

# HSA

Historical Site Assessment

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MYAPC

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**HSA ID# 106**

# **Soil & Sediment Sample History in Vicinity of Maine Yankee**

### Soil Sample History in Vicinity of Maine Yankee Atomic Plant

Year	Location <sup>A</sup>	Depth	Ac-228 pCi/g	Bi-214 pCi/g	K-40 pCi/g	Cs-137 pCi/g	Cs-134 pCi/g	Co-58 pCi/g	Co-60 pCi/g	Mn-54 pCi/g	Reference
1972	Foxbird Island	0-15 cm	1.2	0.7	14	0.94	< 0.03	< 0.025	< 0.03	< 0.02	1
1972	Eaton Farm	0-15 cm	0.34	0.63	14.9	0.87	< 0.015	< 0.012	< 0.015	< 0.01	1
1972	Bailey Farm	0-15 cm	1.24	0.81	14.6	1.67	< 0.03	< 0.025	< 0.03	< 0.02	1
1972	Youngs Creek	0-15 cm	0.88	1.075	18.2	0.8	< 0.03	< 0.025	< 0.03	< 0.02	1
1972	Knight Cemetery	0-15 cm	1.21	0.7	11.2	4.96	< 0.03	< 0.025	< 0.03	< 0.02	1
1972	Westport Firehouse	0-15 cm	1	0.92	11.8	1.11	< 0.03	< 0.025	< 0.03	< 0.02	1
1972	Chewonki Neck	0-15 cm	1.1	1.37	13.2	3.34	< 0.03	< 0.025	< 0.03	< 0.02	1
1972	Cowseagan Narrows	0-15 cm	0.3	0.73	13.4	2.62	< 0.03	< 0.025	< 0.03	< 0.02	1
1972	Bluff Head	0-15 cm	0.66	1.1	11.3	2.03	< 0.03	< 0.025	< 0.03	< 0.02	1
AVERAGE			0.88	0.89	13.62	2.04					
1974	Foxbird Island	0-15 cm	0.9	0.7	7.3	4.6	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Eaton Farm	0-15 cm	1.1	1.1	17.4	2.5	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Bailey Farm	0-15 cm	1.1	1.1	17.6	1.8	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Youngs Creek	0-15 cm	1.1	0.8	19.4	0.7	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Knight Cemetery	0-15 cm	1.7	1.3	11.3	4	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Westport Firehouse	0-15 cm	0.8	0.8	11.8	3.2	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Chewonki Neck	0-15 cm	0.9	0.7	15.1	1.5	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Cowseagan Narrows	0-15 cm	1	1	14.3	1.3	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Bluff Head	0-15 cm	1.2	1.6	13.3	1.2	< 0.035	< 0.025	< 0.03	< 0.015	1
AVERAGE			1.09	1.01	14.17	2.31					

A See attached maps

1 EPA Technical Note ORP/EAD-76-3

**Sediment Sample History in Vicinity of Maine Yankee Atomic Plant**

Date	Location <sup>1</sup>	Depth	Ac-228 pCi/g	Bi-214 pCi/g	K-40 pCi/g	Cs-137 pCi/g	Cs-134 pCi/g	Co-58 pCi/g	Co-60 pCi/g	Mn-54 pCi/g	Reference
1972	Foxbird Island	0-2 cm	0.25	0.5	15	0.35	< 0.015	< 0.012	< 0.015	< 0.01	1
1972	Murphy's Corner	0-2 cm	1.66	0.74	15.2	0.45	< 0.03	< 0.025	< 0.03	< 0.02	1
1974	Foxbird Island	0-2 cm	0.9	0.9	20.5	1	1	21	2.42	0.45	1
1974	Murphy's Corner	0-2 cm	0.9	0.8	18	0.5	< 0.035	< 0.025	< 0.03	< 0.015	1
1974	Bailey Cove Area 1	0-1 cm			7.48	0.19		0.30	0.12		1
1974	Bailey Cove Area 1	0-1 cm			6.36	0.13		0.09	0.07		1
1974	Bailey Cove Area 1	0-1 cm			6.54	0.11		0.18	0.09		1
1974	Bailey Cove Area 1	0-1 cm			5.95	0.09		0.13			1
1974	Bailey Cove Area 1	0-1 cm			5.69	0.12			0.08		1
1974	Bailey Cove Area 1	0-1 cm			6.82	0.16		0.58			1
1974	Bailey Cove Area 1	0-1 cm			6.00	0.19		0.22	0.06		1
1974	Bailey Cove Area 1	0-1 cm			6.38	0.13		0.62	0.05		1
1974	Bailey Cove Area 1	0-1 cm			6.23	0.17		0.42	0.09		1
1974	Bailey Cove Area 1	0-1 cm			5.84	0.14		0.16	0.08		1
1974	Bailey Cove Area 1	0-1 cm			5.17	0.08		0.18			1
1974	Bailey Cove Area 1	0-1 cm			4.84	0.10		0.44			1
1974	Bailey Cove Area 1	0-1 cm			6.69	0.09		0.47			1
1974	Bailey Cove Area 1	0-1 cm			4.20	0.16		0.32	0.06		1
1974	Bailey Cove Area 1	0-1 cm			7.26	0.21		0.24			1
1974	Bailey Cove Area 1	0-1 cm			6.63	0.16		0.79	0.13		1
1974	Bailey Cove Area 1	0-1 cm			7.28	0.24		1.16	0.09		1
1974	Bailey Cove Area 1	0-1 cm			6.54	0.18		0.51	0.10		1
1974	Bailey Cove Area 1	0-1 cm			5.08	0.14		0.65	0.08		1
1974	Bailey Cove Area 1	0-1 cm			7.62	0.64		0.84	0.09		1
1974	Bailey Cove Area 1	0-1 cm			7.34	0.70		0.90	0.09		1
1974	Bailey Cove Area 1	0-1 cm			5.67	0.62		0.47	0.11		1
1974	Bailey Cove Area 1	0-1 cm			6.37			0.22	0.07		1
1974	Bailey Cove Area 1	0-1 cm			7.95	0.42		1.25	0.19		1
1974	Bailey Cove Area 1	0-1 cm			7.46	0.38		0.58	0.08		1
1974	Bailey Cove Area 1	0-1 cm			5.53	0.39		0.63	0.08		1
1974	Bailey Cove Area 1	0-1 cm			5.20	0.27		0.44			1
AVERAGE					6.30	0.24		0.49	0.09		

**Sediment Sample History in Vicinity of Maine Yankee Atomic Plant (continued)**

Date	Location <sup>1</sup>	Depth	Ac-228 pCi/g	Bi-214 pCi/g	K-40 pCi/g	Cs-137 pCi/g	Cs-134 pCi/g	Co-58 pCi/g	Co-60 pCi/g	Mn-54 pCi/g	Reference
1974	Bailey Cove Area 2	0-1 cm			8.23	0.77		6.06	0.53		1
1974	Bailey Cove Area 2	0-1 cm			7.97	0.57		0.26			1
1974	Bailey Cove Area 2	0-1 cm			4.94	0.31		1.30	0.09		1
1974	Bailey Cove Area 2	0-1 cm			5.48	0.21		0.89	0.08		1
1974	Bailey Cove Area 2	0-1 cm			6.46	0.33		2.51	0.15		1
1974	Bailey Cove Area 2	0-1 cm			6.38	0.22		2.05	0.14		1
1974	Bailey Cove Area 2	0-1 cm			6.59	0.18		1.50	0.16		1
1974	Bailey Cove Area 2	0-1 cm			8.44	0.32		1.32	0.11		1
1974	Bailey Cove Area 2	0-1 cm			5.20	0.15		0.88	0.08		1
1974	Bailey Cove Area 2	0-1 cm			6.22	0.34		1.44	0.09		1
1974	Bailey Cove Area 2	0-1 cm			6.61	0.19		0.78	0.11		1
				AVERAGE	6.59	0.33		1.73	0.15		
1974	Bailey Cove Area 3	0-1 cm			6.14	0.29		2.00	0.15		1
1974	Bailey Cove Area 3	0-1 cm			7.86	0.39		2.19	0.15		1
1974	Bailey Cove Area 3	0-1 cm			6.99	0.15		1.21	0.09		1
1974	Bailey Cove Area 3	0-1 cm			5.80	0.32		2.68	0.24		1
1974	Bailey Cove Area 3	0-1 cm			7.71	0.38		1.75	0.12		1
1974	Bailey Cove Area 3	0-1 cm			6.72	0.27		1.24	0.15		1
				AVERAGE	6.87	0.30		1.84	0.15		
1974	Bailey Cove Area 4	0-1 cm			5.59	0.38		1.79	0.15		1
1974	Bailey Cove Area 4	0-1 cm			6.77	0.40		2.36	0.20		1
1974	Bailey Cove Area 4	0-1 cm			7.40	0.14		1.91	0.16		1
1974	Bailey Cove Area 4	0-1 cm			6.60	0.43		1.22	0.13		1
1974	Bailey Cove Area 4	0-1 cm			8.03	0.37		2.07	0.17		1
1974	Bailey Cove Area 4	0-1 cm			7.85	0.29		1.55	0.12		1
				AVERAGE	7.04	0.33		1.82	0.16		

**Sediment Sample History in Vicinity of Maine Yankee Atomic Plant (continued)**

Date	Location <sup>1</sup>	Depth	Pu-238	Pu-239/40	K-40	Cs-137	Cs-134	Co-58	Co-60	Fe-55	Reference
			pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	
1974	Bailey Cove Area 3	0-6 cm	0.0018	0.043		0.40	< 0.009				2
1975	Bailey Cove Area 3	0-6 cm	0.0014	0.032		12.48	7.21				2
1975	Bailey Cove Area 3	0-6 cm	0.0016	0.044		3.72	1.73				2
1975	Back River Area 5	0-6 cm	0.0010	0.028		0.46	0.15				2
1975	Bailey Cove Area 2	0-6 cm	0.0034	0.078		8.65	4.77				2
1977	Bailey Cove Area 3	0-1 cm	0.0014	0.034						3.95	2
1977	Bailey Cove Area 3	1-2 cm	0.00068	0.018						2.08	2
1977	Bailey Cove Area 3	2-3 cm	0.00050	0.014						1.53	2
1977	Bailey Cove Area 3	3-4 cm									2
1977	Bailey Cove Area 3	4-5 cm	0.00018	0.0066						0.30	2
1977	Bailey Cove Area 3	5-6 cm									2
1977	Bailey Cove Area 3	6-7 cm	0.00015	0.0036						0.23	2
1977	Bailey Cove Area 3	7-8 cm									2
1977	Bailey Cove Area 3	8-9 cm	0.000023	0.0015						0.20	2
1977	Bailey Cove Area 3	9-10 cm									2
1977	Bailey Cove Area 3	10-11 cm	0.000032	0.0022						0.15	2
Date	Location <sup>1</sup>	Depth	Ac-228	Bi-214	K-40	Cs-137	Cs-134	Co-58	Co-60	Ag-110m	Reference
			pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	pCi/g	
1993	Back River Area 5	n/a				0.063		0.063	0.066	0.038	3
1993	Back River Area 5	n/a				0.10		<MDA	0.044	0.033	3
1993	Back River Area 5	n/a				0.083		<MDA	0.057	0.023	3
1993	Back River Area 5	n/a				0.12		<MDA	<MDA	<MDA	3
1993	Back River Area 5	n/a				0.040		<MDA	0.026	<MDA	3
1993	Back River Area 5	n/a				<MDA		<MDA	<MDA	<MDA	3
1993	Back River Area 5	n/a				0.24		0.080	0.22	<MDA	3
1993	Back River Area 5	n/a				<MDA		<MDA	<MDA	<MDA	3

**Sediment Sample History in Vicinity of Maine Yankee Atomic Plant (continued)**

Date	Location <sup>1</sup>	Depth	Ac-228 pCi/g	Bi-214 pCi/g	K-40 pCi/g	Cs-137 pCi/g	Cs-134 pCi/g	Co-58 pCi/g	Co-60 pCi/g	Mn-54 pCi/g	Reference
1994	Bailey Cove Area 1	0-5 cm	0.79		19.4	0.30					4
1994	Bailey Cove Area 1	5-10 cm	0.67		17	0.27					4
1994	Bailey Cove Area 1	10-15 cm	0.91		18.03	0.35					4
1994	Bailey Cove Area 2	0-5 cm	0.69		17.02	0.19					4
1994	Bailey Cove Area 2	5-10 cm	0.89		16.49	0.14					4
1994	Bailey Cove Area 2	10-15 cm	0.88		17.15	0.09					4
1994	Bailey Cove Area 3	0-5 cm	0.74		19.7	0.31					4
1994	Bailey Cove Area 3	5-10 cm	0.73		22.6	0.34					4
1994	Bailey Cove Area 3	10-15 cm	0.91		18.77	1.75					4

A See attached maps

1 EPA Technical Note ORP/EAD-76-3

2 NUREG/CR-1658

3 MYC-1647

4 Yankee Atomic - Bolton - Memorandum E. Cumming to P. Anderson, June 24, 1994

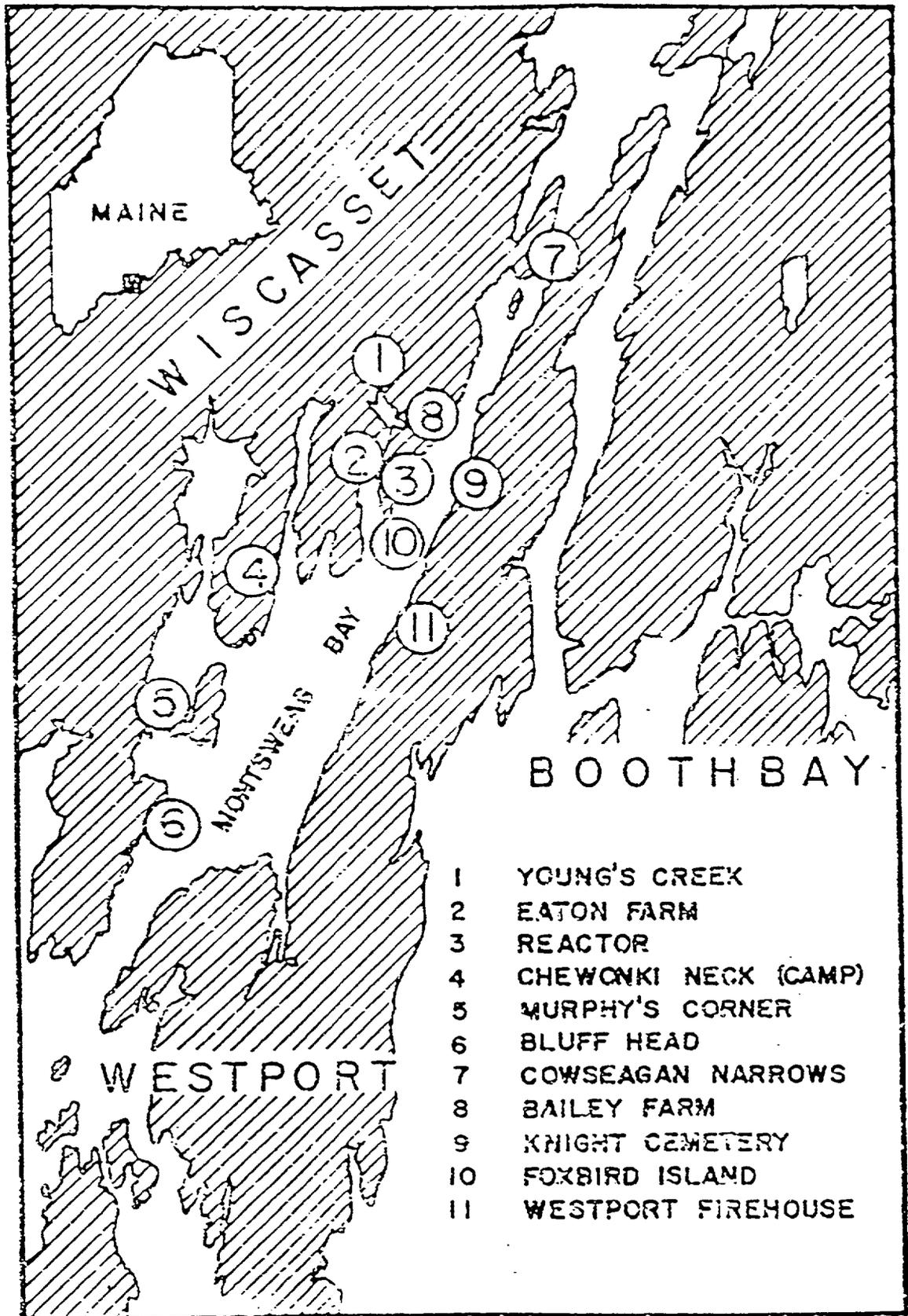
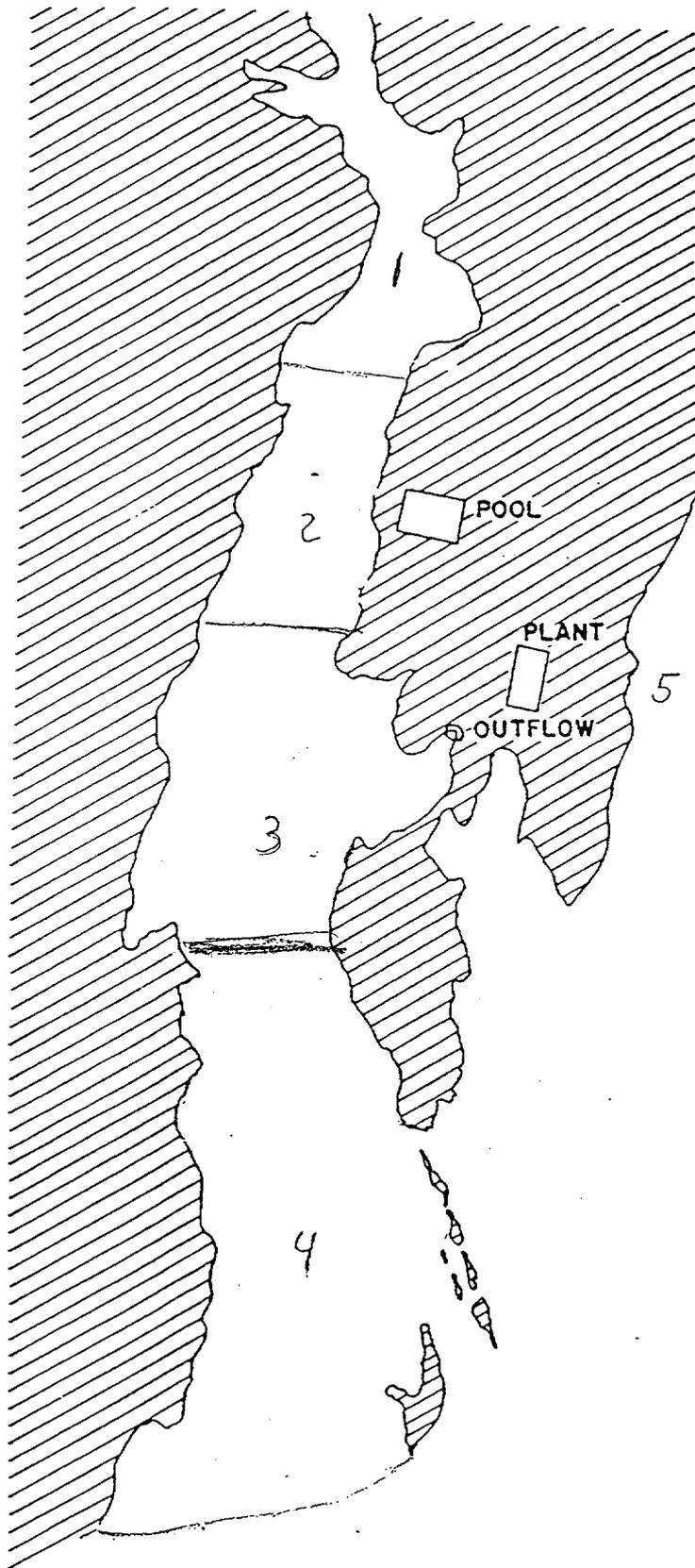


Figure 1. Region near Maine Yankee Atomic Power Plant, Wiscasset, Maine



# PRE OPERATIONAL TRITIUM SAMPLES

Year	Location <sup>^</sup>	Type		H-3 nCi/l		H-3 pCi/l
1972	Foxbird Island	Estuarine	<	0.39	<	390
1972	Eaton Farm	Well	<	0.4	<	400
1972	Bailey Farm	Well	<	0.34	<	340
1972	Youngs Creek	Surface	<	0.14	<	140
1972	Chewonki Neck	Well	<	0.3	<	300
1972	Cowseagan Narrows	Estuarine	<	0.09	<	90
1972	Bluff Head	Estuarine	<	0.4	<	400
				Ave	<	294
				Min	<	90
				Max	<	400

**HSA ID# 107**

Measurement of Radionuclides as a Function of Position in the  
Estuary of the Maine Yankee Atomic Power Plant

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C. T. Hess and A. Price

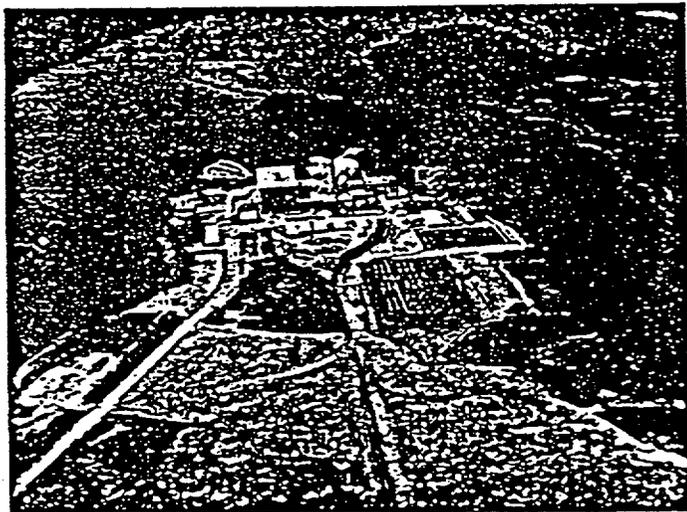
(Presented at American Physical Society Conference,  
New England Sector; April 19, 1975)

## Measurement of Radionuclides as a Function of Position in the Estuary of the Maine Yankee Atomic Power Plant

This paper is primarily a presentation of an experimental survey which was performed on the sediment of the estuary of the Maine Yankee Power Plant in Wiscasset, Maine. The purpose of the survey was to determine the distribution of radionuclides as a function of position in the estuary.

Figure 1 is an aerial photograph of the power plant and its bay estuary. Marked on the figure are the inflow, where the cold water is taken in to condense the steam after it has turned the turbines and the outflow, where the hot water is discharged. It is at the outflow that the radionuclides are introduced into the estuary. Radionuclides are discharged along with the outflow water. Again, it was the purpose of this survey to determine how the radionuclides were distributed after being introduced into the estuary. This was done by taking fifty sediment samples from various points in the estuary. The samples were taken along transect lines, that is, lines perpendicular to the current flow. The samples were gathered in such a way that only the top 1 cm of sediment was collected. Enough sediment was scooped up to fill a 1-liter plastic cylindrical NaI-gener-jar. These jars were labeled according to sample site number and were transported to the University of Maine at Orono, Maine.

Figure 1: Aerial Photograph of the Power Plant and Estuary.



At Orono the samples were measured for gamma-ray emitting nuclides for 5000 seconds using a lead shield and Ge(Li) detector apparatus. The purpose of the lead shield was to block outside radiation so that only the activity of the sample was measured. The Ge(Li) detector is a high resolution, low noise detector. Information from this detector was sent to a multichannel analyzer where the energy spectrum of the sample was recorded. (See Figures 5 and 6 in the back for two typical energy spectra of two samples.) Each spectrum was punched on paper tape which was converted to computer cards which were used as data for our spectrum analysis program. This program identified the nuclides present in each sample and gave their concentration in units of pCi/Kg.

Figure 2 is a drawing of the estuary showing sample locations and numbers. Note again that samples were collected along transect lines which are perpendicular to current flow. Sample numbers 1 through 27 are downstream from the outflow. Sample numbers 28 through 50 are either cross stream or upstream from the outflow. For most samples there were four nuclides of interest which were significantly above background. These were  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{40}\text{K}$ . The first three of these are exclusively man-made products whereas the last one,  $^{40}\text{K}$ , is a natural isotope. Of the man-made isotopes  $^{58}\text{Co}$  and  $^{60}\text{Co}$  are solely reactor products while  $^{137}\text{Cs}$  is a combination of reactor discharge and bomb fallout.

Table I tabulates the amounts of these four isotopes in each of the 50 samples. Looking at the  $^{58}\text{Co}$  column of Table I we see that the concentration below the outflow (site numbers below 28) was generally below 1000, whereas for locations above the outflow, the concentration was generally well above

Figure 2: Locations of sediment sample points-1 in = 650'

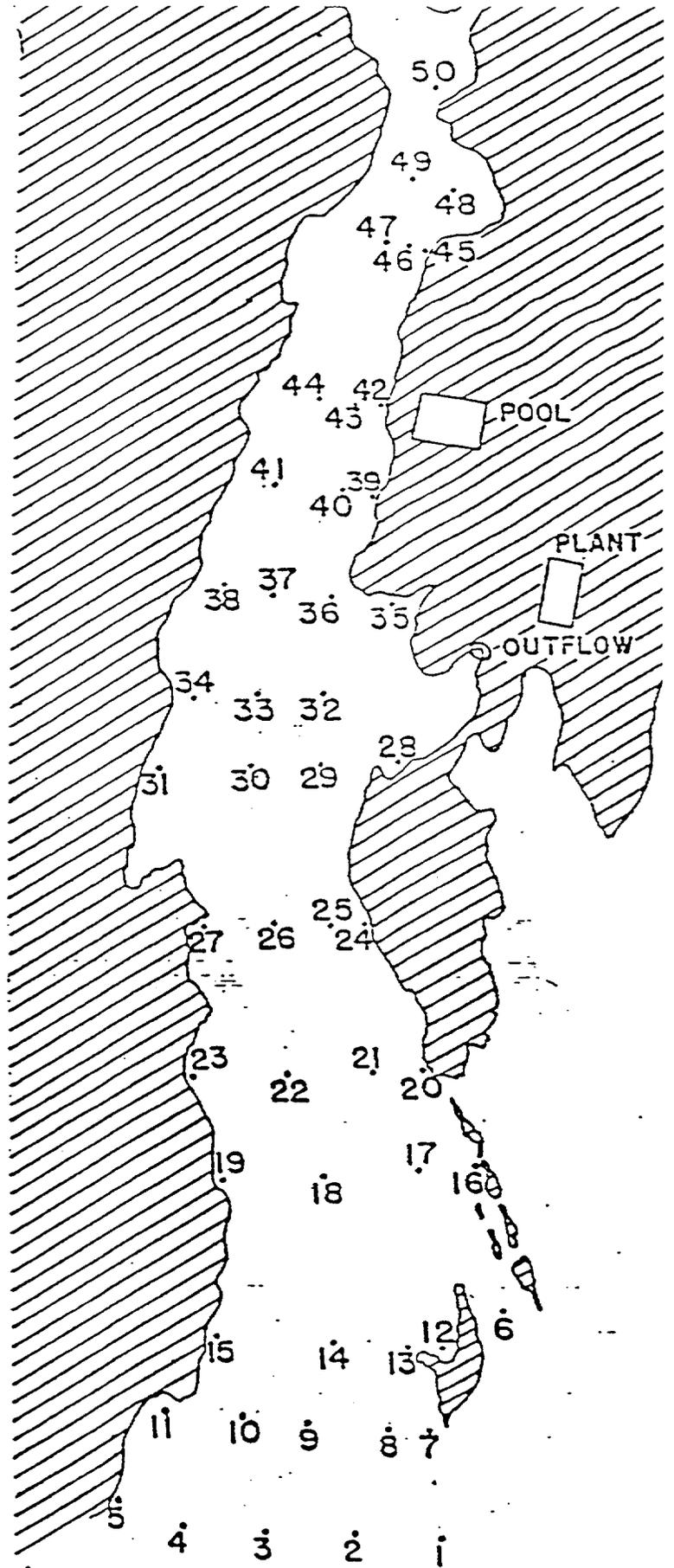


Table I  
 Concentration of Radionuclides per Transect Site Number

Site No.	<sup>58</sup> Co PCi/Kg	<sup>60</sup> Co PCi/Kg	<sup>137</sup> Cs PCi/Kg	<sup>40</sup> K PCi/Kg
1	298	122	191	7478
2	85	67	133	6364
3	175	88	106	6544
4	125	-	87	5953
5	-	77	123	5686
6	579	-	158	6821
7	222	56	191	6004
8	619	49	134	6375
9	422	89	167	6234
10	157	80	144	5844
11	184	-	84	5171
12	441	-	96	4844
13	466	-	92	6694
14	318	63	162	4198
15	236	-	212	7262
16	790	130	158	6634
17	1163	88	239	7278
18	511	98	182	6536
19	645	79	136	5076
20	838	92	641	7623
21	899	92	695	7340
22	472	113	623	5674
23	219	74	-	6369
24	1251	185	420	7951
25	584	81	382	7461
26	634	78	390	5529
27	439	-	268	5197
28	6057	527	769	8228
29	263	-	565	7970
30	1295	91	310	4935
31	893	82	212	5483
32	2506	152	326	6457
33	2050	140	217	6380
34	1500	156	179	6588

Table I (Continued)

Site No.	$^{58}\text{Co}$ PCi/Kg	$^{60}\text{Co}$ PCi/Kg	$^{137}\text{Cs}$ PCi/Kg	$^{40}\text{K}$ PCi/Kg
35	1319	112	318	8441
36	881	83	153	5203
37	1440	90	343	6216
38	783	105	189	6610
39	1996	146	288	6142
40	2192	147	388	7857
41	1210	93	146	6990
42	2679	238	321	5800
43	1750	115	383	7705
44	1236	146	270	6720
45	1791	154	377	5589
46	2356	195	404	6765
47	1906	156	135	7402
48	1224	132	428	6597
49	2072	173	366	8031
50	1546	122	288	7847

1000. Numbers from the  $^{60}\text{Co}$  column display much the same behavior, the concentration was generally higher above the outflow than below the outflow. This means that instead of being flushed out into the ocean which is the desirable effect, much of the radionuclides are backing up with the tide and settling out upstream which is, of course, the undesirable effect.

