been shortchanged, and furthermore that we were led, without being at fault, to shortchange NRC. Obviously, there can be no thought of correcting, or of ameliorating, our unhappy situation; it would be possible, and we urge NRC to consider it, for the Agency to change its practices in contract administration.

Summary

The data collected so far concerning the amounts of long-lived radionuclides in wastes released from the two reactors at Millstone Point, and from the Maine Yankee reactor, confirm that enough ^{55}Fe, ^{60}Co, $^{134,137}Cs$ are released to provide the bases for very useful tracer studies of their biogeochemistry; at least sporadically this appears also to be true of Pu and Am nuclides. Changes made during the period, in 1977-78, of our study at Millstone Point appear to have reduced the levels of Cm released below those likely to be useful, and we have so far no evidence that useful -- or even measurable -- amounts of Cm are released by either Maine Yankee or Pilgrim 1.

Analyses of sediments in the area around Maine Yankee, and of biota around both Maine Yankee and Pilgrim 1 have shown both that enough tracer is available in these environments to be useful, and that we have found enough foci of at least sporadic contamination to provide the framework for the sort of local biogeochemical experiments that were contemplated at the start of this project. At Millstone Point we have not yet found the suitable foci, and it is quite likely that much more work would be needed to do this. We believe, furthermore, that the experience gathered in this project so far has provided us with a generalizable basis that can be used for identifying other reactor sites that have high probability of being suitable loci for biogeochemical studies of their waste products. It is, of course, evident that from the point of view of defending NRC's reactor site acceptability criteria, especially against anti-nuclear forces, the same factors that suggest a site will be a suitable locus for biogeochemical studies, suggest also that it will have greater difficulty in achieving public acceptance.

A set of more-or-less specific conclusions follows:

Conclusions

Nuclides Studied

^{55}Fe is clearly a major waste product of each of the reactors studied, approximating the levels of ^{60}Co released.

^{134}Cs is released, generally, at nearly uniform ratio to ^{137}Cs, so that the change in this ratio with time can be used as a guide to dating movements of released radiocesium.

^{238}Pu is released at very high ratios to $^{239,240}Pu$, ranging from 1 to as high as 5. This should be useful in distinguishing reactor Pu from fallout Pu.

Cm is now being released in only very small amount from Millstone Point, and has not yet been detected from either Maine Yankee or Pilgrim 1.

Release Patterns

The rates of release of radiocesium are not useful predictors of the rates of release either of activation products like ^{55}Fe or ^{60}Co, or of transuranics like Pu, Am or Cm. Further study will be needed to identify those easily measured (by gamma spectrometry) waste nuclides that could be used to identify the occasional pulses of high concentrations of long-lived radionuclide tracers.

Environmental Samples

Millstone Point sampling has shown the following: All ^{55}Fe in the effluent is associated with easily filtrable particles, while only one third of the Pu is similarly associated. This should have led to deposition of these tracers in local sediment accumulations, but we have so far failed to find, and sample, such places. Without a large body of sediment core data for comparison, however, it would have been an unavoidable conclusion, though an incorrect one, that both Millstone Point area cores showed reactor-originated contamination. The only biota samples from this area that clearly show reactor contamination are those taken from the discharge canal. Although much more work should be done, so far no evidence has appeared showing how Millstone Point radioactivity impinges on its local environment.

Maine Yankee occupies a much more restricted, less well flushed, site. We have not yet analyzed any water samples from the area that show evidence of reactor contamination, but sediment samples from all sides of the reactor site prove measurably contaminated. Some of these certainly reflect the conditions before moving and redesign of the waste-stream outflow, but others certainly post-date this change. Both algae

and shellfish from several loci about the area show evidence of reactor wastes. Worms from near the present diffuser have taken up both Cs and Pu radioactivities; comparison of these worms to those near Portland, contaminated only by fallout, suggests that the reactor wastes were measurably different, in chemical-physiological behavior, from the same nuclides from fallout.