In order to display these results more pictorially isocuric maps were drawn of the concentrations of  $^{58}\text{Co}$  and  $^{60}\text{Co}$ . An isocuric map is similar to a topographical map that a civil engineer might draw except that the lines correspond to constant radioactive concentration instead of constant altitude.

Figure 3 is an isocuric map for  $^{58}\text{Co}$ . The gradients were so varied that the lines had to be separated logarithmically. Studing Figure 3 one can see that below the outflow the concentration is characterized by peaks and valleys. Near the outflow there is a very sharp peak where the concentration rises to about 6000 PCi/Kg. Adjacent to this peak is a deep valley where the concentration dips to around 300 PCi/Kg. Upstream from the outflow there is a narrow high concentration ridge.

Figure 4 is an isocuric plot for  $^{60}\text{Co}$ . Studing Figure 4 one can see that it shows roughly the same dependence as Figure 3. Below the outflow the concentration is characterized by peaks and valleys. Near the outflow there is a sharp peak beside which there is a deep valley. Above the outflow there is again a narrow high concentration ridge. Note again in both plots that the concentration was much higher above the outflow than below the outflow.

So, it is apparent from these two maps that  $^{58}\text{Co}$  and  $^{60}\text{Co}$  are distributed in much the same way in the estuary. In order to quantitatively see how

Figure 3: Isocuric map of  $^{58}\text{Co}$  concentration - PCi/Kg  
1 in = 600'

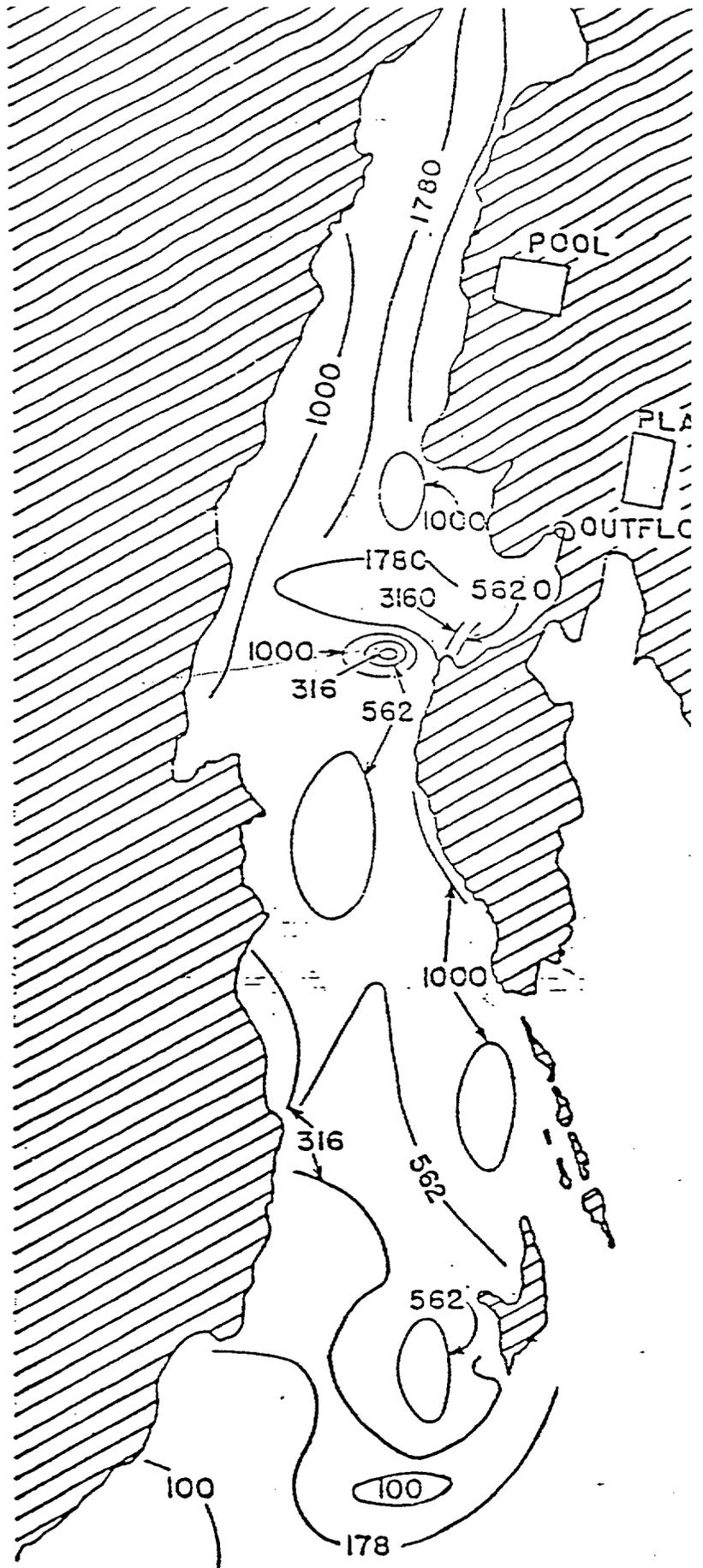
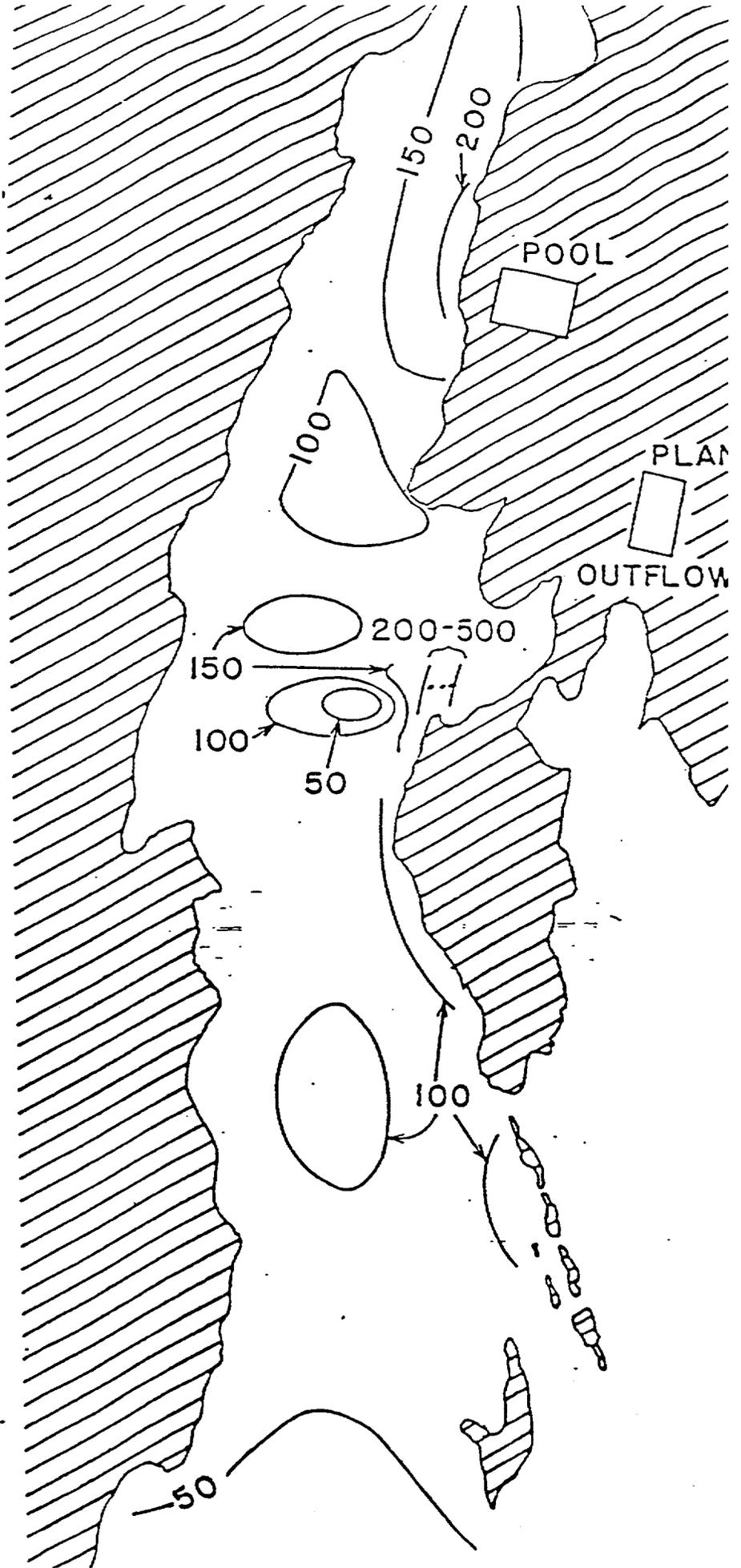


Figure 4: Isocuric map of  $^{60}\text{Co}$  concentration  
PCI/Kg - 1 in = 550'



similar the two distributions were, I applied the correlation coefficient formula to the data in Table I. (In this case, the correlation coefficient can range from 0 to 1, 1 signifying a perfect correlation between two sets of variables and 0 indicating no correlation between two sets of variables.) For the entire estuary the correlation coefficient for the data of  $^{58}\text{Co}$  and  $^{60}\text{Co}$  was .87. In the lower end of the estuary it wasn't so good, it was only .47. However, upstream from the outflow the correlation coefficient was .985. This is exceptionally high. The correlation coefficient between  $^{58}\text{Co}$  and  $^{137}\text{Cs}$  was .503. The coefficient between  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  was .509. These lower coefficients should be expected since  $^{137}\text{Cs}$  is a product of both reactor discharge and bomb fallout and also,  $^{137}\text{Cs}$  has a different chemistry than the Cobalts and should be expected to be distributed differently. The correlation coefficients between  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ; and  $^{40}\text{K}$  are .369 and .364 respectively.

Offhand, one would expect distributions for  $^{58}\text{Co}$  and  $^{60}\text{Co}$  to be similar since they are both solely reactor products and they both have the same chemistry. The paradox arises when one takes into consideration that the half-life of  $^{58}\text{Co}$  is about 70 days whereas the half-life of  $^{60}\text{Co}$  is in excess of 5 years. Thus one should see  $^{58}\text{Co}$  discharged from only the last few months since any release previous to that would have decayed. However, the  $^{60}\text{Co}$  concentration should be a superposition of the  $^{60}\text{Co}$  deposited for the last few years.

Why then are the two distributions so similar? I have some possible explanations for this. One, remember that only the top 1 cm of sediment was gathered. If the sedimentation rate is about 1 cm for every few months this would explain why a superposition of  $^{60}\text{Co}$  for the last few years would not be seen. This would

be a very high sedimentation rate. In the next year we hope to measure the sedimentation rate directly. Also it is not known by what mechanism the radionuclides find their way into the sediment. If this is a reversible mechanism then they should also be able to leave the sediment. If the half-life of this mechanism is about in the ballpark of  $^{58}\text{Co}$  this would also explain why a superposition of  $^{60}\text{Co}$  was not seen.

Early this summer another more extensive sediment survey will be conducted. Also, additional data will be gathered which will be used for a model of the sedimentation of radionuclides.

I would like to express thanks and gratitude to the Darling Center for Oceanography of the University of Maine in Walpole Maine and the Maine Yankee Atomic Power Plant in Wiscasset, Maine for their cooperation in this project.

Figure 5: Energy spectrum of sediment sample # 28 - 2 KeV/channel

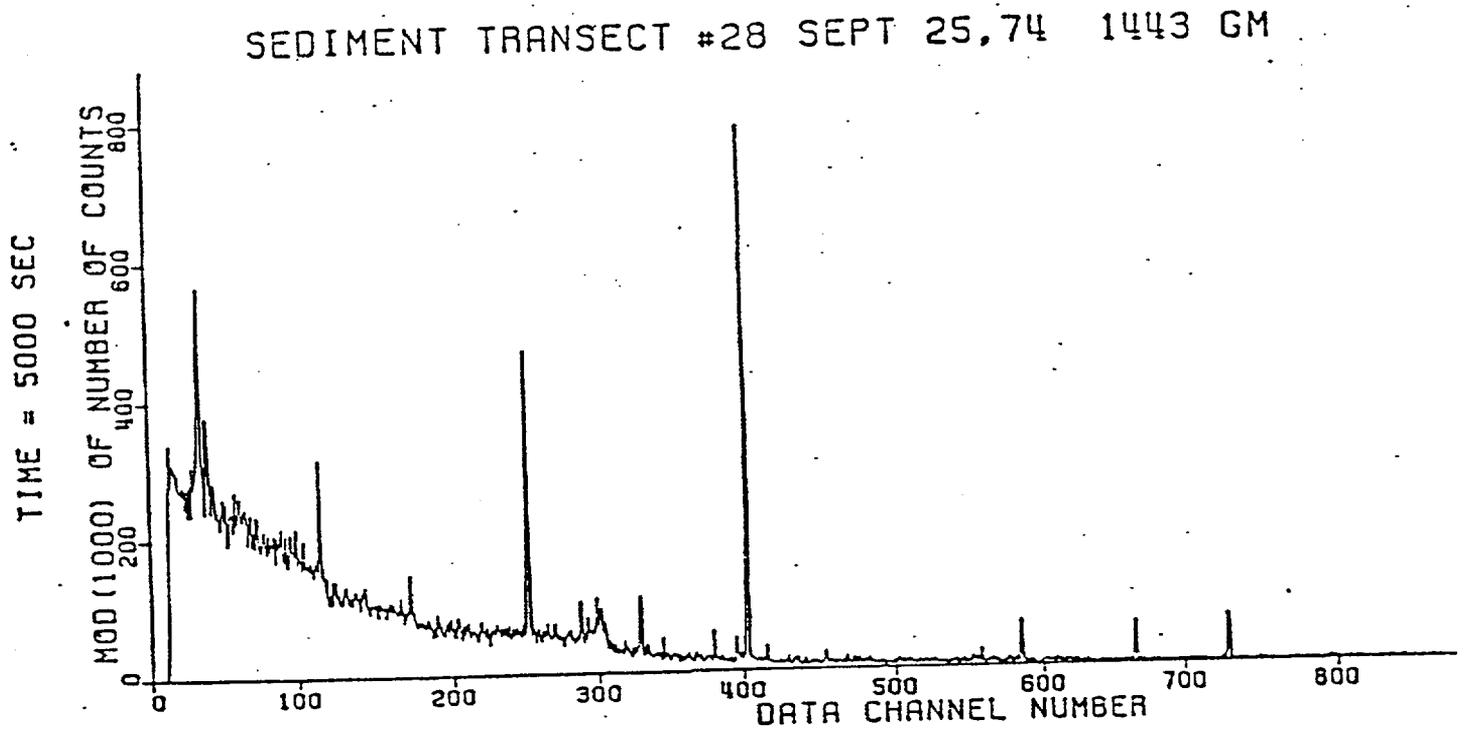
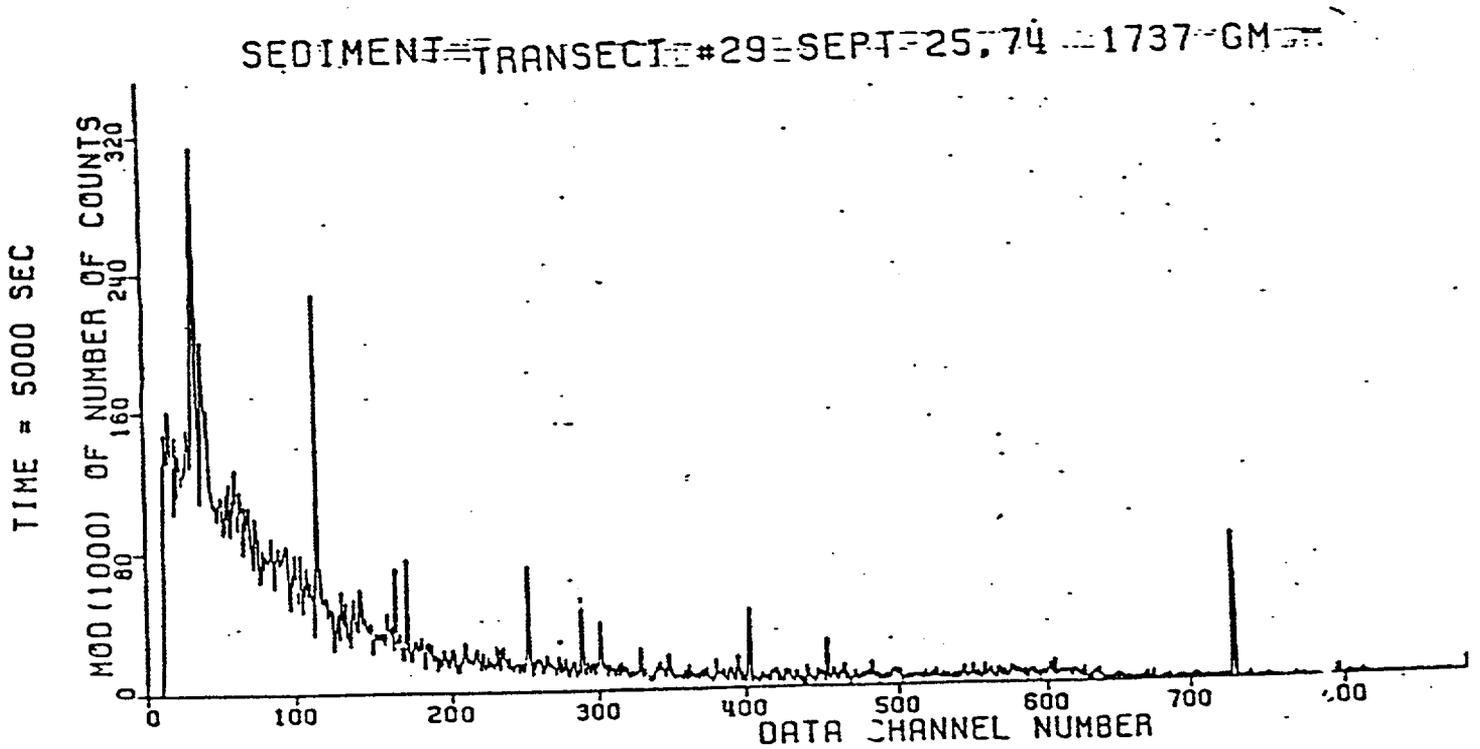


Figure 6: Energy spectrum of sediment sample # 29 - 2 KeV/channel



**HSA ID# 108**

## A Radioactive Isotopic Characterization of the Environment Near Wiscasset, Maine: A Preoperational Survey in the Vicinity of the Maine Yankee Atomic Power Plant<sup>1</sup>

Dr. C. T. Hess, Dr. C. W. Smith, H. A. Kelley, and F. C. Rock<sup>2</sup>

An environmental radioactivity survey was conducted in the vicinity of the Maine Yankee Atomic Power Plant prior to the operation of this facility. Measurements of environmental radioactivity were made on samples of soil, sediment, well water, surface water, estuarine water, air particulates, air moisture, and precipitation.

Natural radioisotopes in the uranium and thorium series and potassium-40 were found in most of the samples. Cesium-137 also was found in some samples.

An environmental radioactivity survey in the vicinity of the Maine Yankee Atomic Power Plant (6.4 km south southwest of Wiscasset, Maine on Bailey Point) was conducted prior to the operation of this facility. The study employed standard surveillance methods as recommended by the Bureau of Radiological Health, U.S. Public Health Service (1). The study was designed to isotopically characterize the radionuclear environment in a quantitative and qualitative manner, establish a meaningful and useful background baseline to which data from future radiological studies can be compared, document average values for the radiation levels from various environmental media, and locate and identify any regions of "other than average character" or contributions to the background by "manmade" radionuclides.

Measurements of the radioactivity in the environment were made on soil, sediment, well water, surface water, estuarine water, air particulate, air moisture, and precipitation in the form of snow and rain.

### *Study design*

The basic approach used in the study design was to employ modern standard methods of measuring environmental radioactivity in typical environmental media with emphasis on measuring individual isotopes. The study was divided into two parts. Field measurements were made employing a portable multichannel analyzer system. This system, along with appropriate support equipment, was used in the field to measure various environmental media in situ (2-5). Laboratory measurements on collected samples were carried out at the Northeastern Radiological Health Laboratory (NERHL), Winchester, Mass. This dual approach exercised a variety of standard surveillance techniques.

The scope of this survey encompassed the three zones of environmental media, terrestrial (soil and sediment), aquatic-estuarine (well water, surface water and estuarine water) and air (air particulate, air moisture and precipitation in the form of snow and rain). Sample locations were chosen in general to be as natural and undisturbed as possible, typical of the media they represent and reproducible in the sense that measurements can be repeated in the future at the sites used in this study (6). The study was 8 weeks in duration beginning June

<sup>1</sup>This work was carried out under Environmental Protection Agency Contract Number 83-01-5711.  
<sup>2</sup>Department of Physics, University of Maine, Orono, Maine 04473.

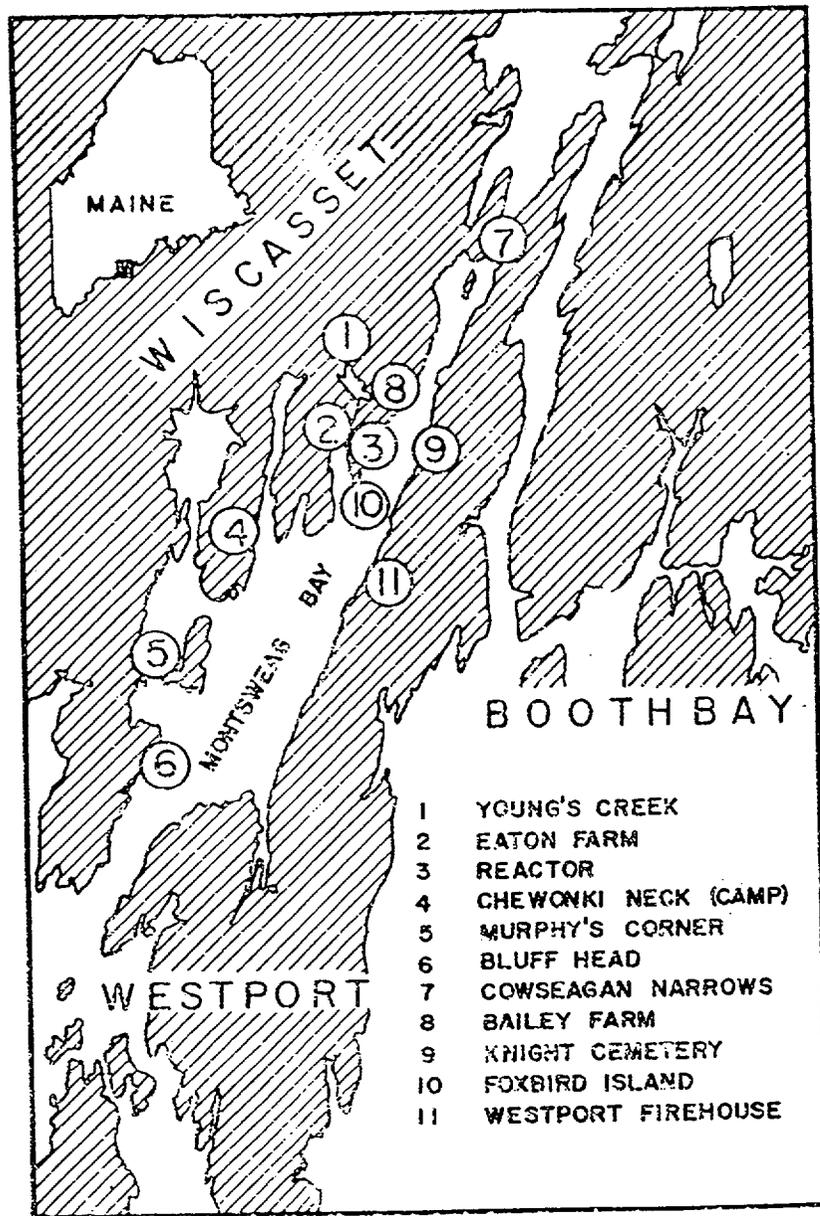


Figure 1. Region near Maine Yankee Atomic Power Plant, Wiscasset, Maine

Eaton Farm, Bailey Farm, Young's Creek, Knight Cemetery, Westport Firehouse, Chewonki Neck (camp), Cowseagan Narrows, and Bluff Head. Sediment samples were collected at Foxbird Island on the outfall side of the causeway and at Murphy's Corner. Soil samples were collected using a disc-cutter sampler which would cut a cylindrical soil sample, 15 cm in diameter by 15 cm deep. Four soil samples this size, each

centered at the corner of a square grid, 25 cm on an edge, were collected at each of the nine sites. The four soil samples including surface vegetation were mixed and the root mats pulverized. The sample was then screened and all material smaller than 5 mm retained. The sample was then dried at 150°C. for 24 hours. In the case of the sediment samples, the top 2 centimeters were collected. These samples were screened and dried in an identical manner. In

ments for potassium-40 and for cesium-137, respectively, would be in an order of magnitude agreement. This is also clear from these tables. However, because the field measurements and the laboratory measurements are quite different (instrumentation, calibration, and sample character) one would not expect exact numerical agreement. Still we can see that for relative rank by activity, both measurements indicate Eaton Farm and Bailey Farm to be higher in potassium-40 with Westport Firehouse being significantly lower, and both measurements indicate Knight Cemetery and Bluff Head to be higher in cesium-137 with Eaton Farm and Young's Creek lower. Because of the quite different calibration procedures used in these two measurements (see appendix A) one cannot directly compare the activities in the two naturally occurring decay series. Nevertheless, in the case of relative rank by activity, both measurements indicate (using actinium-228 as the indicator) higher activity at the Knight Cemetery and Bailey Farm sites for the thorium-232 series, with a significantly lower activity at Young's Creek. In the case of the uranium-238 series (using radium-226 as the indicator), the Eaton Farm and Bluff Head sites are significantly higher than Murphy's Corner. Considering the layering nature of the field samples and the homogeneous nature of the laboratory samples, one cannot expect a much higher correlation than observed.

Soil and sediment classification and analysis can be found in appendix B. Classification was carried out in the field by a soil specialist from the Plants and Soils Department of the University of Maine. The same department did the laboratory analyses for pH and the exchangeable components of phosphorus, potassium, calcium, and magnesium (acetic acid extraction method). Based on soil chemistry, the measurements verify the expectation that there is a fairly consistent relationship exists between the exchangeable component of potassium in the soil and the potassium-40 activity, the magnesium in the soil and the radium-226 activity, and the phosphorus in the soil and the bismuth-214 activity. Good agreement also exists between the potassium in the soil and the soil type and the cesium in the soil and the depth of the organic

mat (layers  $O_1$  and  $O_2$ , see appendix B). From this, one concludes that the soil chemistry is a main feature in determining the retention capabilities in these soils for the isotopes mentioned and that the organic layers may play a dominant role in the uptake and retention of cesium.

## RADIONUCLIDES IN THE WATER

### *Water field measurements*

Radionuclides in the water were measured at Foxbird Island, Young's Creek, Cowseagan Narrows, and Bluff Head (figure 1). The portable multichannel analyzer system and support equipment used for the soil field measurements was used for these water measurements. The only modification necessary was to float the shield-detector assembly in a canoe over the water being measured. At all times, a minimum depth of 1 meter was maintained below the detector during measurement. All water field measurements were taken on half memory (512 channels) for 4 000 seconds. At the end of two runs, the multichannel analyzer was returned to the Maine Yankee Environmental Studies Center at Bailey Point and the memory outputted into the teletype. The water field measurements were only qualitatively analyzed because for reasonable in-the-field counting times (on the order of an hour), the count rate was so low that the gamma-ray photopeaks for nominal environmental concentrations were substantially masked by the counting statistics. However, several general observations can be drawn from the field data. The major contributors to the spectra were potassium-40 and the daughters of radon-222. The potassium concentration is the same for Foxbird Island, Cowseagan Narrows, and Bluff Head and is indicative of the common salinity of these estuarine samples (it corresponds to about 10 lps/liter). The potassium-40 contribution to the Young's Creek spectrum is essentially zero, as expected. The bulk of the rest of the spectra is made up of the daughters of radon-222 with the exception of the Bluff Head sample, which contained very little radioactivity. In general, the water samples were a full order of magnitude less active than the soil field measurements.

two groups (Bailey Farm and Eaton Farm on 6/30/72 and Chewonki Neck (camp) and Westport Firehouse on 7/3/72) separated by 72 hours and an intervening cold-front rain shower, the data are essentially identical. The exhalation rate of radon from the soils in this region is not documented, the attachment of decay products on natural aerosols is a function of the natural aerosol size distribution and therefore, this size distribution was not measured, and the filter efficiency of the fiberglass filters as a function of the aerosol size distribution is not known.

#### *Air particulate laboratory measurements*

Air particulates for laboratory analysis of radionuclides were collected at three sites (Eaton Farm, Bailey Farm, and Westport Firehouse). A Millipore pump type-xx600000 was used to pull air through a 47 mm diameter Millipore absolute aerosol 0.8  $\mu\text{m}$  filter, type AAWP04700. The throughput of this system was metered for all runs with a calibrated gas volume flow meter, American Meter Company Model 10-300-PR1264. This meter was capable of reading up to 99 999.9 liters. The meter, pump, and filter holder were housed in an instrument box with a rubber hose from the filter holder to the outside. A nominal sampling time was 50 hours with a nominal sampling volume being on the order of 100 000 liters. The filters were weighed before and after collection. They were packaged carefully in separate plastic boxes and sent to the Northeastern Radiological Health Laboratory, Winchester, Mass. for gamma-ray analysis. Each sample was prepared for analysis by digestion of the filter and contents in hydrofluoric acid. Water was then added to make the total volume of the prepared sample 200 milliliters. The samples were counted using the same Ge(Li) detector system used for the laboratory soil and sediment measurements, and using a 250 milliliter geometry. The amount of particulate collected on each filter was nominally a few milligrams. The Eaton Farm filter was counted for 3840 minutes, and the Bailey Farm and Westport Firehouse filters were counted for 900 minutes each. Full spectral analysis of all three samples indi-

cated no peaks statistically above background levels. Background peaks were observed at 0.077 MeV, 0.094 MeV, 0.351 MeV (lead-214), 0.510 MeV (thallium-203), 0.603 MeV (bismuth-214), 0.910 MeV (actinium-228), and 1.459 MeV (potassium-40). This would indicate that the activities for these isotopes (for 100 000 liters of air) are below their respective minimum detectable levels (15 pCi for lead-214, 85 pCi for thallium-203, 15 pCi for bismuth-214, 100 pCi for actinium-228, and 100 pCi for potassium-40).

#### *Discussion of the air particulate results*

For the standard measurements employed in this section and for the nominal count times and collection techniques, the bulk of the activity is attributed to the daughters of radon-222. Careful analysis of the gamma-ray peaks for lead-212 (0.239 MeV and 0.300 MeV) and lead-214 (0.295 MeV, 0.242 MeV and 0.352 MeV), daughters of radon-220 and radon-222, respectively, indicates the presence of radon-220. The relative abundance of radon-220 to radon-222 shows a fairly good correlation with the ratio of the activity in the thorium-232 series to the activity in the uranium-238 series as indicated by the field measurements of soil. This ratio was larger for Westport Firehouse and significantly lower for Bailey Farm (the Westport Firehouse site showed evidence of radon-220).

The laboratory air particulate measurements were analyzed for the uranium-238 series, the thorium-232 series, cesium-137, potassium-40, zirconium-niobium-95 and manganese-54. None of these isotopes was found at levels above its respective minimum detectable amount. Apparently, for low efficiency detectors like Ge(Li) to yield statistically meaningful data for air particulate measurements, larger particulate mass must be accumulated. Using the minimum detectable levels for the laboratory soil measurements, one must scale the air particulate data by at least a factor of 10 000. This would require the filtering of  $10^6$  liters of air having the relatively low particulate content found in this study in order to bring the activities of the various isotopes above their respective minimum detectable levels.

## TRITIUM IN WATER, AIR MOISTURE, AND PRECIPITATION

### *Tritium in water*

Tritium in water was measured in samples collected at seven locations, table 5. Each water sample was filtered as described earlier. A 50 milliliter sample from each site was sent to NERHL for tritium analysis using the direct-counting liquid scintillation method (9, 10). (The water samples were processed to remove impurities that cause quenching and a scintillation fluor was added.) The samples were counted in a low background chamber. The results of these measurements are presented in table 5.

Table 5. Tritium in water

Location and distance from reactor site	Date collected (1972)	Type	Measured results (nCi/liter $\pm 2\sigma$ )	Interpreted results*
Foxbird Island, 0.1 km S	6/13	Estuarine	0.39 $\pm$ 0.2	zero
Eaton Farm, 0.4 km W	6/13	Well	.40 $\pm$ .2	zero
Bailey Farm, 0.8 km NE	6/13	Well	.34 $\pm$ .2	zero
Young's Creek, 1.0 km N	6/13	Surface	.14 $\pm$ .2	zero
Cheewahki Neck (camp) 1.9 km SW	6/13	Well	.30 $\pm$ .2	zero
Cowesagan Narrows, 3.2 km NE	6/13	Estuarine	.09 $\pm$ .2	zero
Bluff Head, 4.0 km SSW	6/13	Estuarine	.40 $\pm$ .2	zero

\* The minimum detectable level for the analysis of tritium by the direct liquid scintillation counting method used in this study is 0.4 nCi/liter. All values equal to or less than 0.4 nCi/liter will be reported by convention as zero. Values greater than 0.4 nCi/liter but which round to this value will be reported as 0.4 nCi/liter.

### *Tritium in air moisture*

Tritium in air moisture was measured in samples collected at Eaton Farm, Bailey Farm, and Westport Firehouse. Air moisture samples were collected by drawing air through a plastic cylinder (8-cm diameter by 24-cm long) containing a desiccant (11). The desiccant used was indicator type Dryrite. The cylinder of desiccant was weighed before the air was pulled through and weighed afterward to insure the collection of between 20 and 30 milliliters of air moisture. Nominal collection time was between 2 and 4 hours depending on the relative humidity. The desiccant cylinders were sealed and sent to NERHL for tritium analysis. The water was thermally driven from the desiccant in a closed system. The samples were processed for tritium

analysis in the usual way. The results of these measurements are listed in table 6.

Table 6. Tritium in air moisture

Location and distance from reactor site	Date collected (1972)	Quantity measured (ml)	Measured results (nCi/liter $\pm 2\sigma$ )	Interpreted results (nCi/liter)*
Eaton Farm, 0.4 km W	6/29	33.7	0.60 $\pm$ 0.2	0.6
Bailey Farm, 0.8 km NE	6/15	33.1	19.1 $\pm$ .2	19.1
Westport Firehouse, 1.8 km S	6/27	33.4	.60 $\pm$ .2	.6

\* The minimum detectable level for the analysis of tritium by the direct liquid scintillation counting method used in this study is 0.4 nCi/liter. All values equal to or less than 0.4 nCi/liter will be reported by convention as zero. Values greater than 0.4 nCi/liter but which round to this value will be reported as 0.4 nCi/liter.

### *Tritium in precipitation*

Tritium in precipitation was measured in snow samples collected at Knight Cemetery and Westport Firehouse on March 11, 1972 and rain samples collected at Eaton Farm and Bailey Farm on June 30, 1972. These samples were filtered in the same manner used for the water samples. Fifty milliliter samples were sent to NERHL for tritium analysis using the liquid scintillation method. The results of the analysis are listed in table 7.

Table 7. Tritium in precipitation

Location and distance from reactor site	Date collected (1972)	Type	Measured results (nCi/liter $\pm 2\sigma$ )	Interpreted results (nCi/liter)*
Eaton Farm, 0.4 km W	6/30	Rain	0.40 $\pm$ 0.2	zero
Bailey Farm, 0.8 km NE	6/30	Rain	1.10 $\pm$ .2	1.1
Knight Cemetery, 1.1 km E	3/11	Snow	.26 $\pm$ .2	zero
Westport Firehouse, 1.8 km S	3/11	Snow	.17 $\pm$ .2	zero

\* The minimum detectable level for the analysis of tritium by the direct liquid scintillation counting method used in this study is 0.4 nCi/liter. All values equal to or less than 0.4 nCi/liter will be reported by convention as zero. Values greater than 0.4 nCi/liter but which round to this value will be reported as 0.4 nCi/liter.

### *Discussion of tritium measurements*

Tritium has a half-life of 12.5 years. It is produced naturally by cosmic radiation and artificially by nuclear weapons and nuclear reactors. The nuclear weapons tests of the early fifties and subsequent tests have generated considerable amounts of tritium so that now most of the tritium found in the interchangeable environmental reservoirs is manmade tritium and

## APPENDIX A

### *Calibration method for the field measurements*

The field measurements in this study were computer analyzed using a least squares method. The computer program employed was developed at the Oak Ridge National Laboratory and is a gamma-ray spectrum fitting program known as ALPHA-M. This program uses a library of standard gamma-ray spectra, taken with the same detector and shield geometry as was used for the sample spectra being analyzed. For these measurements, spectra for potassium-40, cesium-137, the thorium-232 series and the uranium-238 series were used. The top shielded  $2\pi$  geometry was simulated by uniformly distributing a known amount of radionuclide in an adequately large cylindrical volume of washed quartz sand. The size of this cylinder was determined experimentally using the natural abundance of potassium-40 in potassium chloride. Potassium-40 was chosen because the gamma ray from this radionuclide falls at the energy for minimum absorption for sand (13). Thus, an adequately large volume (a volume for which the addition of further material does not increase the observed activity) as determined in this manner will be adequate for all other gamma-ray energies, both larger and smaller than that for potassium-40 (1.459 MeV). The volume used was 16.8 cm high by 105 cm in diameter (the mass of dry washed sand contained in this volume is 25.45 kilograms). In order to insure that the radionuclide was dispersed uniformly throughout the sand and in order to further simulate the absorption coefficient of soil, the cylinder of dry sand was saturated with 5 liters of a water solution of the

radionuclide. The standard spectra were then measured using the same detector, shield, cable and multichannel analyzer that were used for the field measurements. The detector-shield assembly was placed on the cylinder of the water-saturated sand in the same manner as used for the field measurements. All spectra were corrected for background activity by using a sand-and-water-only sample.

Calculation of the activity of each standard was carried out knowing the amount of radionuclide dispersed in the cylindrical volume and its branching ratio. The potassium-40 standard was calculated based upon the natural abundance of potassium-40 in natural potassium. Potassium chloride was used as the host. The cesium-137 standard was calculated based upon the known activity of a sample of cesium-137 provided by the Analytical Quality Control Service of the Environmental Protection Agency. The thorium-232 series standard was calculated based upon a sample of thorium nitrate known to be in secular equilibrium. This sample was 25 years old or older and its spectrum was calibrated in the usual manner. The uranium-238 series standard was calculated based upon a sample of uranium ore. The spectrum of the uranium ore was calibrated in the usual manner.

This calibration method was designed to simulate the top shielded  $2\pi$  geometry and to take into account the absorption of gamma rays in the soil. It does not take into account the variations in the soil due to layering. These variations are primarily variations in radionuclide composition from layer to layer, with gamma-ray absorption gradations usually being a secondary consideration.

## APPENDIX C

### *Discussion of error analysis used in the Compton continuum subtraction method*

The Compton continuum subtraction method was used for the quantitative analysis of all laboratory measurements employing the Ge(Li) detector system. The measurements involved were soil, sediment, undissolved solids in water and air particulates, as well as the counting efficiency of the detector itself and the measurement of the background for these laboratory measurements.

For the Compton continuum subtraction method, once the photopeak was located, the most reproducible procedure is to use the peak channel and an equal number of channels on either side of that channel. We chose to use the photopeak channel and five channels on each side. A line estimating the base of the photopeak under consideration is then constructed. There are two numbers involved in the analysis: the sum of the 11 channels encompassing the photopeak, and the sum of the 11 values making up the baseline. We now associate with each of these numbers an uncertainty. The statistical analysis of random variables tells us in this case that each individual value  $N$  has associated with it a probable absolute error equal to  $\sqrt{N}$ . Calling the number of counts in the 11 channels

of the baseline  $B$ , the area of the photopeak will be  $P-B$ . The absolute error associated with this area will be  $\sqrt{P} + \sqrt{B}$  since the absolute error in an addition or subtraction is equal to the sum of the absolute errors in the input quantities. We find reported in tables 1, 2, 4, and appendix D values with errors. These values and errors are based upon the photopeak area and associated absolute error, that is  $(P-B) \pm (\sqrt{P} + \sqrt{B})$ .

When the photopeak area  $(P-B)$  is less than  $\sqrt{P} + \sqrt{B}$  we say that the amount of the particular isotope involved is less than the minimum detectable level. The minimum detectable level for a particular isotope is based upon the absolute error  $\sqrt{P} + \sqrt{B}$ , the detector efficiency calibration, and the appropriate branching ratio for that radionuclide. There are of course complicating circumstances for which this straightforward procedure would have to be modified. These more complicated cases were handled on an individual basis. The most frequent complication confronted in this study was cases in which the branching ratio was known to less accuracy than the statistical limits placed on the photopeak area. The area in these cases, dominated by the branching ratio information, was calculated based on the relative error in the product of the numbers involved as well as the statistical limitations on these numbers.

**HSA ID# 109**

TECHNICAL NOTE  
ORP/EAD-76-3

**RADIOACTIVE ISOTOPIC CHARACTERIZATION OF THE  
ENVIRONMENT NEAR WISCASSET, MAINE USING PRE  
AND POST-OPERATIONAL SURVEYS IN THE VICINITY OF  
THE MAINE YANKEE NUCLEAR REACTOR**



**U.S. ENVIRONMENTAL PROTECTION AGENCY**  
**Office of Radiation Programs**

**RADIOACTIVE ISOTOPIC CHARACTERIZATION OF THE ENVIRONMENT NEAR  
WISCASSET, MAINE USING PRE- AND POST-OPERATIONAL SURVEYS IN THE  
VICINITY OF THE MAINE YANKEE NUCLEAR REACTOR.**

**MAY 1976**

**ENVIRONMENTAL ANALYSIS DIVISION  
OFFICE OF RADIATION PROGRAMS  
U.S. ENVIRONMENTAL PROTECTION AGENCY  
WASHINGTON, D.C. 20460**

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OF DR. C. T. HESS, DR. C. W. SMITH, C. H. CHURCHILL AND G. F. BURKE.**

PREFACE

The Office of Radiation Programs is concerned with the evaluation of radiation exposure to man and his environs. Nuclear power plants release radioactive materials to the environment from normal operations which become a potential source of exposure to the population. The Environmental Analysis Division has responsibilities for evaluating the environmental and public health impacts resulting from such releases.

This study was performed on contract to this Division by the University of Maine for the purpose of isotopic characterization of environmental radioactivity outside the plant site boundary of the Maine Yankee Pressurized Water Nuclear Power Reactor during 1973. It compares the pre-operational survey with the postoperational survey to determine the amount of contamination resulting from operations of this power reactor.

*Floyd L. Galpin*

Floyd L. Galpin  
Director  
Environmental Analysis Division  
Office of Radiation Programs

RADIOACTIVE ISOTOPIC CHARACTERIZATION OF THE ENVIRONMENT  
NEAR WISCASSET, MAINE USING PRE- AND POST-OPERATIONAL SURVEYS  
IN THE VICINITY OF THE MAINE YANKEE NUCLEAR REACTOR

ABSTRACT

A comparison of identical surveys of the pre- and post-operational environmental radioactivity is made for the vicinity of the Maine Yankee Atomic Power Reactor, Wiscasset, Maine. Radionuclides are measured in laboratory samples of soil, sediment, well water, surface water, estuarine water, air particulate, air moisture, and precipitation. Field measurements of gamma-ray emitting radionuclides and high pressure ion chamber measurements are also presented. The changes in radionuclide concentration and dose are evaluated using the Maine Yankee Environmental Impact Statement. The most significant changes occur for radionuclides in soils and sediments. Details of the distribution of sediment radionuclides near the outflow of the reactor in Bailey Cove are presented with dose estimates of 10.6 mrem/year at the sites of greatest specific activity. Radionuclides in the water were mainly natural  $^{222}\text{Rn}$  and daughters. Tritium concentrations were at minimum detectable level. Air particulate showed traces of  $^7\text{Be}$  and  $^{95}\text{ZrNb}$  at fallout levels which were not significantly different from the preoperational levels. High pressure ion chamber dose rates ranged from 8.9 to 12.6  $\mu\text{R/hr}$  at the 12 measured sites.

Time variations of radionuclide content were measured for oysters cultured in the reactor effluent and associated sediments. Doses calculated for ingestion of radionuclides by consumption of oysters would be 0.27 mrem/yr for  $^{58}\text{Co}$  and 0.004 mrem/yr for  $^{54}\text{Mn}$ .

## FORWARD

This report presents the procedures and results of an environmental radioactivity survey in the vicinity of the Maine Yankee Atomic Power Plant (a 855 MWE pressurized water reactor) 6.4 km south southwest of Wiscasset, Maine on Bailey Point. Prior to the operation of this facility measurements of environmental radioactivity were made (1972) on samples of soil, sediment, well water, surface water, estuarine water, air particulate, air moisture, and precipitation. The procedures and results of that survey were reported in "Radiation Data and Reports", volume 15, number 2, February 1974.

The post-operational survey (1974) represents the follow-up survey to the pre-operational survey referenced above. The study design employs the same techniques, environmental media, and sampling sites as the pre-operational survey. The objective of this dual survey is to assess changes in environmental radioactivity.

## ACKNOWLEDGMENTS

The work reported here represents a cooperative effort by several institutions and facilities. Essential to this study was the work of the staff of the Eastern Environmental Radiation Facility, Montgomery, Alabama for off-site laboratory measurements, with special thanks to Mrs. Ann B. Strong, Mr. Charles Phillips and Mr. Thomas Reavey. We gratefully acknowledge the participation by the Ira C. Darling Center for Oceanographic Research, University of Maine in the estuarine aspects of this study, the transect study of Bailey Cove and the aquaculture of the oysters for the uptake modeling, with special thanks to Mr. A. H. Price and Dr. H. Hidu. Cooperation and assistance by the Maine Yankee Atomic Electric Company, with special thanks to Mr. V. Thompson, was gratefully appreciated. We acknowledge the use and cooperation of the University of Maine Physics Department, Computer Center and Office of Grant Support. The information included in this report as Appendix D was part of a study funded by the National Oceanic and Atmospheric Administration. We appreciate their willingness to have this material included in this report.

We are especially grateful for the useful discussions throughout this study with Mr. C. L. Weaver of EPA's Office of Radiation Programs and for the assistance of Mr. Charles Robbins of the Environmental Analysis Division who coordinated the publication of this report.

Dr. C. T. Hess  
Dr. C. W. Smith

University of Maine

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## 1. INTRODUCTION

### 1.1 Purpose of the Study

This study consists of a pre-operational (1972) and post-operational (1974) environmental radioactivity survey in the vicinity of the Maine Yankee Nuclear Power Reactor (6.4 km S.S.W. of Wiscasset, Maine on Bailey Point, See Fig. 1). The study measures any changes in the radionuclide content of the soil, estuarine sediment, estuary, well, and surface water, air moisture precipitation, and air particulate for this region. The study identifies the pathways in these environmental media which tend to collect or reconcentrate radionuclides and the regions in which reconcentration occurs.

### 1.2 Goals for this Study

A. To broadly survey several environmental media (soil, sediment, water, air moisture and air particulate) for changes in radionuclide content (nuclide by nuclide rather than gross activity) resulting from the operation and refueling of the Maine Yankee Nuclear Power Reactor.

B. To compare observed changes with the Maine Yankee Semi-annual Report of Release of Radioactive Materials<sup>(1)</sup> and with predictions of the Maine Yankee Environmental Impact Statement<sup>(2)</sup>.

C. To examine in detail the largest change observed in the study.

### 1.3 Study Design

Studies of pressurized water reactors as outlined by Kahn<sup>(3)</sup>, Rowe<sup>(4)</sup> and Lentsch<sup>(5)</sup> et.al. are based on a combination of knowledge of radionuclide releases, use of very sensitive environmental measurements and a carefully planned pre-operational survey. This study similarly employs these features. A nuclide by nuclide comparison of a pre-operational and post-operational survey of several environmental media forms the overall structure of this study. Field and laboratory measurements of primarily gamma-ray emitting radionuclides were used to determine which of the radionuclides listed in the Semi-annual Report of Release of Radioactive Materials<sup>(1)</sup> are being retained in the environment in the vicinity of the reactor. Tritium, gross beta and gross alpha measurements, along with high pressure ion chamber measurements also were employed. The major radionuclide in the liquid effluent is tritium and the major radionuclides in the gaseous effluent is  $^{133}\text{Xe}$  and tritium. Several other radionuclides in trace amounts (See Appendix A) are also released in the liquid and gaseous effluents.

To achieve both the sensitivity and diversity of measurements to cover this broad survey this study utilized the facilities of the Eastern Environmental Radiation Facility, Montgomery, Alabama for laboratory measurements for the post-operational survey, the Northeastern Radiological Health Laboratory, Woburn, Massachusetts for laboratory measurements for the pre-operations survey and the University of Maine, Department of Physics for the field measurements and some laboratory measurements for both surveys<sup>(6,7)</sup> The two surveys employ the same sampling sites and as far as possible the same or similar instrumentation. Details of each measurement are presented

in the following sections along with the experimental results. Post-operational and pre-operational results are presented in tables in each section. Comparisons are made, changes are discussed and conclusions are presented in each section. The largest change was observed for sediments in the Bailey Cove estuary and is presented as Section 2.14 Sediment Transect Survey and Analysis.

## 2.1 RADIONUCLIDES IN THE SOIL AND SEDIMENT

### 2.11 Laboratory Soil and Sediment Measurements and Their Analysis

Soil samples were collected at the following sites: (Foxbird Island, 0.1 km, S.; Eaton Farm, 0.4 km, W.; Bailey Farm, 0.8 km, N.E.; Young's Creek, 1.0 km, N.; Knight Cemetary, 1.1 km, E.; Westport Firehouse, 1.8 km, S.; Chewonki Neck (Camp), 1.9 km, S.W.; Cowseagan Narrows, 3.2 km, N.E. and Bluff Head, 4.0 km, S.S.W., see map). Sediment samples were collected at Foxbird Island, 0.1 km, S, on the outfall side of the causeway and at Murphy's Corner, 2.8 km, S.W. Soil samples were collected using a disc-cutter sampler which would cut a cylindrical soil sample 15 cm in diameter by 15 cm deep. Four soil samples this size, each centered at the corner of a square grid 25 cm on an edge were collected at each of the nine sites. The four soil samples including surface vegetation were mixed and the root mats pulverized. The sample was then screened and all material smaller than 5 mm retained. The sample was then dried at 110<sup>0</sup>C for 24 hours. In the case of the sediment samples, the top two centimeters were collected. These samples were screened and dried in an identical manner. In each case the samples were divided and a dry kilogram of the sample material was shipped to the Eastern Environmental Radiation Facility, Montgomery, Alabama for gamma-ray analysis using a Ge(Li) detector and on-line computer-analyzer. Analysis was carried out by hand using the Compton continuum subtraction method<sup>(8)</sup>. All samples were counted for a nominal 900 minutes (with a nominal 10 percent dead time) with a 2048 channel analysis. The results of the analysis are listed in Table I. The results of the pre-operational analysis are listed in Table II.