Pilgrim 1 occupies a well flushed site, that is near enough to our laboratory so we have sampled quite extensively in its neighborhood. We have found no water samples or sediment samples so far, that exhibit reactor contamination. We have, however, found a good deal of evidence of reactor contamination of biota. In each case these have been collected from north of the reactor along the mainland shore. And in each case there are enough data to show that, locally, contamination is sporadic and relatively quickly lost. In no case were the levels of radionuclides measured high enough to cause concern -- generally, when confirmed, levels approximated those characteristic of fallout contamination in the 1970-71 period. It is, however, evident that with just a little cooperation from the reactor operators, the Plymouth area could be used for an elegant series of environmental experiments that would invaluably clarify our ideas of the biogeochemical fates and rates of movement of long-lived radionuclides of reactor origin. Information would be generated that would support much more detailed, and much more plausible, models than those now available for prediction of the effects of waste spills or of reactor accidents on local marine environments or food supplies.

Appendix Table

Common Names and Taxonomic Details for

Organisms listed in Tables 7, 11, 12, 14A-B, 15

Plants[1]	Common Names
A. Brown Algae	
Fucus vesiculosus	Rock Weed
Ascophyllum nodosum	" "
Laminaria longicruris	Kelp
B. Red Algae	
Chondrus crispus	Irish Moss
Mixture: *Ceramium rubrum* } chiefly	None
Spermothamnion Turner }	"
Phycodrus rubens } some	"
Phyllophora brodiaei }	"
Animals	
A. Annelida[2]	Worms
Nereis virens	Sandworm; Clamworm
B. Molluscs [2]	Shellfish
Bivalves [2]	
Crassostrea virginiana	Oyster
Spisula solidissima	Surf Clam
Mytilus edulis	Blue Mussel
Gastropods [2]	Snails
Lunatia heros	Moon snail
Buccinum undatum	Waved Whelk
Littorina littorea	Common Periwinkle
C. Crustacea[2]	
Balanus balanoides	Barnacle
Homarus americanus	Lobster
D. Echinodermata [2]	
Strongylocentrotus drobachiensis	Green Sea-urchin
E. Birds [3]	
Somateria mollissima	Common Eider duck

Notes: [1] - Taxonomy from Kingsbury, 1969.
[2] - Taxonomy from Smith, 1964.
[3] - Taxonomy from Robbins, et al, 1966.

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16. ABSTRACT (200 words or less)

Release data are reported for three coastal water-cooled nuclear reactors: Millstone Point No. 1 and No. 2 (for the period January 1977 through April 1978), and Maine Yankee (for the period 20 June 1977 through 25 March 1978); release samples were analyzed for ^{55}Fe, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, ^{242}Cm, and ^{244}Cm, but not all nuclides on every sample. Radioiron is a major component of the releases measured; the transuranium nuclides are less significant components than was expected, but levels have occasionally reached microcuries per month. Pulses of this size are adequate for tracer studies.

Environmental samples (water, sediments, and biota) have been analyzed from about the two reactor sites noted, and that of the Pilgrim No. 1 reactor. No water samples remote from reactor outflows have unequivocally shown reactor contamination. No sediment samples from near Millstone Point or Pilgrim 1 have shown reactor contamination; this has been clearly evident in several sediment collections from near Maine Yankee. Biota so far measured from near Millstone Point show reactor contamination only when taken from the effluent canal. From the Maine Yankee and Plymouth areas, however, biota samples frequently prove to show slight, but definite, reactor contamination. In these two areas biogeochemical studies of the fates of long-lived waste radionuclides could easily be carried out, and would be very profitable.

17. KEY WORDS AND DOCUMENT ANALYSIS

17a. DESCRIPTORS: --

water-cooled reactors
reactor effluents
environmental radioactivity
iron-55
cobalt-60
radiocesium
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