TABLE I  
POST-OPERATIONAL LABORATORY SOIL AND SEDIMENT GAMMA-RAY ANALYSIS

SAMPLE IDENTIFICATION				THORIUM SERIES	URANIUM SERIES	OTHER NATURAL	RADIONUCLIDES				
LOCATION	DATE COLLECTED	DATE COUNTED	TYPE	$^{228}\text{Ac}$ pCi/kg $\pm 2\sigma$	$^{214}\text{Bi}$ pCi/kg $\pm 2\sigma$	$^{40}\text{K}$ pCi/kg $\pm 2\sigma$	$^{137}\text{Cs}$ pCi/kg $\pm 2\sigma$	$^{134}\text{Cs}$ pCi/kg $\pm 2\sigma$	$^{58}\text{Co}$ pCi/kg $\pm 2\sigma$	$^{60}\text{Co}$ pCi/kg $\pm 2\sigma$	$^{54}\text{Mn}$ pCi/kg $\pm 2\sigma$
Foxbird Island	8/14/74	11/16/74	soil	900 $\pm 90$	700 $\pm 200$	7300 $\pm 950$	4600 $\pm 180$	< 35	< 25	< 30	< 15
Eaton Farm	8/14/74	11/18/74	soil	1100 $\pm 120$	1100 $\pm 120$	17400 $\pm 1200$	2500 $\pm 130$	< 35	< 25	< 30	< 15
Bailey Farm	8/14/74	11/20/74	soil	1100 $\pm 150$	1100 $\pm 170$	17600 $\pm 1300$	1800 $\pm 100$	< 35	< 25	< 30	< 15
Young's Creek	8/14/74	11/18/74	tidal marsh soil	1100 $\pm 120$	800 $\pm 70$	19400 $\pm 780$	700 $\pm 50$	< 35	< 25	< 30	< 15
Knight Cemetery	8/14/74	11/19/74	soil	1700 $\pm 225$	1300 $\pm 180$	11300 $\pm 1300$	4000 $\pm 160$	< 35	< 25	< 30	< 15
Westport Firehouse	8/14/74	11/17/74	soil	800 $\pm 90$	800 $\pm 100$	11800 $\pm 700$	3200 $\pm 100$	< 35	< 25	< 30	< 15
Chewonki Neck (Camp)	8/14/74	11/14/74	soil	900 $\pm 90$	700 $\pm 80$	15100 $\pm 600$	1500 $\pm 50$	< 35	< 25	< 30	< 15
Cowseagan Narrows	8/14/74	11/15/74	soil	1000 $\pm 70$	1000 $\pm 100$	14300 $\pm 570$	1300 $\pm 50$	< 35	< 25	< 30	< 15
Bluff Head	8/14/74	11/19/74	soil	1200 $\pm 48$	1600 $\pm 80$	13300 $\pm 530$	1200 $\pm 50$	< 35	< 25	< 30	< 15
Foxbird Island	8/14/74	11/15/74	tidal marsh sediment	900 $\pm 300$	900 $\pm 270$	20500 $\pm 1600$	1000 $\pm 130$	1000 $\pm 130$	21000 $\pm 400$	2420 $\pm 250$	450 $\pm 100$
Murphy's	8/14/74	11/20/74	tidal flat sediment	900 $\pm 90$	800 $\pm 72$	18000 $\pm 500$	500 $\pm 35$	< 35	< 25	< 30	< 15

All measurements based on dry weight

TABLE II  
PRE-OPERATIONAL LABORATORY SOIL AND SEDIMENT GAMMA-RAY ANALYSIS

SAMPLE IDENTIFICATION				THORIUM SERIES	URANIUM SERIES	OTHER NATURAL	RADIONUCLIDES				
LOCATION	DATE COLLECTED	DATE COUNTED	TYPE	<sup>228</sup> Ac pCi/kg ±2σ	<sup>214</sup> Bi pCi/kg ±2σ	<sup>40</sup> K pCi/kg ±2σ	<sup>137</sup> Cs pCi/kg ±2σ	<sup>134</sup> Cs pCi/kg ±2σ	<sup>58</sup> Co pCi/kg ±2σ	<sup>60</sup> Co pCi/kg ±2σ	<sup>54</sup> Mn pCi/kg ±2σ
Foxbird Island	6/29/72	7/20/72	soil	1200 ±100	700 ±90	14000 ±1000	940 ±85	< 30	< 25	< 30	< 20
Eaton Farm	6/12/72	7/17/72	soil	340 ±120	630 ±70	14900 ±400	870 ±45	< 15	< 12	< 15	< 10
Bailey Farm	6/12/72	7/12/72	soil	1240 ±300	810 ±150	14600 ±1300	1670 ±110	< 30	< 25	< 30	< 20
Young's Creek	6/12/72	7/14/72	tidal marsh soil	880 ±250	1075 ±120	18200 ±400	800 ±80	< 30	< 25	< 30	< 20
Knight Cemetery	6/12/72	6/26/72	soil	1210 ±200	700 ±80	11200 ±1200	4960 ±110	< 30	< 25	< 30	< 20
Westport Firehouse	6/12/72	6/21/72	soil	1000 ±350	920 ±120	11800 ±900	1110 ±85	< 30	< 25	< 30	< 20
Chewonki Neck (Camp)	6/12/72	7/20/72	soil	1100 ±250	1370 ±300	13200 ±1200	3340 ±130	< 30	< 25	< 30	< 20
Cowseagan Narrows	6/13/72	7/24/72	soil	300 ±300	730 ±180	13400 ±1250	2620 ±130	< 30	< 25	< 30	< 20
Bluff Head	6/12/72	6/22/72	soil	660 ±300	1100 ±180	11300 ±1240	2030 ±110	< 30	< 25	< 30	< 20
Foxbird Island	6/29/72	7/28/72	tidal marsh sediment	250 ±130	500 ±180	15000 ±350	350 ±32	< 15	< 12	< 15	< 10
Murphy's Corner	7/3/72	7/27/72	tidal flat sediment	1660 ±280	740 ±120	15200 ±1200	450 ±80	< 30	< 25	< 30	< 20

The Thorium series as represented by  $^{228}\text{Ac}$  is found to be the same ( $\pm 2\sigma$ ) as the pre-operational measurements with significant changes only at Eaton Farm soil. At present there is no obvious explanation of the difference. The Uranium series as represented by  $^{214}\text{Bi}$  is found, in all cases, to be the same ( $\pm 2\sigma$ ) as pre-operational measurements. The concentration of  $^{40}\text{K}$  is found to be the same ( $\pm 2\sigma$ ) for all cases except Foxbird Island soil and Foxbird Island sediment. In the case of Foxbird Island soil, a decrease is observed and for Foxbird Island sediment, increase is observed.  $^{137}\text{Cs}$  is observed to be the same ( $\pm 2\sigma$ ) in all cases except Foxbird Island soil, Eaton Farm soil, Westport Firehouse soil, Chewonki Neck soil, Bluff Head soil, and Foxbird Island sediment. Finally,  $^{134}\text{Cs}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$  and  $^{54}\text{Mn}$  are all found to be present in Foxbird Island sediment. These four isotopes were not present in the sediment at the time of the pre-operational study.

This change is not in agreement with the Maine Yankee Environmental Impact Statement<sup>(2)</sup>. Specifically, page V-15, states - "Recreational and other uses of shorelines and waters near the plant will be permitted by the Applicant. Therefore, direct exposure to radiation from nuclides in the waters of the bay will be experienced. The individual receiving the highest radiation dose would probably be one who earns his livelihood by digging blood and sand worms in the mud flats in the vicinity of the reactor discharge. For this calculation, concentrations of radionuclides deposited on the mud flats were assumed to result from undiluted effluent water. Such a person was also postulated to be exposed to the mud flats for 2000 hr/yr. Based on these assumptions, the dose to the total body from radionuclides associated with the mud would be about 6 mrem per year for the individual receiving maximum exposure. Nearly all this exposure is from  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  deposited on the mud flats where the worms are harvested. The exposure to the hands of the worm diggers would be somewhat higher than their total-

body exposure since they sift through the mud while harvesting the worms."

We find that over (0.92) of the exposure will come from  $^{54}\text{Mn}$ ,  $^{58}\text{Co}$  and  $^{60}\text{Co}$  with the remaining portion (.08) coming from  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .

Furthermore, high pressure ion chamber measurements at Foxbird Island and Murphy's Corner show an increase of the intensity over sediment. At Foxbird Island, for example, this change in intensity is 5.3 (6.58)calc microrem per hour which comes to a dose change (for the 2000 hr/yr worm digger) of 10.6 (13.6)calc millirem per year. See Section 2.4 High Pressure Ion Chamber for details of these measurements. The range and extend of radio-nuclides in the sediment are presented in greater detail in Section 2.14 Sediment Transect Survey and Analysis.

## 2.12 Field Soil and Sediment Measurements and their Analysis

As prescribed by our pre-scans of soil samples described in 2.11 radionuclides in the soil were measured in the field at the following six locations. (Foxbird Island, 0.1 km, S.; Eaton Farm, 0.4 km, W.; Bailey Farm, 0.8 km, N.E.; Westport Firehouse, 1.8 km, S.; Chewonki Neck (Camp), 1.9 km, S.W.. On-site sediment measurements were taken at low tide at Murphy's Corner, 2.9 km, S.W. the location of a tidal mud flat of commercial importance to the local blood worm industry and Foxbird Island the site of the outflow. A portable multichannel analyzer system with a 5 cm by 5 cm NaI(Tl) detector was employed. The portable multichannel analyzer system consisted of a Northern Scientific, Inc. NS-710 multichannel analyzer powered by a Cornell-Dubilier Powercon sine wave inverted Model 12ESW25 and a 12 v.d.c., 96 amp-hour battery. The detector, a 5 cm by 5 cm NaI(Tl) crystal was an integral crystal-photomultiplier assembly by Teledyne Isotopes, Inc., Model S-88-I with 8.4 percent resolution at the  $^{137}\text{Cs}$  photo-peak. It was powered by a Northern Scientific, Inc. high voltage battery power pack NS-308 with a matched cable-base assembly NS-309. The detector was connected to the multichannel analyzer with 50 m of coaxial cable (RG-59). A 100 lb. lead shield consisted of a cylinder 28 cm high, and 18 cm in diameter with a 6 cm diameter concentric hole the full length of the cylinder. The detector was housed in this shield for each on-site measurement. The shield was placed on the ground, the detector lowered into the shield and a 3 cm thick lead cap covered the upper end of the shield. The lower end of the shield was open to the soil so that the circular face of the detector was placed on the ground (a 1/8 inch insulating layer of plywood was used to protect the crystal from thermal shock). All

field soil and sediment gamma-ray spectra were taken using this top shielded  $2\pi$  geometry<sup>(7)</sup>. It is felt that this geometry has two advantages: (i) it is reproducible and (ii) it is without bias in that it samples the soil in a nondestructive non-mixed manner. All field soil and sediment spectra were taken on one quarter memory (512 channels) for 4000 seconds. At the end of 4 runs the multichannel analyzer was returned to the University of Maine, Physics Department and the memory outputted into a model KSR-33 teletype. The teletype provided a listing of the counts in each channel and a punched paper tape. The punched paper tapes were converted into cards and the gamma spectra analyzed at the University of Maine Computer Center. The least squares method was used to obtain the best estimates of the amounts of each radionuclide present in each sample and an evaluation of the errors of these results. The computer program used was the Oak Ridge National Laboratory spectrum fitting program, Alpha-M<sup>(9)</sup>. It employs the least squares method to analyze the data for which a "best fit" is mathematically computed such that the sum of the squares of the deviations between the actual spectra and the "best fit" is minimized. The program also uses automatic gain shift and automatic threshold shift routines to optimize the fit. The program works with a library of standard spectra from which it synthesizes the "best fit" spectra. The library used for the analysis of the soil and sediment on-site spectra was composed of standard spectra for  $^{40}\text{K}$ ,  $^{137}\text{Cs}$ , the  $^{232}\text{Th}$  series,  $^{134}\text{Cs}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ , and the  $^{238}\text{U}$  series. Output from the program consists of the estimated amount of each standard spectra needed to synthesize the sample spectrum, the estimated error in the amount used for each library standard, the gain shift (if any) used to match the spectrum to the standards, the threshold or zero shift (if any) used to match the spectrum to the standards, the residuals for each channel and a listing of suspicious channels whose residuals lie outside two standard deviations.

In all cases convergence was obtained in less than 20 iterations. The results of the analysis are listed in Table III. Table IV lists results from the pre-operation survey.

One can see that in general all spectra contained  $^{40}\text{K}$ , and the natural decay series for  $^{232}\text{Th}$  and  $^{238}\text{U}$ . A significant amount of  $^{137}\text{Cs}$ , a non-natural isotope, is contained in all spectra. The field measurements at Foxbird Island tidal mud flats indicates  $^{58}\text{Co}$ , and  $^{60}\text{Co}$  at this site. Comparison of the levels of natural isotopes  $^{232}\text{Th}$ ,  $^{238}\text{U}$  and  $^{40}\text{K}$  and  $^{137}\text{Cs}$  which is partly due to fallout are similar to pre-operational levels: with the exception of the Foxbird Island measurement which had more  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and less  $^{137}\text{Cs}$ , and may be due to disturbances done by nearby construction of a diffuser for the reactor. Eaton Farm also shows more  $^{238}\text{U}$  series and is not explained.

Measurements at Foxbird Island and Murphy's Corner show  $^{58}\text{Co}$  and  $^{60}\text{Co}$ .  $^{60}\text{Co}$  was also measured at Bailey Farm and Knight Cemetery while  $^{58}\text{Co}$  was measured at Eaton Farm. These cobalt isotopes were not present at the time of the pre-operational study. Measurements at Foxbird Island indicate  $^{238}\text{U}$  and  $^{232}\text{Th}$  has increased and  $^{137}\text{Cs}$  decreased as compared to the pre-operational study. These changes in these three isotopes were probably caused by the extensive dust due to construction dredging trucking and back filling with estuarine sediment during construction of the diffuser channel and head basin.

TABLE III  
POST-OPERATIONAL FIELD SOIL AND SEDIMENT GAMMA-RAY ANALYSIS

LOCATION	DATE MEASURED	TYPE	<sup>232</sup> Th pCi/kg ±2σ	<sup>238</sup> U pCi/kg ±2σ	<sup>40</sup> K pCi/kg ±2σ	<sup>137</sup> Cs pCi/kg ±2σ	<sup>58</sup> Co pCi/kg ±2σ	<sup>60</sup> Co pCi/kg ±2σ
Foxbird Island	8/24/74	soil	2000 ±70	1250 ±120	17400 ±750	3800 ±230	< 30	150 ±90
Eaton Farm	8/22/74	soil	2300 ±80	1300 ±120	24700 ±740	2800 ±260	210 ±30	< 75
Bailey Farm	8/22/74	soil	2100 ±95	1000 ±130	18600 ±880	3380 ±370	< 30	60 ±90
Knight Cemetery	8/22/74	soil	3100 ±80	660 ±120	23200 ±790	6900 ±310	< 30	150 ±80
Murphy's Corner	8/22/74	sediment	1300 ±40	280 ±50	11000 ±370	590 ±130	180 ±30	70 ±40
Foxbird Island	8/24/74	sediment	1500 ±60	580 ±80	16500 ±470	1400 ±200	730 ±20	100 ±70

TABLE IV  
PRE-OPERATIONAL FIELD SOIL AND SEDIMENT GAMMA-RAY ANALYSIS

LOCATION	DATE MEASURED	TYPE	THORIUM SERIES pCi/kg	URANIUM SERIES pCi/kg	<sup>40</sup> K pCi/kg	<sup>137</sup> Cs pCi/kg	<sup>58</sup> Co pCi/kg	<sup>60</sup> Co pCi/kg
Foxbird Island	6/29/72	soil	1600 ±100	600 ±100	14000 ±3500	3590 ±800	< 30	< 75
Eaton Farm	6/26/72	soil	2400 ±100	700 ±100	21000 ±5000	5600 ±2700	< 30	< 75
Bailey Farm	6/27/72	soil	2000 ±100	800 ±200	16000 ±6000	3300 ±600	< 30	< 75
Young's Creek	6/27/72	soil	1600 ±400	600 ±200	14000 ±9000	4000 ±1400	< 30	< 75
Knight Cemetery	6/27/72	soil	3100 ±300	600 ±200	20000 ±6000	9500 ±1600	< 30	< 75
Westport Firehouse	6/27/72	soil	1500 ±100	500 ±100	14000 ±3000	7600 ±2000	< 30	< 75
Chewonki Neck (Camp)	6/26/72	soil	1900 ±100	500 ±150	20000 ±4000	5000 ±1100	< 30	< 75
Cowseagan Narrows	6/27/72	soil	1700 ±100	500 ±100	15000 ±3000	5400 ±800	< 30	< 75
Bluff Head	6/28/72	soil	2000 ±100	800 ±300	16000 ±5000	6300 ±1400	< 30	< 75
Murphy's Corner	7/3/72	sediment	1400 ±100	400 ±200	9000 ±4000	7000 ±1900	< 30	< 75

## 2.14 Sediment Transect Survey and Analysis

As a result of the high readings for the pre-scan and in situ gamma-ray measurements of sediment from Foxbird Island and as a result of suggestions from our collaborator Mr. Charles Phillips of Eastern Environmental Radiation Facility, Montgomery, Alabama, we decided to make a survey along tidal transects of the sediments in Bailey Cove. Using the cooperative efforts of the Ira C. Darling Center for marine research and members of the bloodworm research project, under the direction of Marine Biologist A. H. Price, 50 samples of estuarine sediments were collected at 50 ft. transects in Baily Cove adjacent to the outflow of the reactor. The locations of the sites are shown in Figure 2. These sediments were counted for 5000 sec. using the University of Maine, Physics Department's Ge(Li) detector and low background shield. Results of the determination of the concentration of gamma-ray emitting isotopes in the sediment is shown in pCi/kg. Figures 3 and 4 illustrate activity maps for  $^{58}\text{Co}$  and  $^{60}\text{Co}$ , respectively. It should be noted that the highest levels of radioactivity are for the outflow site (29) and for the upper reaches of the cove. The constant picocurie lines seem to follow the flow of water out of the cove at low tide and into the upper cove at high tide and thus suggest that the isotopes are transported by the outflow water and reconcentrated into the sediments. This reconcentration is consistent with a diffusion theory for fallout radionuclides in sediments as suggested by Lerman<sup>(10)</sup> and with chemical precipitation of  $^{58}\text{Co}$  in the effluent of reactors as suggested by Fukui<sup>(11)</sup>. Comparison of the average flow velocities of the water from the outflow suggests sedimentation as another mechanism which reconcentrates the nuclides. Some indication of

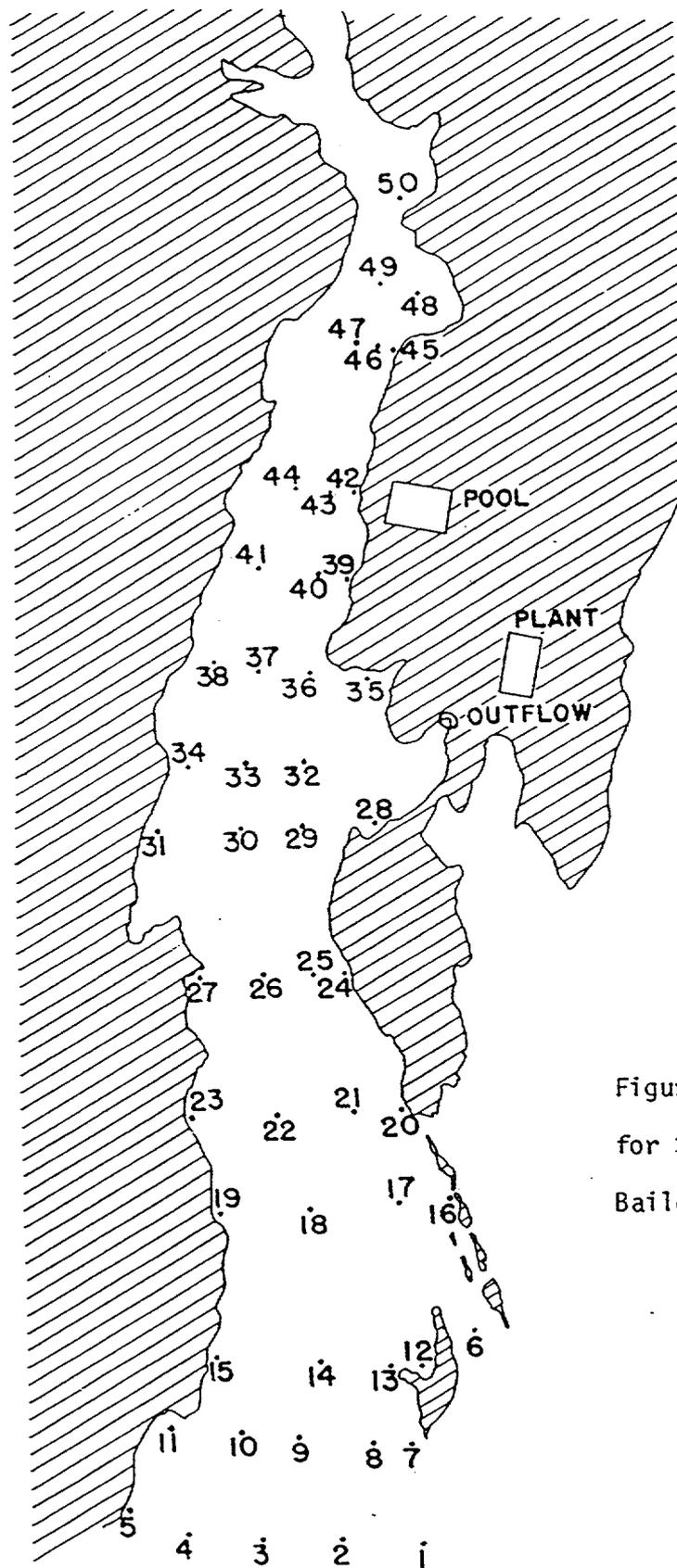


Figure 2, Site Location  
for Sediment Transect  
Bailey Cove

175 meters

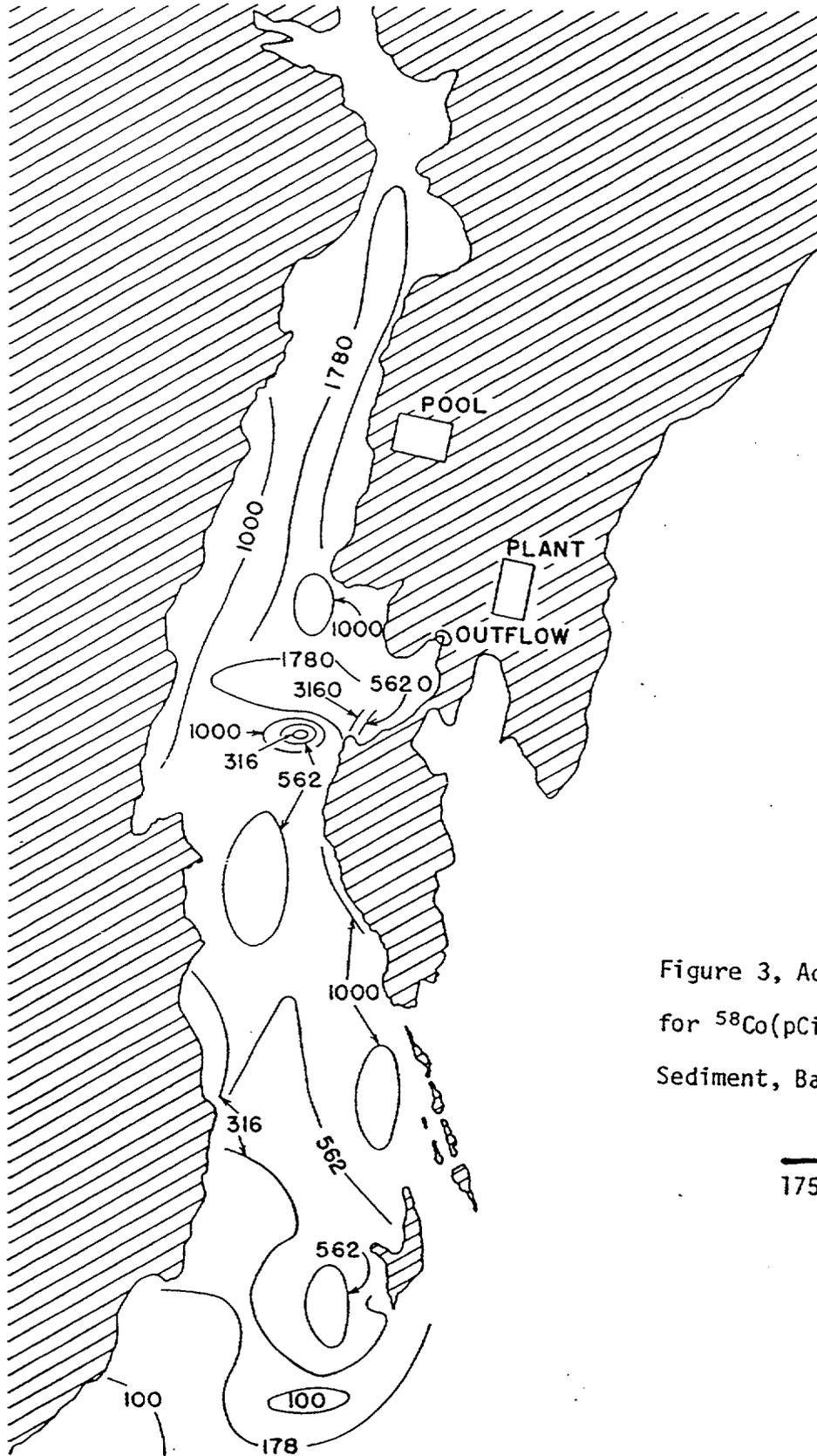


Figure 3, Activity Ma  
for  $^{58}\text{Co}$ (pCi/kg) in  
Sediment, Bailey Cove

175 meters

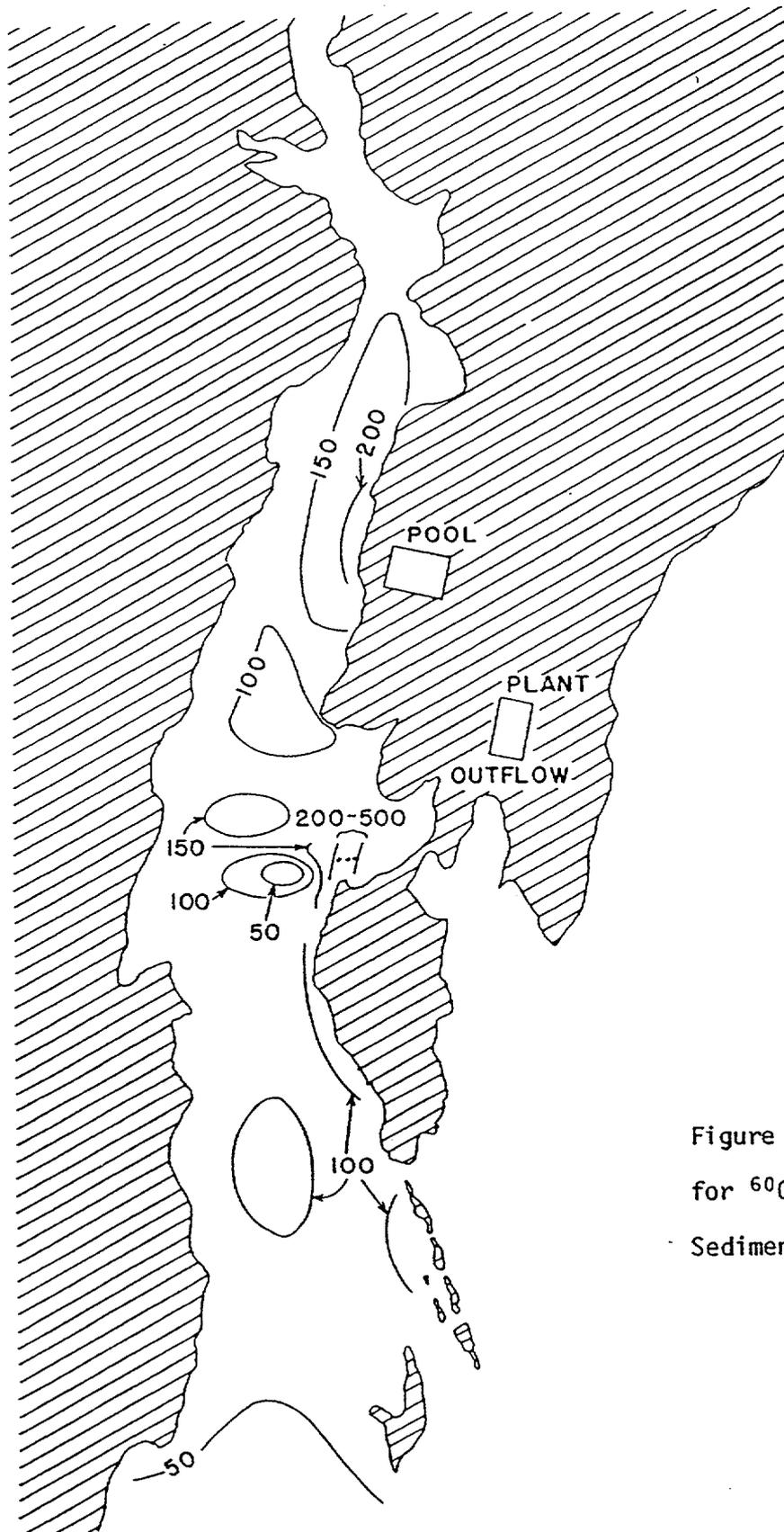


Figure 4, Activity  
for  $^{60}\text{Co}$ (pCi/kg) i  
Sediment, Bailey C

175 meter

this may be seen in the reduced level of nuclides along the most direct channel of the outflow. The radionuclides  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$  bear this out by having similar behavior to the  $^{58}\text{Co}$ . Since the Maine Yankee Environmental Impact Statement<sup>(2)</sup> does not specify range or distribution guidelines or estimates no comparison is made. However, implicit in the model<sup>(12)</sup> upon which the dose calculations were based is an assumption of uniform distribution of effluent discharge. This is not the case in the vicinity of Maine Yankee as shown by this transect survey in Bailey Cove. One radioactive particle containing 7700 pCi of  $^{60}\text{Co}$  was observed in the sediment transect #19 and had a total activity ( $^{58}\text{Co}$ , 530 pCi;  $^{46}\text{Sc}$ , 670 pCi;  $^{54}\text{Mn}$ , 120 pCi) of more than 9000 pCi in a mass less than 20  $\mu$  grams. Photographs and x-rays powder patterns were made and it was found to be cubic cobalt, alpha iron, nickel, alpha and gamma manganese and possibly chromium and vanadium. This stainless steel like composition suggests reactor origin. The particle was also checked for the oxides, chlorides, nitrates, sulphates and hydroxides of these metals with negative results<sup>(13)</sup>.

## 2.2 RADIONUCLIDES IN THE WATER

### 2.21 Field Water Measurements

Based upon the data obtained during the pre-operational field water measurements, the post-operational measurements showed no detectable amount of gamma activity above background. It is felt that field measurement of in-situ water with portable multichannel analyzer-detector equipment is significantly less sensitive than laboratory measurement and yields no additional information. Laboratory measurement, with the advantages larger detectors and massive shielding together with the practicality of longer measuring time, outweighs the advantages of preserving temperature, particulate and salinity gradients as is done in the laboratory water measurement.

### 2.22 Laboratory Water Measurements and Their Analysis

Water samples were collected at the following seven sites Foxbird Island, 0.1 km, S.; Eaton Farm, 0.4 km, W.; Bailey Farm, 0.8 km, N.E.; Young's Creek, 1.0 km, N.; Chewonki Neck (Camp), 1.9 km, S.W.; Cowseagan Narrows, 3.2 km, N.E.; and Bluff Head, 4.0 km, S.S.W., see map Figure 1. Each water sample was separated, by filtering the dissolved and the undissolved solids into two components. This separation was done in the field at the time of collection. The water was filtered using a Millipore high pressure filtering unit. A Millipore 90 mm filter holder was loaded with a Whatman No. 1 paper filter, a Millipore AP3207500 spacer, a Millipore SCWP09025 cellulose ester 8.0 $\mu$  filter, a Millipore AP3207500 spacer and a Millipore HAWP09025 cellulose ester 0.45 $\mu$  filter for each water sample. Ten liters of water from each site was filtered using the above arrangement.

The filtered water (dissolved component) was placed in one gallon plastic shipping containers to which 40 ml of nitric acid was added to stabilize the sample. Prior to acidification a 50 ml sample from each site was placed in a plastic bottle for tritium analysis, (see section 2.23). The dissolved component, (one gallon samples) and the undissolved component (filters) were sent to the Eastern Environmental Radiation Facility, Montgomery, Alabama for gamma-ray analysis.

The dissolved component was gamma-ray analyzed using a NaI(Tl) detector and 3.5 liter Marinelli beaker geometry in a massive shield. The results of this analysis show no detectable gamma activity in the dissolved component. (See Appendix E for pre-operational laboratory dissolved water component).

The undissolved component was analyzed in the following manner. The filters were weighed before use. After use they were dried and reweighed to determine the amount of undissolved material. The filters were sent to the Eastern Environmental Radiation Facility, Montgomery, Alabama for gamma-ray analysis. The three filters for each site were first measured for gross alpha and gross beta activity and then counted in a 40 ml NaI(I well-crystal 10 cm x 13 cm with 3 cm deep well. The results of these measurements are presented in Table V. Table VI lists the results for the pre-operational survey.

TABLE V  
 POST-OPERATIONAL LABORATORY WATER MEASUREMENT  
 UNDISSOLVED COMPONENT (FILTER) COLLECTED 8/14/74

LOCATION	WATER TYPE	SAMPLE MASS (GRAMS)	GROSS ALPHA pCi/10 liters $\pm 2\sigma$	GROSS BETA pCi/10 liters $\pm 2\sigma$	GROSS GAMMA pCi/10 liters $\pm 2\sigma$
Foxbird Island	estuarine	0.165 $\pm$ 0.015	ND	1.4 $\pm$ 81%	ND
Eaton Farm	well	0.005 $\pm$ 0.015	18.9 $\pm$ 85%	2.6 $\pm$ 39%	ND
Bailey Farm	well	0.002 $\pm$ 0.015	60.7 $\pm$ 33%	21.1 $\pm$ 22%	ND
Long Ledge Creek (Young's Creek)	surface	0.621 $\pm$ 0.045	2.1 $\pm$ 59%	4.8 $\pm$ 59%	ND
Chewonki Neck (Camp)	well	0.005 $\pm$ 0.015	6.2 $\pm$ 66%	1.6 $\pm$ 57%	ND
Cowseagan Narrows	estuarine	0.122 $\pm$ 0.015	ND	1.3 $\pm$ 69%	ND
Bluff Head	estuarine	0.149 $\pm$ 0.015	ND	ND	ND

TABLE VI  
 PRE-OPERATIONAL LABORATORY WATER MEASUREMENT  
 UNDISSOLVED COMPONENT (FILTER) COLLECTED 6/13/72

LOCATION	WATER TYPE	SAMPLE MASS (GRAMS)	$^{228}\text{Ac}$	$^{212}\text{Bi}$	$^{208}\text{Tl}$	$^{214}\text{Pb}$	$^{214}\text{Bi}$	$^{40}\text{K}$
Foxbird Island	estuarine	0.131 ± 0.005	< 45	100 ± 20	< 25	< 10	< 10	< 25
Eaton Farm	well	0.000 ± 0.005	< 45	< 10	< 25	30 ± 25	< 10	< 25
Bailey Farm	well	0.007 ± 0.005	200 ± 45	< 10	< 25	< 10	< 10	< 25
Long Ledge Creek (Young's Creek)	surface	0.045 ± 0.005	< 45	< 10	< 25	< 10	< 10	< 25
Chewonki Neck Camp	well	0.002 ± 0.005	< 45	< 10	< 25	< 10	< 10	< 25
Cowseagan Narrows	estuarine	0.114 ± 0.005	< 45	< 10	30 ± 25	< 10	< 10	< 25
Bluff Head	estuarine	0.163 ± 0.005	< 45	< 10	70 ± 25	< 10	< 10	< 25

## 2.22 Discussion of the Water Gamma-Ray Analysis

Several observations can be made from the data in Table V. The well water in this region of Maine contains considerable dissolved  $^{222}\text{Rn}$ . This is due primarily to leaching from uranium oxide in the pegmatite deposits in the area. This is also reflected in the alpha and beta activity in the well water from the sites measured (Eaton Farm, Bailey Farm and to some extent Chewonki Neck Camp). In general the water measurements at all sites show no significant change from the pre-operational survey shown in Table VI and, with the exception of the well water, have typical activities for this type of environmental media. The specific activity of the undissolved component in the estuarine water is of the same magnitude as the specific activity in the estuarine sediments. The specific activity of the well water samples was about two orders of magnitude greater but from the pre-operational survey this is known to be of natural origin.

## 2.23 Tritium in the Water, Air Moisture and Precipitation

### 2.231 Tritium in the Water

Tritium in the water was measured in samples collected at the following seven locations, Foxbird Island, 0.1 km, S.; Eaton Farm, 0.4 km, W.; Bailey Farm, 0.8 km, N.E.; Young's Creek, 1.0 km, N.; Chewonki Neck (Camp) 1.9 km, S.W.; Cowseagan Narrows, 3.2 km, N.E. and Bluff Head, 4.0 km, S.S.W.; (see Fig. 1). Each water sample was filtered as described in section 2.2. A 50 milliliter sample from each site was sent to the Eastern Environmental Radiation Facility, Montgomery, Alabama for tritium analysis using the direct counting liquid scintillation method. The samples were counted in a low background chamber. The results of these post-operational measurements are presented in Table VII. Pre-operational measurements are presented in Table VIII.

TABLE VII  
POST-OPERATIONAL TRITIUM IN WATER

LOCATION	DATE COLLECTED	TYPE	ACTIVITY nCi/1±2σ	INTERPRETED RESULT
Foxbird Island	8/15/74	estuarine	0.2 ± 0.2	at m.d.l.
Eaton Farm	8/15/74	well	< 0.2	below m.d.l.
Bailey Farm	8/15/74	well	0.2 ± 0.2	at m.d.l.
Long Ledge Creek (Young's Creek)	8/15/74	surface	0.3 ± 0.2	above m.d.l.
Chewonki Neck (Camp)	8/15/74	well	0.2 ± 0.2	at m.d.l.
Cowseagan Narrows	8/15/74	estuarine	< 0.2	below m.d.l.
Bluff Head	8/15/74	estuarine	< 0.2	below m.d.l.

The minimum detectable level (m.d.l.) for the analysis of tritium is 0.2 nCi/l.

TABLE VIII  
PRE-OPERATIONAL TRITIUM IN WATER

LOCATION	DATE COLLECTED	TYPE	ACTIVITY nCi/1±2σ	INTERPRETED RESULT
Foxbird Island	6/13/72	estuarine	0.39 ± 0.2	zero
Eaton Farm	6/13/72	well	0.40 ± 0.2	zero
Bailey Farm	6/13/72	well	0.34 ± 0.2	zero
Young's Creek	6/13/72	surface	0.14 ± 0.2	zero
Chewonki Neck (Camp)	6/13/72	well	0.30 ± 0.2	zero
Cowseagan Narrows	6/13/72	estuarine	0.09 ± 0.2	zero
Bluff Head	6/13/72	estuarine	0.40 ± 0.2	zero

TABLE IX  
POST-OPERATIONAL TRITIUM IN AIR MOISTURE

LOCATION	DATE COLLECTED	WATER QUALITY MEASURED	AIR VOLUME	MEASURED RESULTS $\pm 2\sigma$ nCi/l WATER	INTERPRETED RESULTS nCi/l
Eaton Farm	8/27/74	32.8 ml	1280 l	< 0.2	zero
Bailey Farm	8/27/74	33.0 ml	1990 l	< 0.2	zero
Westport Firehouse	8/27/74	25.4 ml	1730 l	< 0.2	zero

TABLE X  
PRE-OPERATIONAL TRITIUM IN AIR MOISTURE

LOCATION	DATE COLLECTED	WATER QUALITY MEASURED	AIR VOLUME	MEASURED RESULTS $\pm 2\sigma$ nCi/l WATER	INTERPRETED RESULTS nCi/l
Eaton Farm	6/29/72	33.7 ml	2502.7 l	0.60 $\pm$ 0.2	0.6
Bailey Farm	6/15/72	33.1 ml	2000.0 l	19.1 $\pm$ 0.2	19.1
Westport Firehouse	6/27/72	33.4 ml	2500.0 l	0.60 $\pm$ 0.2	0.6

TABLE XI  
POST-OPERATIONAL TRITIUM IN PRECIPITATION

LOCATION	DATE COLLECTED	TYPE	MEASURED RESULTS nCi/l $\pm 2\sigma$	INTERPRETED RESULTS nCi/l
Eaton Farm	8/27/74	RAIN	< 0.2	zero
Bailey Farm	8/27/74	RAIN	0.3 $\pm$ 0.2	0.3
Knight Cemetery	4/9/74	SNOW	< 0.2	zero

TABLE XII  
PRE-OPERATIONAL TRITIUM IN PRECIPITATION

LOCATION	DATE COLLECTED	TYPE	MEASURED RESULTS nCi/l $\pm 2\sigma$	INTERPRETED RESULTS nCi/l
Eaton Farm	6/30/72	RAIN	0.40 $\pm$ 0.2	zero
Bailey Farm	6/30/72	RAIN	1.10 $\pm$ 0.2	1.1
Knight Cemetery	3/11/72	SNOW	0.26 $\pm$ 0.2	zero
Westport Firehouse	3/11/72	SNOW	0.17 $\pm$ 0.2	zero

### 2.232 Tritium in the Air Moisture

Tritium in the air moisture was measured in samples collected at the following three sites, Eaton Farm, 0.4 km, W.; Bailey Farm, 0.8 km, N.E.; and Westport Firehouse, 1.8 km, S. Air moisture samples were collected by drawing nominally 2000 liters of air through a plastic cylinder (8 cm diameter by 24 cm long) containing a desiccant<sup>(14)</sup>. The desiccant used was indicator type Dryrite. The cylinder of desiccant was weighed before the air was pulled through and weighed afterward to insure the collection of between twenty and thirty milliliters of air moisture. Nominal collect time was between two and four hours depending on the relative humidity. The desiccant cylinders were sealed and sent to the Eastern Environmental Radiation Facility in Montgomery, Alabama for tritium analysis. Water was exchanged with the desiccant moisture in a closed system. The resulting samples were processed for liquid scintillation tritium analysis in the usual way. The results of these measurements indicate the level for tritium in the air moisture was below the minimum detectable level of 0.2 nCi/l at all locations sampled on the data sampled (8/27/74).

### 2.233 Tritium in Precipitation

Tritium in precipitation was measured in two snow samples collected at Knight Cemetery, 1.1 km, E. on April 9, 1974 and rain samples collected at Eaton Farm, 0.4 km, W. and Bailey Farm, 0.8 km, N.E. on August 27, 1974. These samples were filtered in the same manner used for the water samples (See section 2.22) Fifty milliliter samples were sent to the Eastern Environmental Radiation Facility, Montgomery, Alabama for tritium analysis using the liquid scintillation method. (See section 2.231) The results

of the analysis of tritium activity in the rain at Eaton Farm showed it to be below the minimum detectable level of 0.2 nCi/liter and the level at Bailey Farm to be  $0.3 \pm 0.2$  nCi/liter, greater than the minimum detectable level. Tritium activity in the snow samples was below the minimum detectable level.

#### 2.24 Discussion of Tritium Measurements

The purpose of tritium measurements in this study is to document typical values for the HTO component in water, air moisture and precipitation. In general the results of tritium analysis indicate no significant change. There is no evidence of an accumulation of tritium in the interchangeable environmental reservoirs such as wells, surface water or the Montsweag Bay estuary. Even though the Maine Yankee Environmental Impact Statement<sup>(2)</sup> lists a bioaccumulation factor of 1 for tritium, the authors feel that in light of the fact that tritium is one of the major radionuclide in the liquid and gaseous effluent, that tritium monitoring should be carried out on a regularly scheduled basis for a period of one half life (12.5 years) in order to check for possible long term accumulation. The only significant correlation between the pre-operational survey for tritium and the post-operational survey is that the rain samples at the Bailey Farm location contain tritium above the minimum detectable level. The large amount of tritium in the air moisture observed on 6/15/72 in the pre-operational survey was not observed in the post-operational study.

## 2.3 Radionuclides in Air Particulates

### 2.31 Field Air Particulate Measurements and Analysis

Radionuclides in air particulates were measured at three sites: Eaton Farm, Bailey Farm and Westport Firehouse. A Staplex large volume air sampler was used in conjunction with a 20.5 cm (8 1/16 inch) by 25.4 cm (10 inch) filter holder. Nuclear Associates, Inc. fiberglass filters number 08-780 were used. Approximately 3400 ft<sup>3</sup> of air were filtered at each site. The filter was folded three times (8 layers) and placed on the circular surface of the NaI(Tl) detector assembly used for the soil and sediment field measurements, and covered with a 3 cm lead shield. All air particulate field data were counted as soon after collection as possible (normally within a few minutes) on quarter memory (512 channels) for 2000 seconds. The multichannel analyzer was outputted on the teletype. Qualitative analysis shows that the bulk of the activity was due to daughters of <sup>222</sup>Rn and <sup>220</sup>Rn. After a period of 24 hours the activity of particulate on the filters was found to be long half-life radon daughters.

### 2.32 Laboratory Air Particulate Measurements

Air particulates for laboratory analysis were also collected at these same sites (Eaton Farm, Bailey Farm, Westport Firehouse). A Millipore pump type XX6000000 was used to pull air through a 47 mm diameter Millipore absolute aerosol 0.8 μm filter, type AAWP4700. The throughput of this system was metered for all runs and a calibrated gas flow meter, American Meter Company Model 10-300-PR1264. The meter, pump, and filter holder were housed in an instrument box with a rubber hose from the filter holder to the outside. A nominal sampling time was 100 hours, with a nominal sampi

volume being 200,000 liters. The filters were weighed before and after collection. They were packaged carefully in separate plastic boxes and sent to the Eastern Environmental Radiation Facility, Montgomery, Alabama, for gamma-ray analysis. Each sample was counted for gamma-rays in the 40 ml NaI(Tl) well-counter which is 11 cm x 15 cm and has a 3 cm well. The results of these measurements are given in Table XIII. The gross-alpha measurements were counted in an internal proportional counter and the gross-beta measurements were counted in a low background beta counter. The results of these measurements are also given in Table XIII. The pre-operational measurements are listed in Table XIV.

## Discussion of the Air Particulate Results

The short term field air particulate collections and measurements reveal the presence of  $^{220}\text{Rn}$  and  $^{222}\text{Rn}$  as the most important source of activity, essentially identical to the pre-operational study of 1972.

Large volume laboratory air particulate measurements as presented in TABLES XIII and XIV show no significant changes.

### 2.4 High Pressure Ion Chamber

#### 2.41 High Pressure Ion Chamber Measurements

High pressure ion chamber measurements had been carried out in 1971 by Wesley R. Van Pelt of Environmental Analysis, Inc. for Maine Yankee. Due to the relevance for population dose calculations and for comparison with the gamma-ray field studies, high pressure ion chamber measurements were carried out in collaboration with Mr. Charles Phillips of the Eastern Environmental Radiation Facility, Montgomery, Alabama. Both studies employed calibrated Reuter Stokes high pressure ion chambers. In the present study the ion chamber was RSS-111 (Reuter Stokes environmental monitor), with Rustrak Recorder. The monitor was placed 1 meter above the surface at 11 soil sites and 2 sediment sites. The soil sites were Foxbird Island, 0.1 km, S.; Eaton Farm, 0.4 km, W.; Bailey Farm, 0.8 km, N.E.; Young's Creek, 1.0 km, N.; Knight Cemetery, 1.1 km, E.; Westport Firehouse, 1.8 km, S.; Chewonki Neck (Camp) 1.9 km, S.W.; Cowseagan Narrows, 3.2 km, N.E.; Bluff Head, 4.0, S.S.W.; Cromwells 1.0 km, E.; and sediment sites Foxbird Island, 0.1 km, S.; and Murphy's Corner, 2.8 km, S.W.. The results of these measurements are shown in Table XV. Table XV lists high pressure ion chamber measurements of dose rate ( $\mu\text{R/hr}$ ) for the post-operational and pre-operational<sup>(15)</sup> surveys. The most significant change occurred at the

Foxbird Island (sediment) site at 42 percent increase in dose rate was measured. The last column lists post-operational calculated dose rate based upon laboratory measurement of specific activity for all significant gamma emitting radionuclides observed. The method used, due to Beck, is discussed in Appendix C<sup>(16)</sup>.

TABLE XIII  
POST-OPERATIONAL AIR PARTICULATE ANALYSIS

LOCATION	COLLECTION DATE	AIR VOLUME LITERS	PARTICULATE MASS (GRAMS)	α-ACTIVITY pCi/FILTER ±2σ	β-ACTIVITY pCi/FILTER ±2σ	γ-ACTIVITY pCi/FILTER ±2σ
Bailey Farm	8/19/74	166,230	0.016 ± 0.001	5.3 ± 68%	29.3 ± 73%	34 ± 37% ( <sup>7</sup> Be)
Eaton Farm	8/22/74	197,350	0.004 ± 0.001	6.0 ± 70%	25.4 ± 10%	4 ± 18% ( <sup>95</sup> ZrNb)
Westport Firehouse	8/27/74	191,960	0.005 ± 0.001	7.7 ± 55%	4.4 ± 11%	NONE

TABLE XIV  
PRE-OPERATIONAL AIR PARTICULATE ANALYSIS

LOCATION	COLLECTION DATE	AIR VOLUME LITERS	PARTICULATE MASS (GRAMS)	γ-ACTIVITY ZrNb	γ-ACTIVITY ( <sup>7</sup> Be)
Bailey Farm	6/19/72	97,740	0.002 ± 0.001	<15 pCi ZrNb	<200 pCi
Eaton Farm	7/5/72	86,730	0.001 ± 0.001	< 5 pCi	< 50 pCi
Westport Firehouse	6/29/72	101,810	0.004 ± 0.001	<15 pCi	<200 pCi

TABLE XV  
 COMBINED PRE-OPERATIONAL AND POST-OPERATIONAL  
 HIGH PRESSURE ION CHAMBER MEASUREMENTS

SITE NAME (measured over soil unless otherwise noted)	POST-OPERATIONAL MEASUREMENTS DATE	POST-OPERATIONAL DOSE RATE $\mu\text{R/hr}$	PRE-OPERATIONAL MEASUREMENTS DATE	PRE-OPERATIONAL DOSE RATE $\mu\text{R/hr}$	POST-OPERATIONAL CALCULATED DOSE RATE ( $\mu\text{R/hr}$ )
Foxbird Island	8/24/74	10.1	-	-	8.9
Eaton Farm	8/22/74	10.2	9/20/71	9.5	12.0
Bailey Farm	8/22/74	9.7	9/20/71	9.5	12.0
Young's Creek	8/22/74	9.7	-	-	11.7
Knight Cemetery	8/22/74	11.2	-	-	13.0
Westport Firehouse	8/22/74	9.5	9/21/71	11.4	9.6
Chewonki Neck Camp	8/22/74	9.9	-	-	10.2
Cowseagan Narrows	8/22/74	10.2	-	-	10.9
Bluff Head	8/22/74	10.8	-	-	12.3
Foxbird Island (sediment)	8/24/74	12.6	9/21/71	7.3	14.51
Murphy's Corner (sediment)	8/22/74	8.9	9/21/71	7.91	10.8
Cromwells	8/22/74	9.1	-	-	-

APPENDIX A  
LIQUID EFFLUENT INVENTORY IN CURIES

TABLE XVI

	JULY-DEC 72	JAN-JUNE 73	JULY-DEC 73	JAN-JUNE 74	JULY-DEC 74	
<sup>140</sup> BaLa	NDA	NDA	NDA	6.09 x 10 <sup>-5</sup>	NDA	6.09 x 10 <sup>-</sup>
<sup>133</sup> I	NDA	NDA	NDA	2.93 x 10 <sup>-3</sup>	-	2.93 x 10 <sup>-</sup>
<sup>131</sup> I	4.12 x 10 <sup>-3</sup>	1.62 x 10 <sup>-3</sup>	4.12 x 10 <sup>-3</sup>	2.76 x 10 <sup>-1</sup>	8.20 x 10 <sup>-2</sup>	0.37
<sup>133</sup> Xe	1.49 x 10 <sup>-3</sup>	7.05 x 10 <sup>-2</sup>	1.49 x 10 <sup>-3</sup>	11.7	1.04 x 10 <sup>-2</sup>	11.78
<sup>135</sup> Xe	NDA	NDA	NDA	7.78 x 10 <sup>-3</sup>	NDA	7.78 x 10 <sup>-</sup>
<sup>137</sup> Cs	NDA	NDA	NDA	6.15 x 10 <sup>-3</sup>	9.81 x 10 <sup>-1</sup>	9.9 x 10 <sup>-</sup>
<sup>134</sup> Cs	NDA	NDA	NDA	2.53 x 10 <sup>-3</sup>	7.31 x 10 <sup>-1</sup>	7.3 x 10 <sup>-</sup>
<sup>60</sup> Co	2.04 x 10 <sup>-4</sup>	3.34 x 10 <sup>-3</sup>	2.04 x 10 <sup>-4</sup>	5.41 x 10 <sup>-3</sup>	1.46 x 10 <sup>-2</sup>	2.3 x 10 <sup>-</sup>
<sup>58</sup> Co	5.48 x 10 <sup>-3</sup>	6.56 x 10 <sup>-2</sup>	5.48 x 10 <sup>-3</sup>	2.17 x 10 <sup>-2</sup>	2.60 x 10 <sup>-1</sup>	3.58 x 10 <sup>-</sup>
<sup>51</sup> Cr	NDA	2.68 x 10 <sup>-2</sup>	NDA	3.78 x 10 <sup>-2</sup>	-	4.04 x 10 <sup>-</sup>
<sup>54</sup> Mn	8.06 x 10 <sup>-4</sup>	1.12 x 10 <sup>-2</sup>	8.06 x 10 <sup>-4</sup>	1.99 x 10 <sup>-3</sup>	NDA	1.48 x 10 <sup>-</sup>
<sup>103</sup> Ru	NDA	NDA	NDA	NDA	-	NDA
<sup>90</sup> Sr	LOST	NDA	LOST	1.40 x 10 <sup>-5</sup>	NDA	1.40 x 10 <sup>-</sup>
<sup>99</sup> Mo	1.5 x 10 <sup>-5</sup>	NDA	1.50 x 10 <sup>-5</sup>	2.30 x 10 <sup>-3</sup>	-	2.33 x 10 <sup>-</sup>
<sup>95</sup> Zr	-	3.1 x 10 <sup>-3</sup>	-	1.54 x 10 <sup>-3</sup>	-	4.64 x 10 <sup>-</sup>
<sup>95</sup> Nb	-	2.9 x 10 <sup>-3</sup>	-	2.62 x 10 <sup>-3</sup>	-	5.52 x 10 <sup>-</sup>
<sup>59</sup> Fe	-	5.95 x 10 <sup>-3</sup>	-	-	1.19 x 10 <sup>-3</sup>	7.14 x 10 <sup>-</sup>
<sup>97</sup> Zr	1.5 x 10 <sup>-5</sup>	-	1.5 x 10 <sup>-5</sup>	-	-	3.0 x 10 <sup>-5</sup>
UN	1.34 x 10 <sup>-3</sup>	-	-	-	-	1.34 x 10 <sup>-</sup>
<sup>133m</sup> Xe	-	-	-	6.48 x 10 <sup>-5</sup>	-	6.48 x 10 <sup>-1</sup>
<sup>57</sup> Co	-	-	-	9.38 x 10 <sup>-6</sup>	NDA	9.38 x 10 <sup>-1</sup>
<sup>89</sup> Sr	-	-	-	-	2.94 x 10 <sup>-5</sup>	2.94 x 10 <sup>-1</sup>
<sup>3</sup> H	9.22	75.67	77.9	115	104	3.81 x 10 <sup>2</sup>

GASEOUS EFFLUENT INVENTORY IN CURIES

	OCT-DEC 72	JAN-JUNE 73	JULY-DEC 73	JAN-JUNE 74	JULY-DEC 74
<sup>137</sup> Cs	NDA	NDA	NDA	3.51 x 10 <sup>-5</sup>	NDA
<sup>140</sup> BaLa	NDA	NDA	NDA	NDA	NDA
<sup>90</sup> Sr	NDA	NDA	NDA	-	-
<sup>134</sup> Cs	NDA	NDA	NDA	1.37 x 10 <sup>-5</sup>	NDA
<sup>89</sup> Sr	-	-	-	-	-
<sup>58</sup> Co	-	3.9 x 10 <sup>-6</sup>	8.91 x 10 <sup>-5</sup>	2.3 x 10 <sup>-5</sup>	3.32 x 10 <sup>-4</sup>
<sup>60</sup> Co	-	-	-	5.1 x 10 <sup>-6</sup>	1.72 x 10 <sup>-4</sup>
<sup>54</sup> Mn	-	-	-	-	NDA
<sup>110</sup> Ag	-	-	-	-	NDA
<sup>57</sup> Co	-	-	-	2.2 x 10 <sup>-8</sup>	2.10 x 10 <sup>-5</sup>
<sup>99</sup> Mo	-	-	-	2.9 x 10 <sup>-6</sup>	2.90 x 10 <sup>-6</sup>
<sup>131</sup> I	1.71 x 10 <sup>-6</sup>	9.39 x 10 <sup>-6</sup>	1.62 x 10 <sup>-3</sup>	6.58 x 10 <sup>-2</sup>	4.98 x 10 <sup>-2</sup>
<sup>133</sup> I	NDA	NDA	3.28 x 10 <sup>-4</sup>	3.00 x 10 <sup>-2</sup>	1.48 x 10 <sup>-3</sup>
<sup>135</sup> I	NDA	NDA	NDA	5.79 x 10 <sup>-3</sup>	NDA
<sup>85</sup> Kr	2.05	6.3 x 10 <sup>-3</sup>	1.505	3.50 x 10 <sup>2</sup>	4.36 x 10 <sup>2</sup>
<sup>133</sup> Xe	NDA	6.36	151.56	3.81 x 10 <sup>3</sup>	2.41 x 10 <sup>3</sup>
<sup>88</sup> Kr	NDA	NDA	NDA	NDA	NDA
<sup>87</sup> Kr	NDA	NDA	NDA	NDA	NDA
<sup>138</sup> Xe	NDA	NDA	NDA	NDA	NDA
<sup>135m</sup> Xe	NDA	NDA	NDA	-	NDA
<sup>41</sup> Ar	7.1 x 10 <sup>-2</sup>	NDA	1.00 x 10 <sup>-1</sup>	NDA	NDA
<sup>133m</sup> Xe	-	-	-	NDA	3.30
<sup>131m</sup> Xe	-	8.9 x 10 <sup>-2</sup>	NDA	4.91 x 10 <sup>1</sup>	7.78 x 10 <sup>1</sup>
<sup>88</sup> Rb	NDA	NDA	-	NDA	-
<sup>85m</sup> Kr	-	NDA	NDA	NDA	NDA
<sup>135</sup> Xe	-	-	1.27 x 10 <sup>-1</sup>	1.90 x 10 <sup>2</sup>	5.50 x 10 <sup>-1</sup>
<sup>51</sup> Cr	-	-	-	8.4 x 10 <sup>-6</sup>	-
<sup>95</sup> Nb	-	-	-	1.78 x 10 <sup>-6</sup>	-

APPENDIX B  
PRE-SCAN RESULTS

The purpose of a pre-scan is to determine the optimum locations for the field studies. For this reason, the pre-scan was planned to take place early in the field studies and to provide usable results during the first week of the study. As is mentioned in section 2.11 of off-site soil measurements and their analysis, gamma-ray pre-scans were taken on kilogram samples from nine soil and two sediment sites. Samples were counted for 5000 seconds using the University of Maine Department of Physics Ge(Li) detector with low background shield. The lists of counts were then compared with lists of counts taken on pre-operational samples, collected in 1972. The examination of these lists provided the information for choosing the field locations. Later, punched tapes of these spectra were analyzed at the University of Maine Computer Center. The results of these analyses are shown in Table XVIII. Radionuclide concentrations are shown in picocurie per kilogram of sample. The first five columns of this table contain concentrations of natural isotopes:  $^{228}\text{Ac}$ ,  $^{208}\text{Tl}$  of the Thorium series,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  of the Uranium series, and  $^{40}\text{K}$ . The remaining columns of concentrations are of man-made fallout or reactor origin. Trace amounts of  $^{46}\text{Sc}$  or  $^{65}\text{Zn}$  were observed at Foxbird Island, Eaton Farm, Bailey Farm, Young's Creek, Knight Cemetery, Bluff Head, and Murphy's Corner, but could not be quantitatively determined due to interference from  $^{214}\text{Bi}$ . The radionuclides  $^{95}\text{Nb}$ - $^{95}\text{Zr}$  (200 pCi/kg) were observed at Foxbird Island, Bailey Farm, Young's Creek, Knight Cemetery, Westport Firehouse, Bluff Head, Murphy's Corner and Foxbird Island sediment, and  $^{131}\text{I}$  (300 pCi/kg) may have been observed in Murphy's Corner sediment. Errors are quoted under each number and represent  $\pm 1\sigma$ .

The site which showed largest amounts of man-made radionuclides was

Foxbird Island (sediment). There was no archival pre-scan result for this sample and Murphy's Corner sediment since there were no archival samples. In all other cases, pre-operational and post-operation samples were available and results are given in pairs in the table. Dashes in the table mean that results were below the minimum detectable limit. As a result of studying the pre-scan results (using the original lists) we selected for field studies: Foxbird Island soil and sediment, Eaton Farm, Bailey Farm, Knight Cemetery, soils and Murphy's Corner sediment.

TABLE XVIII PRE-SCAN RADIONUCLIDES pCi/kg  $\pm 1\sigma$ 

LOCATION	DATE OF COLLECTION	DATE OF MEASUREMENT	TYPE	<sup>228</sup> Ac	<sup>208</sup> Tl	<sup>214</sup> Pb	<sup>214</sup> Bi	<sup>40</sup> K	<sup>137</sup> Cs	<sup>58</sup> Co	<sup>60</sup> Co	<sup>134</sup> Cs	<sup>54</sup> Mn
Foxbird	6/29/72	8/15/73	soil	600 ±400	600 ±200	1000 ±100	700 ±200	11000 ±1000	800 ±100	-	-	200 ±60	-
Island	8/14/74	8/17/74	soil	600 ±400		700 ±500	700 ±200	10000 ±1000	5400 ±400	-	60 ±30	-	-
Eaton	6/12/72	8/14/73	soil	1100 ±200	1400 ±200	1100 ±200	900 ±100	15000 ±1200	1900 ±100	-	-	600 ±80	20 ±30
Farm	8/14/74	8/19/74	soil	1300 ±300	1200 ±200	1400 ±200	800 ±200	15000 ±1200	1900 ±100	-	-	400 ±70	-
Bailey	6/12/72	8/14/73	soil	1400 ±300	1600 ±200	2000 ±200	900 ±100	15000 ±1400	1300 ±100	-	-	200 ±60	-
Farm	8/14/74	8/17/74	soil	700 ±200	700 ±200	1000 ±100	800 ±100	15000 ±1200	1800 ±100	-	-	300 ±60	-
Young's	6/12/72	8/16/73	tidal marsh soil	1200 ±200	1400 ±200	1600 ±200	800 ±100	18000 ±1100	500 ±100	-	-	100 ±100	-
Creek	8/14/74	8/19/74	tidal marsh	1400 ±300	1800 ±300	1000 ±100	800 ±200	14000 ±1300	800 ±100	80 ±50	-	300 ±60	-
Knight	6/12/72	9/21/73	soil	2400 ±400	1600 ±200	1600 ±200	1400 ±300	13000 ±1400	6200 ±300	-	70 ±50	300 ±60	-
Cemetery	8/14/74	8/17/74	soil	1700 ±500	1400 ±200	1400 ±200	1100 ±200	10000 ±1200	3600 ±200	-	60 ±50	600 ±80	-
Westport	6/12/72	8/17/73	soil	1100 ±200	800 ±200	1700 ±200	1000 ±100	10000 ±900	1300 ±100	-	40 ±50	300 ±60	-
Firehouse	8/14/74	8/19/74	soil	400 ±200	1100 ±200	1200 ±200	700 ±200	11000 ±1300	2200 ±200	50 ±50	30 ±20	300 ±60	-
Chewonki	6/12/72	9/19/73	soil	1300 ±300	1300 ±300	1500 ±200	1200 ±200	16000 ±1400	5000 ±300	-	160 ±70	100 ±30	-
Neck Camp	8/14/74	8/20/74	soil	600 ±200	700 ±200	800 ±100	600 ±200	10000 ±1200	1700 ±100	-	90 ±44	300 ±60	-
Cowseagan	6/13/72	8/17/73	soil	700 ±300	900 ±300	1600 ±200	800 ±200	17000 ±1700	4000 ±200	120 ±64	40 ±50	200 ±60	-
Narrows	8/14/74	8/19/74	soil	1000 ±200	1100 ±200	1700 ±200	700 ±100	10000 ±1000	-	-	30 ±50	81 ±20	25 ±30
Bluff	6/12/72	8/21/73	soil	1900 ±300	1100 ±200	2000 ±200	1600 ±200	12000 ±1100	2500 ±200	80 ±50	-	100 ±30	-
Head	8/14/74	8/17/74	soil	1300 ±300	1500 ±200	1300 ±200	1100 ±200	12000 ±1100	1800 ±200	-	-	400 ±70	20 ±30
Murphy Corner	8/14/74	8/20/74	Sed.	1100 ±200	1100 ±200	1300 ±200	1000 ±200	12000 ±1000	400 ±200	100 ±40	20 ±30	400 ±70	-
Foxbird Island	8/14/74	8/20/74	Sed.	600 ±100	1400 ±300	2300 ±700	2000 ±200	8000 ±900	910 ±100	19000 ±400	800 ±100	1000 ±80	200 ±60

APPENDIX C

Calculation of Dose Rate at One Meter from Gamma-Ray Emitting  
Soils and Sediments

Calculation of gamma-ray dose rate due to a plane source such as soil or sediment is of practical importance. The factors for conversion from the isotope concentration of the plane medium to dose at one meter above the surface for  $^{40}\text{K}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{232}\text{Th}$ -series and  $^{238}\text{U}$ -series are found in a paper due to Beck<sup>(16,17)</sup>. Two new factors, using Beck's Table 7 and branching ratios from the literature, are calculated for  $^{58}\text{Co}$  and  $^{134}\text{Cs}$ . These eight factors are presented in Table XIX. Dose Calculations for eleven sites are presented in Table XV. The cosmic ray contribution is included using ionization chamber data gathered by W. Van Pelt<sup>(15)</sup> over water in 1971.

TABLE XIX  
 Factors for Computation of Dose Rate at 1 Meter due to Gamma-Ray  
 Emitting Radionuclides in Soil and Sediment

Isotope		Factor
Natural		( $\mu\text{R/h per pCi/g}$ )
	$^{40}\text{K}$	0.179
	$^{238}\text{U}$ -Series	1.82
	$^{232}\text{Th}$ -Series	2.82
		( $\mu\text{R/h per mCi/km}^2$ ) $\times 10^{-3}$
Man Made	$^{137}\text{Cs}$	4.29
	$^{58}\text{Co}$	7.25
	$^{60}\text{Co}$	18.0
	$^{54}\text{Mn}$	6.3
	$^{134}\text{Cs}$	11.4

TABLE XX

POST-OPERATIONAL LABORATORY RESULTS USED IN CALCULATION OF  
DOSE RATE ( $\mu\text{R/h}$ ) 1 METER ABOVE GROUND

LOCATION	$^{40}\text{K}$	$^{238}\text{U}$ -series	$^{232}\text{Th}$ -series	$^{137}\text{Cs}$	TOTAL GAMMA-RAY DOSE RATE	COSMIC RAY*	GRAND TOTAL <sup>†</sup>
Foxbird Island (soil)	1.30	1.24	2.53	.22	5.29	3.60	8.89
Eaton's Farm (soil)	3.14	2.00	3.10	.12	8.36	3.60	11.96
Bailey's Farm	3.15	2.00	3.10	.05	8.30	3.60	11.90
Young's Creek (soil)	3.47	1.45	3.10	.03	8.05	3.60	11.65
Knight Cemetery (soil)	2.02	2.36	4.79	.19	9.36	3.60	12.96
Westport Firehouse (soil)	2.11	1.45	2.25	.15	5.96	3.60	9.56
Chewonki Neck (Camp) (Soil)	2.70	1.27	2.53	.07	6.57	3.60	10.17
Cowseagan Narrows (soil)	2.55	1.82	2.82	.06	7.25	3.60	10.85
Bluff Head (soil)	2.38	2.91	3.38	.06	8.73	3.60	12.33
Foxbird Island (sed.)	3.67	1.64	2.54	.06	7.91	3.60	14.45
Murphy's Corner (sed.)	3.22	1.45	2.53	.02	7.20	3.60	10.80

\*Taken over water in 1971 by W. Van Felt

<sup>†</sup>Including  $^{134}\text{Cs}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$  and  $^{54}\text{Mn}$  ( $2.93\mu\text{R/h}$ )

## APPENDIX D

### A Model for Radionuclides in Oysters and Associated Sediments\*

#### INTRODUCTION

The variation of the concentration of gamma-ray emitting radionuclides has been observed and modeled in a stable population of American oysters (*C. virginica*), and associated estuarine sediments during 12 months at four sites in the Montsweag Estuary in the effluent of the Maine Yankee Nuclear Power Reactor and at a control site in the Damariscotta Estuary. Gamma-ray peaks have been observed at 0.810 MeV ( $^{58}\text{Co}$ ), 1.173 and 1.332 MeV ( $^{60}\text{Co}$ ), 1.732 ( $^{40}\text{K}$ ), 0.835 MeV ( $^{54}\text{Mn}$ ), 0.765 MeV ( $^{95}\text{Nb}$ ), 0.238 MeV ( $^{212}\text{Pb}$ ), and 0.662 MeV ( $^{137}\text{Cs}$ ) using a Ge(Li) detector. The concentration of the most abundant man-made isotopes  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ , and  $^{54}\text{Mn}$  have been compared at selected sites with a mathematical model for the accumulation and loss of these nuclides by the oysters and sediments.

#### PREVIOUS WORK

Radionuclide accumulation and loss have been studied in oysters by Seymour<sup>(18)</sup>, Jefferies and Preston<sup>(19)</sup>, Naidu and Seymour<sup>(20)</sup>, Nelson and Seymour<sup>(21)</sup>, Wolf<sup>(22)</sup>, Lowman, Rice and Richards<sup>(23)</sup>; and radionuclide accumulation has been studied in sediments by Heft, Phillips, Ralston, and Steel<sup>(24)</sup>, Noshkin and Bowen<sup>(25)</sup> and Lentsch, et. al.<sup>(26)</sup> among others. Models, field studies and laboratory studies of accumulation and depuration have been undertaken for radionuclides in reactor effluent for  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  in the marine clam, Harrison<sup>(27)</sup>;  $^{58}\text{Co}$  in the mussel, *Mytilus edulis*, Shimizu et. al.<sup>(28)</sup>; Teleost and Elasmobranch fish, Pentreath<sup>(29)</sup>, and for several organisms by Lowman, et. al.<sup>(30)</sup>.

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Mathematical models for accumulation of radionuclides, are usually based on a constant source of radionuclides, a condition which is generally convenient and applicable for laboratory studies however, must provide for the time variation of the sources of radionuclides since in the case of nuclear reactors the liquid, radionuclide effluents are usually released according to some schedule with waiting periods between releases, as discussed by Heft et.al.<sup>(24)</sup>. This variation of source intensity can be included in the mathematical model by driving the first order linear differential equation for accumulation and decay with a time varying function representing the release schedule<sup>(1)</sup>. This model corresponds mathematically to an impulsively driven relaxator with characteristic relaxation times for the various mechanisms of accumulation and decay.

#### EXPERIMENTAL DESIGN

Five groups of American oysters were grown: 4 Montsweag Estuary locations (intake of the reactor S1, outflow of the reactor S2, upper cove S3 and 0.58 km, N., Long Ledge S4, 1.29 km. S.) and at a control site located on the Damariscotta Estuary S5. At approximately two month intervals, the live oysters were removed from their trays, and transported to the Ge(Li) detector enclosed in a low background shield. Approximately 1 liter of live oysters were counted for 5000 seconds and the resulting data were computer processed using the Compton continuum subtraction method<sup>(8)</sup>. After counting, the oysters were returned to their original locations. The concentrations of radionuclides were determined using standard techniques to calibrate the multichannel analyzer-detector-shield system.

Seven sediment sites were chosen (intake sediment M1, outflow M2, Upper cove M3, Long Ledge M4, Murphy Corner, M5, 2.98 km S.W., Oak Island M6, 2.90 km S.W., and Bluff Head M7, 4.39 km S.W.) At approximately two month intervals, 2 Kg sediment samples were collected at each of the sites and gamma-ray analyzed in the same manner as the oysters.

THEORY

The uptake of radionuclides may be described by a first order linear differential equation

$$\frac{dN}{dt} = -\lambda N + R(t), \tag{1}$$

where  $\frac{dN}{dt}$  is the rate of increase in atoms of nuclide N,  $\lambda N$  is the rate of loss due to radioactive decay, and  $R(t)$  is the rate of introduction of nuclides from an external source (i.e. the nuclear reactor release schedule) Depuration is included by writing a term  $\lambda_p N$  which describes the ("Biological Decay") observed experimentally by Seymour<sup>(18)</sup>.

$$\frac{dN}{dt} + \lambda N + \lambda_p N = R(t). \tag{2}$$

The solution to Eq. 1 may be written

$$N = e^{-\lambda t} \int e^{\lambda t} R(t) dt + ce^{-\lambda t}. \tag{3}$$

We assume that releases of nuclides are made on a sequence of m times,  $\{t_1, t_2, t_3, \dots, t_m\}$  and the amount of nuclide released is given by a function  $f(t)$  which for times greater than or equal to  $t_1$  but less than  $t_2$  is given by  $f_1 \delta(t-t_1)$ , and for times greater than or equal to  $t_2$  but less than  $t_3$  by  $f_2 \delta(t-t_2)$  and so on up to times greater than  $t_m$ . The fraction of the nuclide which was released and is retained is given by U, so that for the accumulation  $N(t)$

$$N(t) = e^{-\lambda t} \int_0^{t_1+\epsilon} e^{\lambda t} f_1 \delta(t-t_1) U dt + \dots$$

$$+ e^{-\lambda t} \int_{t_m-1+\epsilon}^{t_m+\epsilon} e^{\lambda t} f_m \delta(t-t_m) U dt + c e^{-\lambda t} \quad 5)$$

If U. is a constant ratio of retention for all time, we can construct a table of functions for the times between the release time.

$0 \leq t \leq t_1$	$N(t) = ce^{-\lambda t}$	
$t_1 \leq t \leq t_2$	$N(t) = Uf_1 e^{-\lambda(t-t_1)} + ce^{-\lambda t}$	6)
$t_2 \leq t \leq t_3$	$N(t) = Uf_2 e^{-\lambda(t-t_2)} + Uf_1 e^{-\lambda(t-t_1)} + ce^{-\lambda t}$	

These equations may be interpreted as a gradual decay of isotopes from the release-time until the next release-time. Sudden increases occur at each release-time. Graphs of two typical cases are shown in Fig. 5, which shows the results for a half life ( $^{58}\text{Co}$ ) comparable to two release intervals, as well as a half life ( $^{60}\text{Co}$ ) comparable to 60 release intervals.

#### OYSTER RESULTS

The concentration expressed in pCi of the radionuclide  $^{58}\text{Co}$  is shown per gram of oysters in Fig. 6. The broken lines represent the experimental values for this isotope in the four Montsweag Estuary sites and the Damari-scotta Estuary control site from June 73 through July 74. The theoretical results are shown with a solid line from May 73 through June 74. The same peak found in the theory is evident at the outflow, Long Ledge and intake sites and a small increase may be seen in the control site in October. The best agreement occurs (comparison may be made) between the outflow site and the theory. It is evident that the oysters show a faster decrease in activity in Dec. 73 through Feb. 74 than that predicted by theory. This may be

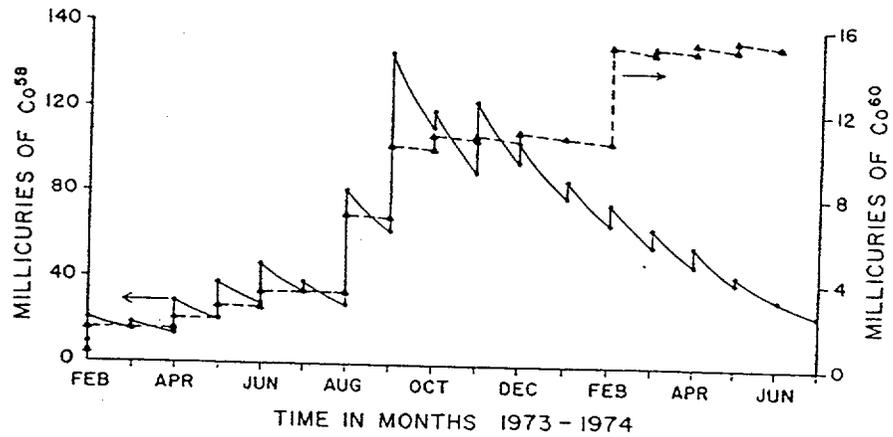


Figure 5. Calculated Accumulation of  $^{58}\text{Co}$  and  $^{60}\text{Co}$  as a Function of Time

The solid line shows the theoretical accumulation of  $^{58}\text{Co}$  in millicuries versus month of year from Feb. 1973 through July, 1973. The dashed line shows the theoretical accumulation of  $^{60}\text{Co}$  in millicuries versus month of the year from Feb. 1973 through June, 1974.

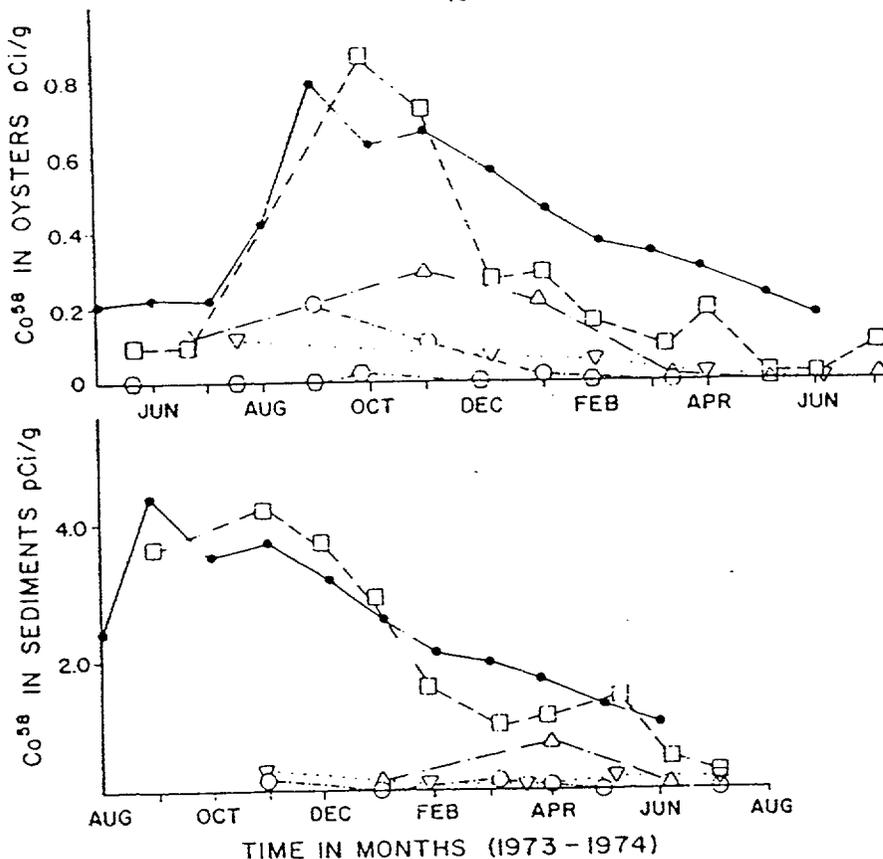


Figure 6. Calculated and Measured Accumulation of  $^{58}\text{Co}$  as a Function of Time

The upper half shows the radionuclide  $^{58}\text{Co}$  in oysters at site S1 (intake) triangle and dot-dash, site S2 (outflow) square and dash, site S3 (Upper cove) circle and dot-dash, site S4 (Long Ledge) triangle and dot, SC (control site) hexagon dash-dot-dot, and theoretical curve for outflow site with filled circle and solid line, versus time in months, for the years 1973-1974. The lower half shows the radionuclides  $^{58}\text{Co}$  in sediments at sediments sites M1 (intake) with triangle dash-dat, M2 (outflow) with square dash, M3 (Upper cove) circle dash-dot, M4 (Long ledge) with triangle dot and theoretical curve for outflow site with cricle solid line versus time in months from Aug. 1973-July, 1974:

explained as the depuration of isotopes from the oysters during these months.

The concentration of the radionuclide  $^{54}\text{Mn}$  for the outflow site was found to be practically flat during this time and compare favorably with theory within statistical errors in the measurement. In addition to the  $^{58}\text{Co}$  and  $^{54}\text{Mn}$  results, small amounts of  $^{60}\text{Co}$  are observed in Oct. 73 at the outflow, Jan. 74 at Long Ledge and Oct. 73 and Jan. 74 at Upper Cove. Small amounts of  $^{54}\text{Mn}$  is also observed on July 73, Dec. 73 and Jan. 74 at Long Ledge and Jan. 74 at the control site. The naturally occurring isotope  $^{40}\text{K}$  is observed at all sites for all measurements and its variation is correlated with variation in the salinity.

#### SEDIMENT RESULTS

The concentration expressed in pCi of  $^{58}\text{Co}$  per gram of sediments is shown in Fig. 6. The broken lines represent the experimental values for this isotope in four Montsweag Estuary sites from Sept. 73 through July 74. The solid line represents the theory for the time from Aug. 73 through June 74. The best comparison can be made from the outflow site which has the largest concentration and shows a broad peak in the months Sept. 73 through Jan. 74. The decrease of  $^{58}\text{Co}$  during Jan - Feb. 74, is more rapid than theory predicts and represents depuration due to loss of radioactivity from the sediments. The other sites had a factor of 10 less isotope than this site, and insufficient data for further conclusions.

The radionuclides  $^{40}\text{K}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{54}\text{Mn}$  observed in the outflow sediment may be averaged, and compared. These radionuclides are expressed in picocuries/gram of sediment and are averaged for monthly values for the time period Aug. 73 - March 74. The average values in order of decreasing concentration are  $^{40}\text{K}$ , 5.7 pCi/gm,  $^{58}\text{Co}$ , 4.9 pCi/gm,  $^{60}\text{Co}$ , 0.8 pCi/gm,

and  $^{54}\text{Mn}$ , 0.2 pCi/gm. All of these isotopes have maximum values in the month of Nov. 73. Most of the isotopes show a decrease in the month of Dec. 73 which is correlated with a salinity drop in the estuary. The decreases in both  $^{60}\text{Co}$  and  $^{54}\text{Mn}$  may be interpreted as the combined effect of radioactive decay and depuration. The effective depuration rate for the outflow sediment during this time was  $150 \pm 30$  days for  $^{60}\text{Co}$ . The depuration rate for the  $^{54}\text{Mn}$  is  $180 \pm 50$  days.

The radionuclides  $^{95}\text{Nb}$ ,  $^{212}\text{Pb}$ ,  $^{137}\text{Cs}$  in the outflow sediment may also be averaged and compared. In order of decreasing concentration, these are  $^{95}\text{Nb}$ , 0.8 pCi/gm,  $^{212}\text{Pb}$ , 0.4 pCi/gm and  $^{137}\text{Cs}$ , 0.2 pCi/gm. These radionuclides show a maximum during the month of Nov. 73 for  $^{95}\text{Nb}$  and March 74 for the  $^{212}\text{Pb}$  and  $^{137}\text{Cs}$ . Since the initial values of  $^{212}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{95}\text{Nb}$   $^{95}\text{Zr}$  in soil and sediments measured prior to the plant operations were large, comparison with theory is not attempted for these nuclides. The sediment sites, M5 M6, and M7 showed only  $^{40}\text{K}$ ,  $^{212}\text{Pb}$  and  $^{137}\text{Cs}$ .

#### DETERMINATION OF UPTAKE RATIO U

Since theory and experiment seem to agree, it is possible to divide the radionuclide in oysters or sediments (Fig. 6) by the calculated accumulation (Fig. 5) to form an uptake ratio U. The values for U indicate that most of the radioactivity is concentrated in the region of outflow, decreasing rapidly with distance. There is also variation of concentration from radionuclide to radionuclide with  $^{58}\text{Co}$  strongest,  $^{60}\text{Co}$  weaker and  $^{54}\text{Mn}$  weakest for both sediments and oysters.

SUMMARY OF APPENDIX D

Oysters have been grown in the effluent of a nuclear reactor and have been used in a longitudinal study of radionuclide uptake and depuration. The radionuclides  $^{58}\text{Co}$ ,  $^{60}\text{Co}$  and  $^{54}\text{Mn}$  have been observed experimentally in the oysters and associated sediments. At selected sites, the variation of radionuclides in both oysters and sediments was found to be in good agreement with predictions of a mathematical model, which incorporates radioactive decay and a time dependent driving source of radionuclides due to the liquid effluent release from the reactor. Values for an average uptake ratio  $U$ , are then calculated for selected sites. Maximum values of  $^{58}\text{Co}$  of 800 pCi/kg results in an annual dose rate of 0.27 mrem/year. Maximum values of  $^{54}\text{Mn}$  of 12 pCi/kg results in an annual dose rate of 0.004 mrem/year.

PRE-OPERATIONAL LABORATORY WATER GAMMA-RAY ANALYSIS  
OF THE DISSOLVED COMPONENT

APPENDIX E

TABLE XXI

LOCATION	TYPE	DATE COLLECTED	DATE COUNTED	<sup>40</sup> K gm/l±2σ	<sup>137</sup> Cs pCi/l±2σ	<sup>222</sup> Rn pCi/l±2σ
Foxbird Island	estuarine	6/13/72	6/22/72	0.18 ±0.06	<2.0	<4.0
Eaton Farm	well	/13/72	6/22/72	<0.10	<2.0	<4.0
Bailey Farm	well	6/13/72 6/13/72	6/22/72 6/29/72	<0.10 <0.10	<2.0 <2.0	81.0 ± 13.0 <4.0
Young's Creek	surface	6/13/72	6/22/72	<0.10	<2.0	<4.0
Chewonki Neck (Camp)	well	6/13/72 6/13/72	6/22/72 6/27/72	<0.10 <0.10	<2.0 <2.0	98.2 ± 19.2 <4.0
Cowseagan Narrows	estuarine	6/13/72	6/22/72	0.19 ±0.06	<2.0	<4.0
Bluff Head	estuarine	6/13/72	6/22/72	0.16 ±0.06	<2.0	<4.0

(All samples were counted for 50 minutes in a 3.5 liter Marinelli beaker geometry)

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217

High Rad Key # 59 is broken.

Notified by D. Lovitt that soil sample taken from Foxbird Island Sample Station had licensed material. Called B. Gann to inform him of discovery. Dispatched technician to area to verify disc. rates. Notified PSS.

⑧ Soil sample station reading 50  $\mu$ R/hr. Area has been flagged by GTS Duratek.

⑨ All high rad keys accounted for. Ops has keys #1 and #62.

RP has keys # 53, 59, 60, 61, 74, 75, 76, MMC #14 and MMC #20.

⑩ Relieved by A. Salib.

Scott Clabey

Date 31 JAN 98 Shift 1500-2300

Technician SALIB

Duratek running  Y /  N

RMS Monitor out of service: PVS PART, SERV H<sub>2</sub>O, SERV BLDG

① Reviewed NO RCSIB, log; relieved watch. AS

② Ops turnover; no items of Rad Con concern. AS

③ Countroom set up. AS

④ Other than hand-held meters daily v/s complete as req'd. AS

⑤ As per Jamie Mallon: (telecom) Security will dress out in PCs and enter CTHT in order to inspect sections of piping that are currently capped / blank flange. (Piping to be used for chem decon.) AS

⑥ High rad key inventory SST: #1, 62 @ OPS

# 53, 59, 60, 61, 74, 75, 76, MMC14, MMC-20 @ HP CKPT. AS

⑦ Relieved by Van Ziemann

Andrew Salib

Date 1-31-98 Shift 2300-0700

Technician Ziemann

Duratek running  Y /  N

RMS Monitor out of service: Service Bldg APD

Relieved the watch, receive turnover

Ops turnover no HP item

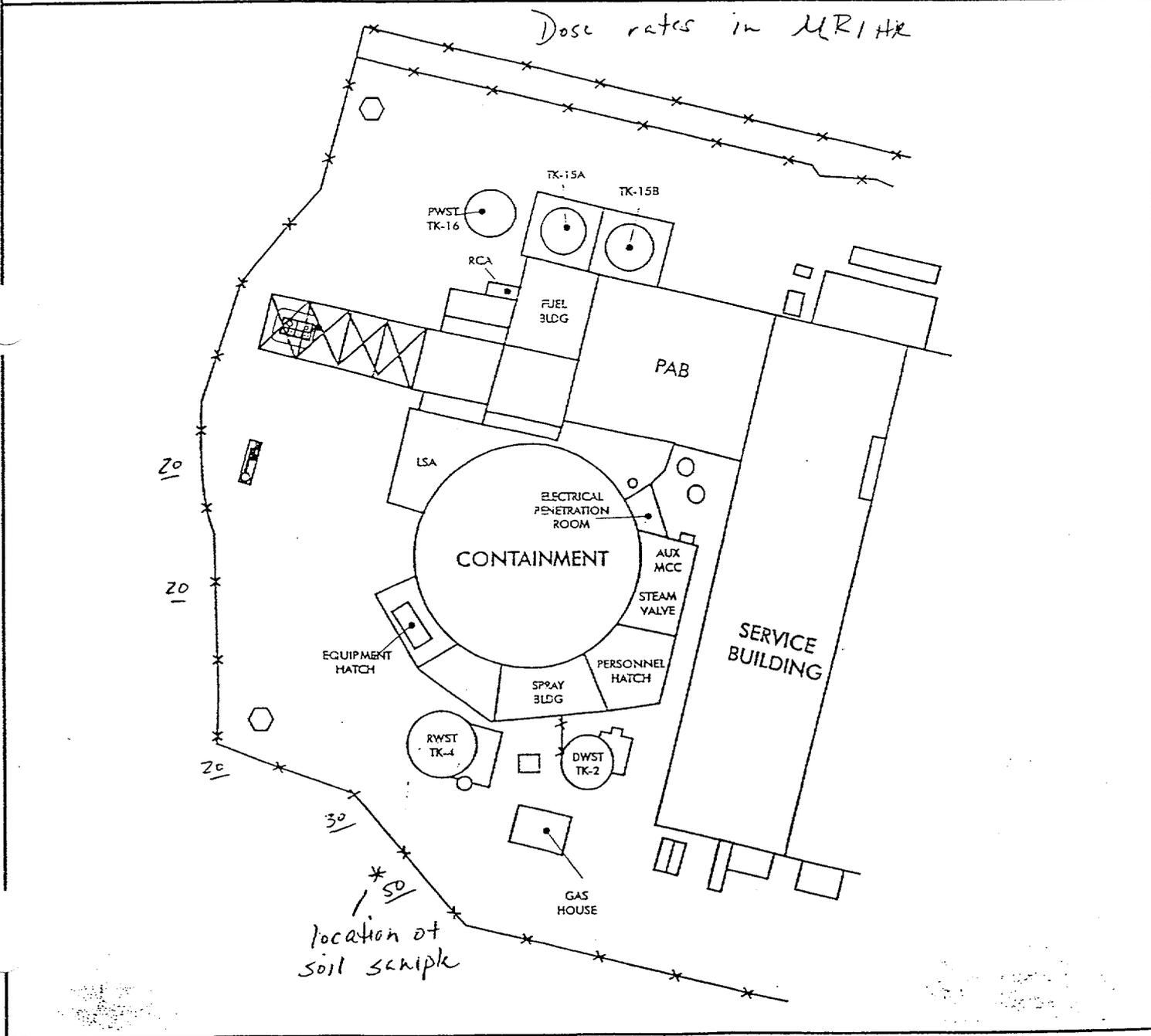


Map#: OSA-002    Date: 1-30-98    Time: 1500    Reactor Pwr %: 0    Tech File Number: 19.20.11.10    RWP's Used: 98-1    Dose Received: 50 mR

Surveyor Name: (Printed) *Ambrose*    Surveyor Name: (Signature) *Ambrose*    Location/Job Description: Outside Restricted Area / Site Characterization Support

Required R.P. Review / Date: *Scott Cluby 1/30/98 MA*    Required ALARA Supervisor Review / Date: *NIA*    REASON FOR SURVEY:  ROUTINE<sup>1</sup>     JOB-COVERAGE<sup>1</sup>     SHIELDING<sup>1,2</sup>     OTHER<sup>1</sup> (Specify):

INSTRUMENTS USED				CONTAMINATION RESULTS								KEY:
MODEL	SERIAL #	CAL DUE	MDA	SAMPLE #1	RESULTS	SAMPLE #1	RESULTS	SAMPLE #1	RESULTS	SAMPLE #1	RESULTS	
Bicron	B297W	7-22-98	N/A									<ul style="list-style-type: none"> <li>Contact exposure rates denoted by: *</li> <li>Smear locations denoted by: ⊙</li> <li>Boundaries or barriers denoted by: -x-x-</li> <li>Dose rates denoted by: /</li> <li>Large area smears denoted by: [ ]</li> <li>Air sample location denoted by: [ ]</li> </ul>
												Sample Continuation Sheet Used: <input type="checkbox"/> YES



Map#: MSC-001    Date: 1-30-98    Time: 1300    Reactor Pwr %: 0    Tech File Number:    RWP's Used: 98-1    Dose Received: 0 mR

Surveyor Name: (Printed) Ambrose    Surveyor Name: (Signature) [Signature]    Location/Job Description: 0/s Gate 6 / Site Characterization Support

Required R.P. Review / Date: [Signature] 1/30/98    Required ALARA Supervisor Review / Date: N/A    REASON FOR SURVEY:  ROUTINE<sup>1</sup>     JOB-COVERAGE<sup>1</sup>     OTHER<sup>1</sup> (Specify): Site Characterization  
 SHIELDING<sup>1,2</sup>

INSTRUMENTS USED				CONTAMINATION RESULTS								KEY:
MODEL	SERIAL #	CAL DUE	MDA	SAMPLE #1	RESULTS	SAMPLE #1	RESULTS	SAMPLE #1	RESULTS	SAMPLE #1	RESULTS	
M-3	72455	3/23/98	1000									• Contact exposure rates denoted by: *
												• Smear locations denoted by: ⊙
												• Boundaries or barriers denoted by: -x-x-
												• Dose rates denoted by: <u>  </u>
												• Large area smears denoted by: <u>  </u>
												• Air sample location denoted by: <u>  </u>
												Sample Continuation Sheet Used: <input type="checkbox"/> YES

Survey of equipment used to drill 16" into soil @ Bailey's Point 0/s  
Protected Area fence

- <math>MDA</math> on all masslms taken at drill base and push pins
- No detectable counts above background on direct frisk of all equipment used for drilling

**HSA ID# 111**

\*\*\*\*\*  
\*  
\* GAMMA SPECTRUM ANALYSIS \*  
\*  
\*\*\*\*\*

ERRA APOGEE V2.4

Canberra Industries, Inc.

22-JAN-98

15:13:19

ANALYSIS PARAMETERS

Spectrum file number : 100.0 Sample no. : 1.0  
MDA unit number : 1 ADC unit number : 1.0  
Detector number : 1 Geometry number : 2  
Search threshold 1 : 2.0 Search threshold 2 : 3.0  
Search FROM channel : 50 Search TO channel : 4095  
Ed energy tolerance : 1.0 Order of background : linear  
Smoothing factor : 0 Random sum corr : disabled  
BRA parameter : 0 Baseline channels : disabled

Confidence threshold index : 0.100  
Confidence levels LLD : 1.645 (95.0%) MDA : 1.645 (95.0%)

Analysis library : SPF\$LIBRARY;SPFANL.LIB;1  
Background subtract : enabled

Sample description : TK-37 SLUDGE Analyzed by: SJW

Wa size : 1.000000E+00 EA Conv. factor : 1.000  
Standard size : 1.000000E+00 EA

Sample taken on : 22-JAN-98 at 14:30:00  
Collect started on : 22-JAN-98 at 15:01:03  
Decay time : 31.0 minutes

Live time : 1000.0 seconds real time : 1000.0 seconds  
Dead time : 0.00 %

Energy calibration used done on 1 / 22 / 1998  
Efficiency calibration used done on 11 / 8 / 1996

\*\*\* P E A K   S E A R C H   R E P O R T   \*\*\*

22-JAN-98

15:18:19

first search channel : 50  
 last search channel : 4095  
 first significance limit for found peaks: 2.00  
 second significance limit for found peaks: 3.00  
 average Gaussian peak width (in channels): 1.49

i	peak channel	peak energy	signif of peak	check-1 signif	check-2 shape	accept channels	number
1	68.817	44.4	2.163	small	check		
2	93.588	46.8	2.328	small			
3	114.000	57.0	2.044	small	check		
4	119.077	59.6	2.929	small			
5	127.806	63.9	2.609	small	check		
6	149.614	74.8	4.709			150	1
7	154.489	77.3	5.858			154	2
8	174.149	87.1	3.659			174	3
9	180.232	90.1	2.017	small			
10	234.968	117.5	2.386	small			
11	259.024	129.5	2.871	small			
12	372.424	186.1	2.004	small	check		
13	477.472	238.6	10.683			477	4
	483.052	241.4	2.359	small			
	513.592	256.7	2.485	small			
16	561.856	280.8	2.043	small			
17	590.734	295.2	5.308			591	5
18	600.825	300.3	2.265	small			
19	676.795	339.3	3.035			677	6
20	704.117	351.9	5.460			704	7
21	770.329	385.0	2.579	small			
22	806.879	403.3	2.067	small	check		
23	925.883	462.8	2.343	small			
24	1022.237	510.9	2.655	small			
25	1166.680	583.1	5.880			1167	8
26	1219.129	609.3	5.654			1219	9
27	1246.789	623.2	2.110	small			
28	1455.313	727.4	2.894	small			
29	1536.929	768.2	2.432	small	check		
30	1545.324	772.4	2.225	small	check		
31	1721.545	860.5	2.060	small	check		
32	1822.992	911.2	4.343		check	1823	10
33	1930.674	965.0	2.526	small			
34	1938.616	968.9	3.238			1939	11
35	2241.815	1120.4	2.630	small			
36	2477.028	1237.9	2.319	small	check		

\*\*\* P E A K F I T R E P O R T \*\*\* 22-JAN-98 15:18:19

pk	nuclide(s)	centroid channel	energy keV	FWHM keV	net counts	error %	gamma per second	error %
M 1	PB-214 NATURAL	149.61	74.82	1.08	136.2	10.09	4.0	14.85
B 1			74.82		2.1	20.88		
M 2	PB-214	154.49	77.26	1.08	211.9	7.73	6.1	12.20
M 3	NP-237 EU-155	174.15	87.08	1.19	44.6	24.80	1.2	26.33
	NATURAL CD-109							
B 4	NATURAL	477.47	238.64	1.21	406.0	5.50	15.1	6.90
			238.64		4.0	10.39		
B 5	PB-214	590.73	295.25	1.33	79.6	12.24	3.4	12.75
B 6	NATURAL	676.79	338.26	1.44	40.3	19.01	2.0	19.26
B 7	PB-214	704.12	351.92	1.45	105.6	10.60	5.6	11.04
B 8	NATURAL	1166.68	583.13	1.57	130.4	9.08	11.1	9.73
B 9	BI-214	1219.13	609.35	1.53	1.1	21.38		
			609.35		71.6	12.59	6.1	13.06
B 9			609.35		0.8	24.59		
B 10	NATURAL	1822.99	911.15	1.74	44.4	16.27	5.3	15.46
B 11	NATURAL RU-105	1938.62	968.93	1.74	30.4	22.17	4.2	22.30

M - Peak is a multiplet

B - Environmental background peak. Will be subtracted from the peak above.

Background subtraction performed using file SPF#DATA: BK0001.MC1

Ground description: 50K BKG 11/24/77



\*\*\* M D A C A L C U L A T I O N R E P O R T \*\*\*

22-JAN-98

15:18:19

Sample description  
 Analyzed by

:TK-37 SLUDGE  
 :SJW

		----- MDA ( uCi/EA ) -----	
		measured	decay corrected
VA-24	:	MDA : 6.13E-05	6.28E-05
AR-41	:	MDA : 5.27E-05	6.41E-05
CR-51	:	MDA : 3.95E-04	3.96E-04
4N-54	:	MDA : 7.61E-05	7.61E-05
4N-56	:	MDA : 5.78E-05	6.64E-05
CO-57	:	MDA : 3.28E-05	3.28E-05
CO-58	:	MDA : 6.37E-05	6.37E-05
FE-59	:	MDA : 9.71E-05	9.71E-05
CO-60	:	MDA : 7.74E-05	7.74E-05
CU-64	:	MDA : 1.23E-02	1.27E-02
VI-65	:	MDA : 2.50E-04	3.22E-04
ZN-65	:	MDA : 1.68E-04	1.68E-04
SE-75	:	MDA : 4.71E-05	4.71E-05
BR-84	:	MDA : 1.22E-04	2.40E-04
CR-85	:	MDA : 1.77E-02	1.77E-02
BR-85	:	MDA : 7.76E-05	7.76E-05
CR-85M	:	MDA : 3.96E-05	4.29E-05
CR-87	:	MDA : 1.01E-04	1.34E-04
CR-88	:	MDA : 1.43E-04	1.62E-04
CR-88	:	MDA : 7.75E-05	7.75E-05
RB-88	:	MDA : 4.03E-04	1.35E-03
RB-89	:	MDA : 8.18E-05	3.30E-04
CR-89	:	MDA : 6.96E-04	6.32E-01
7-91M	:	MDA : 5.19E-05	8.01E-05
BR-91	:	MDA : 1.45E-04	1.50E-04
7-92	:	MDA : 5.05E-04	5.58E-04
BR-92	:	MDA : 1.19E-04	1.35E-04
7-93	:	MDA : 5.33E-04	5.52E-04
7-94	:	MDA : 1.27E-04	4.02E-04
7-95	:	MDA : 2.72E-04	2.20E-03
VB-95	:	MDA : 8.92E-05	8.92E-05
VB-95M	:	MDA : 3.12E-04	3.13E-04
ZR-95	:	MDA : 1.04E-04	1.04E-04
ZR-97	:	MDA : 1.26E-03	1.29E-03
VB-97	:	MDA : 5.45E-05	7.35E-05
4D-99	:	MDA : 3.76E-04	3.78E-04
TC-99M	:	MDA : 3.67E-05	3.69E-05
4D-101	:	MDA : 1.75E-04	7.64E-04
TC-101	:	MDA : 3.70E-05	1.68E-04
TC-102M	:	MDA : 1.56E-04	2.19E-02
RU-103	:	MDA : 4.72E-05	4.72E-05
TC-104	:	MDA : 3.98E-05	1.31E-04
TC-105	:	MDA : 1.76E-04	1.77E-04
TC-105	:	MDA : 1.65E-04	1.79E-04
RU-106	:	MDA : 5.02E-04	5.02E-04
RU-106	:	MDA : 5.02E-04	5.02E-04
AB-108N	:	MDA : 5.31E-05	5.31E-05

DD-109	:	MDA	:	1.53E-03	1.53E-03
AG-110M	:	MDA	:	5.66E-05	5.66E-05
BN-113	:	MDA	:	7.98E-05	7.98E-05
BE-122	:	MDA	:	8.04E-05	8.08E-05
BT 124	:	MDA	:	5.70E-05	5.70E-05
25	:	MDA	:	1.62E-04	1.62E-04
31M	:	MDA	:	2.15E-04	2.18E-04
XE-131M	:	MDA	:	1.58E-03	1.58E-03
BE-131	:	MDA	:	1.19E-04	3.02E-04
I-131	:	MDA	:	4.94E-05	4.95E-05
TE-131	:	MDA	:	4.80E-05	1.14E-04
I-132	:	MDA	:	6.37E-05	7.45E-05
TE-132	:	MDA	:	4.16E-05	4.18E-05
BA-133	:	MDA	:	1.21E-04	1.21E-04
TE-133	:	MDA	:	4.67E-05	2.63E-04
I-133	:	MDA	:	5.58E-05	5.69E-05
XE-133M	:	MDA	:	3.68E-04	3.71E-04
XE-133	:	MDA	:	1.69E-04	1.70E-04
TE-133M	:	MDA	:	1.54E-04	2.28E-04
TE-134	:	MDA	:	1.75E-04	2.92E-04
I-134	:	MDA	:	5.99E-05	9.02E-05
DS-134	:	MDA	:	8.33E-05	8.33E-05
I-135	:	MDA	:	2.34E-04	2.47E-04
XE-135M	:	MDA	:	6.05E-05	2.45E-04
XE-135	:	MDA	:	4.18E-05	4.35E-05
DS-136	:	MDA	:	6.41E-05	6.42E-05
DS-137	:	MDA	:	6.31E-05	6.31E-05
DS-138	:	MDA	:	9.84E-05	1.92E-04
XE-138	:	MDA	:	1.22E-04	5.58E-04
DS-139	:	MDA	:	9.70E-04	9.58E-03
39	:	MDA	:	1.83E-04	2.38E-04
38M	:	MDA	:	6.82E-04	1.14E+00
DE-139	:	MDA	:	3.90E-05	3.90E-05
BA-140	:	MDA	:	1.86E-04	1.86E-04
LA-140	:	MDA	:	9.53E-05	9.55E-05
BA-141	:	MDA	:	8.04E-05	2.61E-04
LA-141	:	MDA	:	2.72E-03	2.98E-03
DE-141	:	MDA	:	7.17E-05	7.17E-05
BA-142	:	MDA	:	2.45E-04	1.83E-03
LA-142	:	MDA	:	9.64E-05	1.21E-04
DE-143	:	MDA	:	1.50E-04	1.51E-04
PR-144	:	MDA	:	4.67E-03	4.67E-03
DE-144	:	MDA	:	2.91E-04	2.91E-04
PR-146	:	MDA	:	1.06E-04	2.60E-04
DE-146	:	MDA	:	6.09E-05	2.87E-04
ND-147	:	MDA	:	1.86E-04	1.86E-04
PR-147	:	MDA	:	1.49E-04	7.24E-04
PM-149	:	MDA	:	1.43E-03	1.44E-03
EU-152	:	MDA	:	9.46E-05	9.46E-05
HF-181	:	MDA	:	5.06E-05	5.06E-05
H-187	:	MDA	:	1.92E-04	1.95E-04
HG-203	:	MDA	:	6.04E-05	6.04E-05
NP-237	:	MDA	:	4.05E-04	4.05E-04
J-237	:	MDA	:	1.38E-04	1.38E-04
NP-239	:	MDA	:	1.45E-04	1.46E-04
341	:	MDA	:	1.46E-04	1.46E-04
54	:	MDA	:	7.03E-05	7.03E-05
E. 35	:	MDA	:	1.50E-04	1.50E-04
NE-74	:	MDA	:	4.42E-05	4.42E-05

**HSA ID# 112**

# Center for Biological Monitoring, Inc.

Sponsor of RADNET: Nuclear information on the Internet

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World Wide Web at <http://home.acadia.net/cbm/Rad.html>

BOX 144 HULLS COVE, ME 04644-0144 207/238-5126

FAX: 207/238-2725 EMAIL: [sbrack@post.acadia.net](mailto:sbrack@post.acadia.net)

*copy - Messner  
Fenn  
Brown  
Hower  
E. Rice  
Maine form  
you agreed  
to develop  
interest  
response to  
this letter  
which we  
may or  
may not  
share  
with others  
MJK*

Fax Cover Sheet

Date: 1/23/98

Number of Pages including cover sheet: 5

To: MARY ANN LYNCH

From: H.G. Brack

Re: Letter to Shirley Jackson

Message: The enclosed letter to Shirley Jackson is intended to alert first the NRC, and then any other interested parties to:

A. Deficiencies in the MYAPC site characterization process and the Duratek site management plan.

B. Controversies pertaining to arbitrary and artificial guideline levels contained in the new DCGLs. Site-specific DCGLs for Maine Yankee have not yet been issued, but for a preview of what's coming down the turnpike consult the Appendices in NUREG-1500.

Recipients of this letter, please be advised that the definition of "contamination" contained in the Jackson letter is extracted from the draft MARSSIM.

DCGL = Derived Concentration Guideline Level

MARSSIM = Multi-Agency Survey and Site Investigation Manual, NUREG-1575

MARSSIM can be viewed on the Internet at <http://www.epa.gov/radiation/cleanup>

Comments, criticisms and additional information welcomed.

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01/23/98

Nuclear Regulatory Commission  
Washington, D.C. 20555-0001

Dear Chairperson Jackson,

This letter is to address some issues which pertain to the Federal Register notice of July 21, 1997, Radiological Criteria for License Termination. This criteria is total effective dose equivalence (TEDE) not exceeding 25 mrem/yr for a maximally exposed person, all radionuclides and all pathways.

An ongoing review of the decommissioning process at the Maine Yankee Atomic Power Company (MYAPC) reveals a series of deficiencies and omissions in the MYAPC site characterization process, particularly with respect to the marine environment which has been historically impacted by the receipt of liquid effluents from MYAPC diffusers at the bottom of Montsweag Bay. The deficiencies and omissions noted originate in part from the *Duratek Site Characterization Management Plan* for MYAPC. In the case of MYAPC, or any decommissioned pressurized water reactor (PWR) or boiling water reactor (BWR), this release criteria mandates systematic pathway analyses for the long-lived isotopes characterizing spent fuel and reactor vessel internal components. The *Duratek* plan makes it clear that this systematic pathway analyses is lacking at MYAPC, and the resulting site characterization is insufficient to meet the criteria for license termination.

There also appear to be a series of generic irregularities and deficiencies within the regulatory literature of the NRC. These raise questions about the validity of the newly issued release criteria and, thus of the ability of the NRC to meet its statutory obligations as described in the Code of Federal Regulations.

**MYAPC: Preliminary observations of deficiencies in site characterization**

Timely, accurate and reliable information about the decommissioning activities at MYAPC is not available to the general public. What little information is available derives from licensee controlled Citizen's Advisory Panel (CAP) monthly meetings. The *Duratek* site management plan has, however, been available for review by CBM and reveals blatant deficiencies in the site characterization process as a precursor to decommissioning activities.

1. The long-standing inadequacies of the annual licensee Radiological Environmental Monitoring Program (REMP) reports are now impacting the site characterization process. These REMPs provide just enough information to identify large areas of the marine environment adjacent to MYAPC as being impacted by plant operations. At the same time, these flawed reports contain a consistent lack of data points such that isocuric characterization of the environmental impact of plant operations has never been possible. They represent the first stage of an NRC licensee failure to meet the statutory obligation of protecting public health and safety, as contained in the Code of Federal Regulations.
2. The *Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) Draft (NUREG-1575)* makes critically clear the important role of "historical site assessment" in evaluating site status in terms of existing contamination (unaffected and affected areas - see Chapter 3). A large body of secondary radiological surveillance reports exist which also document a pattern of plant derived contamination in the marine environment adjacent to MYAPC. This secondary literature ranges from pre- and post-operational analyses of cesium-137 and cobalt-60 done by Charles Hess in behalf of the NRC to a whole series of reports done under the auspices of the Sea Grant program. Some of these other secondary reports include state of Maine monitoring and other reports by Hess and Smith, 1975; Price, Hess and Smith, 1976; Churchill, 1976; Hess, Smith and Price, 1977; McCarthy and Ryder, 1978; McCarthy, Ryder and Antonitis, 1978; Bowen, 1981; Churchill, Hess and Smith, 1980; Lutz and Hess, 1980; Lutz, Inoze and Hess, 1980; and Murray, 1982. In contrast to these many reports in the early years of plant operation, there are no post-1982 independent radiological monitoring reports that the Center for Biological Monitoring can cite which supplement the MYAPC REMPs after this date other than Hess, 1997. None of these older documents appear to play any role in the current Duratek "Site Characterization Management Plan". The result is a grossly inaccurate identification of unaffected as well as affected areas. No credible TEDE can be established for MYAPC until the site characterization process is reviewed and revised.
3. Chapter 6 in MARSSIM discusses field measurement methods and instrumentation and reflects the historical emphasis of the NRC on external radiation as the primary area of concern with respect to acute health effects resulting from radiological contamination. With the advent of the *all radionuclides all pathway clause* of the new release criteria, it is now clear that radiological surveillance must be upgraded to include a consideration of the presence and impact of all the micro-contaminants which characterize long-lived spent fuel wastes (LLSFW): Sr-90, Zr-92, Tc-99, Sn-126, I-129, Cs-137, Pu-238, Pu-239/240, Pu-241, and Am-241. Most of these isotopes are difficult to characterize alpha and pure beta emitters. Comprehensive pathway analyses of these radionuclides is expensive but time consuming and must be done within a laboratory setting. Information is not yet available as to the accuracy of the on-site laboratory recently established at MYAPC and its ability to characterize these isotopes. Questions however have already been raised about the accuracy and scientific reliability of sample collection methods.
4. The controversial presentation made by the licensee at the December CAP meeting denoting extremely limited areas impacted by plant operations combined with the grossly insufficient Duratek site management plan sampling for the marine

environment indicates that insufficient data points will be available to construct a grid, as suggested in MARSSIM. The consequence of insufficient data is the clear inability of the NRC or the licensee to evaluate not only the past environmental impact of plant operations but also the ongoing impact of decommissioning activities. The postponement of accurate site characterization until a final site survey is executed (in the case of MYAPC, 2004) is a possible interpretation of the MARSSIM, which is not a site-specific guide. The MARSSIM makes many references to the possibility of detailed site characterization prior to decommissioning but it also makes it clear that this detailed survey is an option. The failure to execute a thorough survey prior to decommissioning activities at MYAPC is a clear violation of the Code of Federal Regulations general statutory requirement to protect public health and safety.

5. Another deficiency in the Duratek site characterization plan is the choice of the location of the "reference area" as the control for evaluating contamination in "affected" areas. The reference area chosen is a terrestrial location which may have been impacted by plant operations. Site-specific factors such as the location of the two diffusers at the bottom of Montsweag Bay make it mandatory that a second reference or control area be chosen in a marine environment located well away from the bays and estuaries near the Wiscasset facility. A terrestrial reference area is insufficient as a control for analyses done within a marine environment.
6. The net result of the gross deficiencies in historical site assessment of plant operations in the marine environment as well as of the current site characterization process is the undermining of the credibility of radiological criteria as the bases for license termination. In the case of MYAPC or any other NRC licensed facility, the errors and omissions in historical and contemporary site characterization are cumulative. Superficial, one-dimensional characterization of residual radioactivity with over reliance on external radiation exposure from gamma releasing radioisotopes results in the failure to meet the *all radionuclides all pathways* clause of the release criteria (e.g. see the old guidelines in AEC Rule 1-86). Significant additional pathway analyses are needed to characterize the marine environment as a possible repository of LLSFW resulting from micro-contamination from several incidents of spent fuel cladding leakage, grid to rod fretting within the spent fuel assemblies and the general impact of overall operations at the Maine Yankee Atomic Power Company (1972 - 1998). In particular, there is an obvious need for a greatly expanded program of sediment sampling in areas impacted by the liquid diffusers, which due to the strong currents which sweep the bottom of Montsweag Bay have the potential to cover many square miles of bays and estuaries near MYAPC.

Upcoming reports containing specific data collected by the licensee and its contractor Duratek during the site characterization process will hopefully provide additional information about these deficiencies.

#### Irregularities and omissions in NRC regulatory guides

Specific NRC guidelines pertaining to decommissioning NRC licensed facilities begin with the U.S. Atomic Energy Commission Regulatory Guide 1.86 and include NUREG publications 5512, 5849, 1444, 1496 vols. 1 and 2, 1500, 1501, 1505, 1507, MARSSIM and 10 CFR Part 20, et. al. There appear to be a series of discrepancies and omissions in

these regulatory guides which culminate in some questionable definitions in MARSSIM, the most recent of the regulatory guides. In particular, the definition of *derived concentration guideline levels* (DCGL) appears to be sufficiently flawed as a tool for evaluating residual radioactivity as to undermine the credibility of the release criteria as described in 10 CFR Part 20, et. al. The details leading up to this conclusion will be presented in another CBM report this spring. Pre-existing irregularities in regulatory literature and definitions within MARSSIM culminate in an artificial and controversial definition of DCGL. These include the methodology used to determine background radiation, the concept of 5 mrem as indistinguishable from background, minimum detectable concentration (MDC), elevated measurement comparison (EMC), data quality assessment (DQA) and residual radioactivity. A definition that is emblematic of the historical deficiencies in the regulatory guides is that of *contamination* within MARSSIM: "the presence of residual radioactivity in excess of levels which are acceptable for release of a site or facility for unrestricted use" (pg. GL-4). While site-specific DCGLs are not yet available for MYAPC, the appendices in NUREG-1500 are the closest regulatory guides come to setting derived concentration guideline levels. In Appendix B-1, the soil concentration of cesium-137 for the residential scenario necessary to provide an exposure of 24 mrem/yr (just below the TEDE of 25 mrem/yr and thus not *contamination??*) is 17,120 pCi/kg.; for americium-241 2,928 pCi/kg; etc. This gives just a hint of the problems coming down the turnpike in the new DCGLs. The new FDA derived intervention level for americium-241 contamination in foodstuffs is 2 Bq/kg for infants (= 54 pCi/kg). The problems with the DCGLs provide an example of why it is absolutely essential that the NRC re-schedule public hearings for the decommissioning process at all NRC licensed facilities.

The deficiencies and omissions in NRC regulatory literature originate in a fundamental epistemological quandary: the conflict between easily determined dose-related non-stochastic acute health effects of ionizing radiation versus the more controversial stochastic (random e.g. cancer, hereditary defects) health effects of low-levels of chronic contamination. This issue is now manifest in the new DCGLs and NRC's release criteria of a TEDE of 25 mrem/yr. Pathway analyses for long-lived isotopes characterizing spent fuel wastes have been traditionally overlooked in most 20th century environmental radiological monitoring reports. Implicit in the new NRC release criteria is the necessity for more comprehensive analyses of the presence of these isotopes in all pathways, including the marine pathway at MYAPC. The dilatory MYAPC site characterization process is insufficient to meet the obligations implicit in the new site release criteria. Application of spurious DCGLs as they become codified will only exacerbate this problem not only for MYAPC but for all other NRC supervised facilities.

Yours truly,



H. G. Brack

**HSA ID# 113**

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

Woods Hole Oceanographic Institution, MA

Prepared for

Nuclear Regulatory Commission  
Washington, DC

Mar 81

U.S. Department of Commerce  
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# The Environmental Behavior of Transuranic Nuclides Released from Water Cooled Nuclear Power Plants

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Prepared by V. T. Bowen

Woods Hole Oceanographic Institution

Prepared for  
U.S. Nuclear Regulatory  
Commission

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

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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  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{242}\text{Cm}$  and  $^{244}\text{Cm}$ , 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.

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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:

- $^{55}\text{Fe}$ : Labeyrie, Livingston and Gordon, 1975. Nucl. Instr. and Meth. 128, 575-580.
- $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  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  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{242}\text{Cm}$ , and  $^{244}\text{Cm}$ ; 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 9: Wiscasset, Maine, Area: Surficial Sediment Radiochemistry I.
- Table 10: Wiscasset, Maine, Area: Sediment Cores Radiochemistry II.
- Table 11: Wiscasset, Maine, Area: Environmental Samples - Biota I.
- Table 12: Wiscasset, Maine, Area: Environmental Samples - Biota II.
- Figure 3: Sampling Locations around the Pilgrim Nuclear Reactor, Plymouth, Mass.
- Table 13: Plymouth, Mass., Area: Water and Sediment Radiochemistry.
- Table 14: Plymouth, Mass., Area: Environmental Samples - Biota.
- Table 15: Biota Samples for Comparison to Plymouth Area (Table 14A & B).

TABLE 1

MILLSTONE POINT REACTOR #1 MONTHLY DISCHARGE ACTIVITY

Month and Year	Stored Volume (liters)	<sup>55</sup> Fe mCi (DOC) <sup>1</sup>	<sup>60</sup> Co mCi	<sup>134</sup> Cs mCi (DOC) <sup>1</sup>	<sup>137</sup> Cs mCi	<sup>238</sup> Pu nCi	<sup>239,240</sup> Pu nCi	<sup>241</sup> Am nCi	<sup>242</sup> Cm nCi (DOC) <sup>1</sup>	<sup>244</sup> Cm nCi
Jan. 1977	1.07x10 <sup>6</sup>		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			

<sup>1</sup> 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)	<sup>55</sup> Fe mCi (DOC) <sup>1</sup>	<sup>60</sup> Co mCi	<sup>134</sup> Cs mCi (DOC) <sup>1</sup>	<sup>137</sup> Cs mCi	<sup>238</sup> Pu nCi	<sup>238,239</sup> Pu nCi	<sup>241</sup> Am nCi	<sup>242m</sup> Cm nCi (DOC) <sup>1</sup>	<sup>244</sup> Ym nCi
Jan. 1977	640,422			0.30±0.05	0.61±0.09	<40	60±80	<130	<65	<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			

<sup>1</sup> DOC = data decay corrected to date of collection of sample

TABLE 3  
MAINE YANKEE, PERIODIC DISCHARGE ACTIVITY

<u>Date - 1977</u>	<u>Time</u>	<u>Volume (liters)</u>	<u><sup>55</sup>Fe mCi (DOC)<sup>1</sup></u>	<u><sup>134</sup>Cs mCi (DOC)<sup>1</sup></u>	<u><sup>137</sup>Cs mCi</u>	<u><sup>238</sup>Pu nCi</u>	<u><sup>239,240</sup>Pu nCi</u>	<u><sup>241</sup>Am nCi</u>
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 1400	45,526 40,800						
16 August	1200 1752	35,200 38,016		0.148±0.008	0.51 ±0.01	0.11 ±0.14	3.7 ±0.5	
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 1700	46,934 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

<u>Date - 1978</u>	<u>Time</u>	<u>Volume (liters)</u>	<u><sup>55</sup>Fe mCi (DOC) <sup>1</sup></u>	<u><sup>134</sup>Cs mCi (DOC) <sup>1</sup></u>	<u><sup>137</sup>Cs mCi</u>	<u><sup>238</sup>Pu nCi</u>	<u><sup>238,239</sup>Pu nCi</u>	<u><sup>241</sup>Am nCi</u>
3 January	1500	42,229		0.029±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	42,106						
	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

<u>Date - 1978</u>	<u>Time</u>	<u>Volume (liters)</u>	<u><sup>55</sup>Fe mCi (DOC)<sup>1</sup></u>	<u><sup>134</sup>Cs mCi (DOC)<sup>1</sup></u>	<u><sup>137</sup>Cs mCi</u>	<u><sup>238</sup>Pu nCi</u>	<u><sup>238,239</sup>Pu nCi</u>	<u><sup>241</sup>Am nCi</u>
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

<sup>1</sup> 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:

$^{55}\text{Fe}$ : At the time we prepared this proposal, it was the prevailing wisdom at NRC that "reactors produce  $^{59}\text{Fe}$  but not  $^{55}\text{Fe}$ ", and we were accordingly directed to omit  $^{55}\text{Fe}$  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  $^{59}\text{Fe}$  without simultaneously producing  $^{55}\text{Fe}$ ; 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  $^{55}\text{Fe}$  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  $^{55}\text{Fe}$  are being released from each of these reactors, the  $^{55}\text{Fe}$  to  $^{60}\text{Co}$  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  $^{55}\text{Fe}$  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.

$^{134}\text{Cs}$ : $^{137}\text{Cs}$ : Both  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are produced, and released, in considerable amounts in reactor operations. The very different half-lives of the two nuclides ( $T_{1/2}$   $^{134}\text{Cs}$  = 2.1y;  $T_{1/2}$   $^{137}\text{Cs}$  = 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,  $^{134}\text{Cs}$  to  $^{137}\text{Cs}$ , 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.

$^{238}\text{Pu}$ : $^{239,240}\text{Pu}$ : 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  $\text{km}^2$  from global fallout, versus the nanocuries 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  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$ : 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  $^{238}\text{Pu}$ : $^{239,240}\text{Pu}$  ratio.

$^{241}\text{Am}$ : This nuclide in fallout has originated almost entirely from decay of its parent  $^{241}\text{Pu}$ . At the ratios so far observed of  $^{241}\text{Pu}$  to  $^{239,240}\text{Pu}$  in test debris, the ingrowth of  $^{241}\text{Am}$  is not predicted to produce ratios above 0.30  $^{241}\text{Am}$  to  $^{239,240}\text{Pu}$  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  $^{241}\text{Am}$  can be at least partly differentiated from fallout  $^{241}\text{Am}$  by its ratio to  $^{239,240}\text{Pu}$ .

$^{242}\text{Cm}$ ,  $^{244}\text{Cm}$ : 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  $^{60}\text{Co}$  or  $^{55}\text{Fe}$  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  $^{137}\text{Cs}$  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	
	$^{137}\text{Cs}$ $\mu\text{Ci/day}$	$^{239,240}\text{Pu}$ $\text{pCi/day}$	$^{137}\text{Cs}$ $\mu\text{Ci/day}$	$^{239,240}\text{Pu}$ $\text{pCi/day}$
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	-- <sup>1</sup>	19.7	-- <sup>1</sup>
Apr. 1977	3.7	-- <sup>1</sup>	117	<2,700
May 1977	3,533	300	78.7	< 67
June 1977	-- <sup>2</sup>	30.0	-- <sup>1</sup>	< 33
July 1977	1,680	247	1.8	13.3
Aug. 1977	-- <sup>3</sup>	-- <sup>3</sup>	8.7	< 23
Sept. 1977	-- <sup>3</sup>	-- <sup>3</sup>	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

<sup>1</sup> Sample too small for analysis at these concentrations.

<sup>2</sup> No data.

<sup>3</sup> 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  $\mu$ m 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:

$^{55}\text{Fe}$ to $^{137}\text{Cs}$	about	1
$^{134}\text{Cs}$ to $^{137}\text{Cs}$	"	0.39
$^{238}\text{Pu}$ to $^{239,240}\text{Pu}$	"	1.7
$^{244}\text{Cm}$ to $^{241}\text{Am}$	"	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  $^{137}\text{Cs}$ , very slightly depleted in  $^{90}\text{Sr}$ , shows no evidence of enrichment by  $^{134}\text{Cs}$ ,  $^{238}\text{Pu}$  or  $^{239,240}\text{Pu}$ , and has been enriched by 290% in  $^{55}\text{Fe}$ . A larger volume sample would have been needed for measurement of  $^{134}\text{Cs}$  at the concentration indicated. It is of interest that the  $^{55}\text{Fe}$  was essentially 100% associated with particles collected by the 1  $\mu$ m 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  $^{55}\text{Fe}$  from Millstone and that No. 3 shows also some  $^{137}\text{Cs}$ , 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  $^{134}\text{Cs}$  or  $^{238}\text{Pu}$ ) 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  $62\ \mu$ , compared to over 66% less than  $62\ \mu$  for core No. 2. The radionuclide concentrations of the sediments also differ substantially. The Niantic Bay core is much the richer in  $^{55}\text{Fe}$ , slightly poorer in  $^{137}\text{Cs}$ , and much poorer in  $^{239,240}\text{Pu}$ . Neither core showed, even in the top section, any measurable  $^{134}\text{Cs}$ , nor any strong elevation of the  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  inventory of core No. 2, over  $45.5\ \text{mCi/km}^2$  (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<sup>2</sup> of fallout <sup>137</sup>Cs. The inventory of Pu in core No. 2, over 12.1 mCi/km<sup>2</sup>, appears at least as much elevated as that of <sup>137</sup>Cs, whereas the <sup>55</sup>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 <sup>134</sup>Cs and for <sup>238</sup>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 <sup>55</sup>Fe of core No. 3, and the high <sup>137</sup>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 <sup>55</sup>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 <sup>137</sup>Cs, <sup>238</sup>Pu, <sup>239,240</sup>Pu, <sup>241</sup>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 <sup>137</sup>Cs and <sup>239,240</sup>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 <sup>137</sup>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 <sup>137</sup>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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$ . 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  $^{137}\text{Cs}$ ).

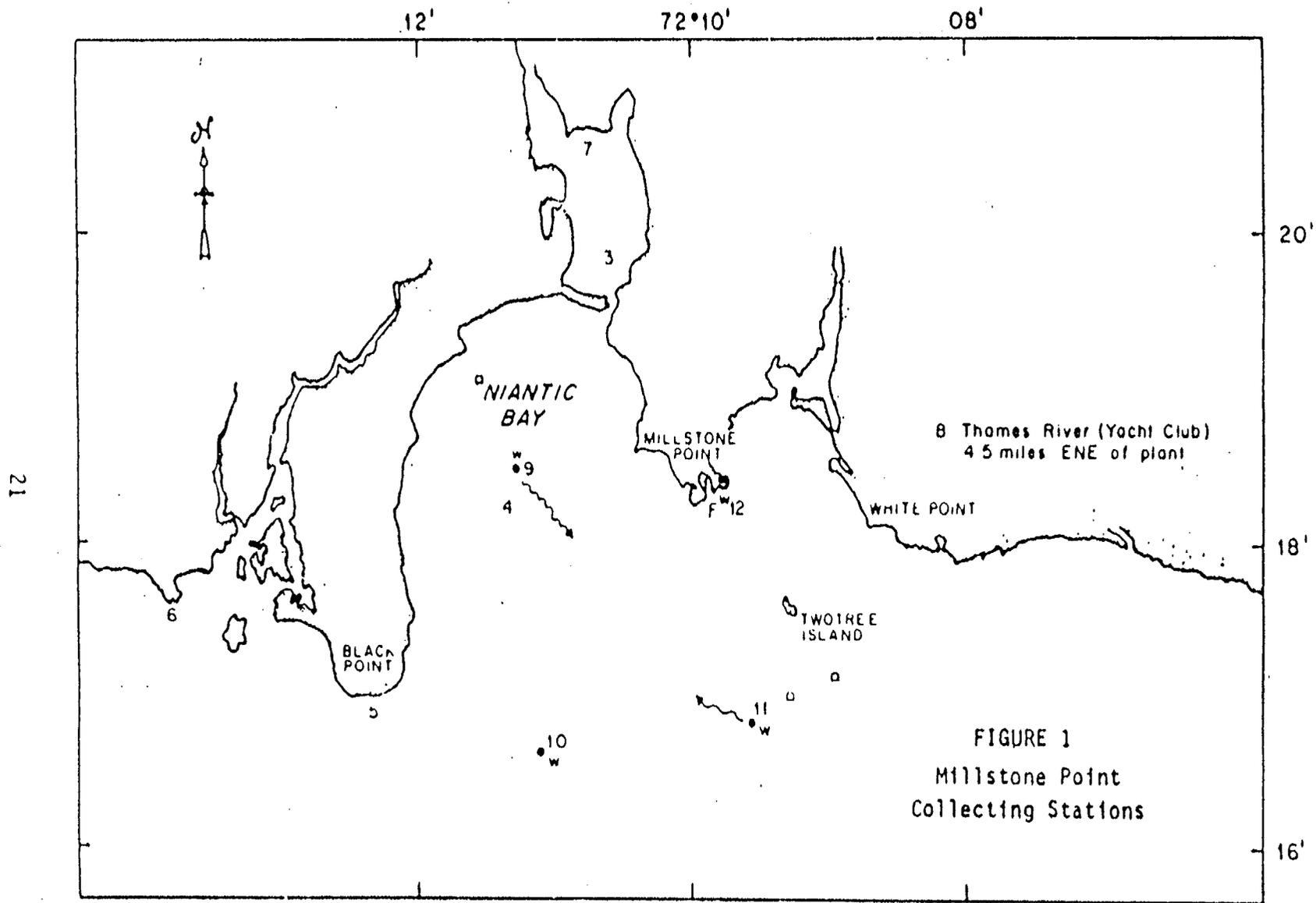


TABLE 5

Millstone Point, Conn.

## Long Island Sound Surface Water\*

(Radionuclides in disintegrations per minute per 100 kg water)

<u>Station</u>	<u><math>^{134}\text{Cs}</math></u>	<u><math>^{137}\text{Cs}</math></u>	<u><math>^{90}\text{Sr}</math></u>	<u><math>^{236}\text{Pu}</math></u>	<u><math>^{239,240}\text{Pu}</math></u>	<u><math>^{55}\text{Fe}</math></u>
#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 >1 $\mu$ , from outflow, inner plume)	---	---	---	0.002±0.001	0.032±0.001	26.7±0.2
#5B(particulates >1 $\mu$ , 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<sup>1</sup> Radiochemistry

(Radionuclides in disintegrations per minute per kg dry sediment)

A. Station 2, Core #3						
Section cm	Wet/ Dry	<sup>55</sup> Fe <sup>8</sup>	<sup>137</sup> Cs	<sup>238</sup> Pu	<sup>239,240</sup> Pu	<sup>241</sup> Am
0-1	1.53 <sup>2</sup>	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 <sup>3</sup>	325±46	600±4 <sup>7</sup>	3.27±0.19	129±3	32.3±1.5 <sup>6</sup>
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 <sup>6</sup>
7-9						
9-11	1.83	388±25	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	46.5±1.6 <sup>6</sup>
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 <sup>6</sup>
				5		5

Inventories,  
mCi/km<sup>2</sup>  
% estim. delivery

30.5  
128%

45.5  
38.5%

12.07  
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 <sup>238</sup>Pu:<sup>239</sup>Pu = 0.028.  
 5 = mean ratio <sup>241</sup>Am:<sup>239</sup>Pu = 0.257.  
 6 = No Curium detectable.  
 7 = <sup>134</sup>Cs less than 1/60 of <sup>134</sup> + <sup>137</sup>Cs.  
 8 = <sup>55</sup>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	$^{137}\text{Cs}$	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{241}\text{Am}$	$^{242}\text{Cm}$	$^{244}\text{Cm}$
Millstone Beach	Nov. 1970	M. edulis	Shell		---	0.98±0.04			
			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	1776±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	bd1	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		
<u>Fallout Contaminated Samples for Comparison:</u>									
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, I.I. 28 mi. SW	1976	M. edulis	Soft parts	2.4±0.4	0.008±0.003	0.09±0.01	0.014±0.006	---	---
	1977	M. edulis	Soft parts	3.1±0.25	0.002±0.002	0.120±0.012	<0.01	---	---
New Haven, Conn. 42 mi. W	1976	M. edulis	Soft parts	Lost	0.011±0.004	0.24 ±0.02	0.042±0.013	---	---
	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. 30 mi. SE	1976	M. edulis	Soft parts	5.1±0.3	0.008±0.005	0.19 ±0.02	0.037±0.015	---	---
	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.  
 bd1 = Below detection limits.  
 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  $^{137}\text{Cs}$  or of Pu were too low for detection. The change in the ratio  $^{137}\text{Cs}$  to  $^{90}\text{Sr}$  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  $^{55}\text{Fe}$ .

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  $^{134}\text{Cs}$  to have certainly come from the Maine

Yankee reactor. The ratio  $^{134}\text{Cs}$  to  $^{137}\text{Cs}$ , 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  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$ , in the sediments of Table 8 with those reported by Livingston and Bowen (1979) suggests there was little or no Pu in the high- $^{137}\text{Cs}$  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  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$  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  $^{134}\text{Cs}$  to  $^{137}\text{Cs}$ .

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  $^{134}\text{Cs}$  to  $^{137}\text{Cs}$ , 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  $^{134}\text{Cs}$  content. The fluctuations, with depth, of the ratios of either  $^{239,240}\text{Pu}$  or  $^{55}\text{Fe}$  to  $^{137}\text{Cs}$ , do not show the patterns that we would expect to result from resolubilization and redistribution within the sediment, as discussed for Pu: $^{137}\text{Cs}$  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  $^{55}\text{Fe}$  to  $^{137}\text{Cs}$  is quite interesting: nowhere less than 50%, 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  $^{55}\text{Fe}$  were from the low end of the range of its concentrations, or of its ratios to  $^{137}\text{Cs}$ . Also evident is the fact that, as was seen at Millstone Point (above),  $^{55}\text{Fe}$  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  $^{55}\text{Fe}$  expected from worldwide fallout: although Joseph et al (1971) estimated that 1962-1966 fallout was characterized by a ratio  $^{55}\text{Fe}$  to  $^{90}\text{Sr}$  of 9.4 (=  $^{55}\text{Fe}$  to  $^{137}\text{Cs}$ , 6.5), the elapse of 4  $^{55}\text{Fe}$  half-lives by 1975 reduced this to  $^{55}\text{Fe}$  to  $^{137}\text{Cs}$  of 0.406 or so. Although geochemical processes will affect Fe and Cs in different ways, fallout  $^{55}\text{Fe}$  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  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$ . 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  $^{55}\text{Fe}$  (250% of fallout) or of  $^{137}\text{Cs}$  ( $\sim 65 \text{ mCi}\cdot\text{km}^{-2}$  vs the range 4 to 15  $\text{mCi}\cdot\text{km}^{-2}$  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  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$ , and the concentration vs depth-in-core curve of  $^{239,240}\text{Pu}$  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  $^{134}\text{Cs}$  to  $^{137}\text{Cs}$  in this core shows unusual variability, with no significant  $^{134}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{238}\text{Pu}$ . 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  $^{55}\text{Fe}$  analyses would help clarify this interpretation, as will  $^{134}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$  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  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$ . 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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  and Pu, although the  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$  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  $^{137}\text{Cs}$  or Pu. The  $^{55}\text{Fe}$  in the second of these samples does look high, however; in Narragansett Bay (and Plymouth area

7) mussels we have found ratios of  $^{55}\text{Fe}$  to  $^{239,240}\text{Pu}$  in the 30 to 50 range, significantly different from the value close to 100 reported here. Too little comparison data is available concerning  $^{55}\text{Fe}$  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  $^{137}\text{Cs}$  and of Pu; whole worms contained  $^{137}\text{Cs}$  at a concentration about 10% and Pu about 22.7% that (estimated ) in the wet sediment from which they were collected. Cleaning reduced the  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$ , and 53% of Pu; also, of these concentrations higher fractions were in the tissues: 42% of  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$ , but 18% of tissue Pu, soluble in formalin, whereas the fallout contaminated worms showed all their tissue  $^{137}\text{Cs}$ , 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.

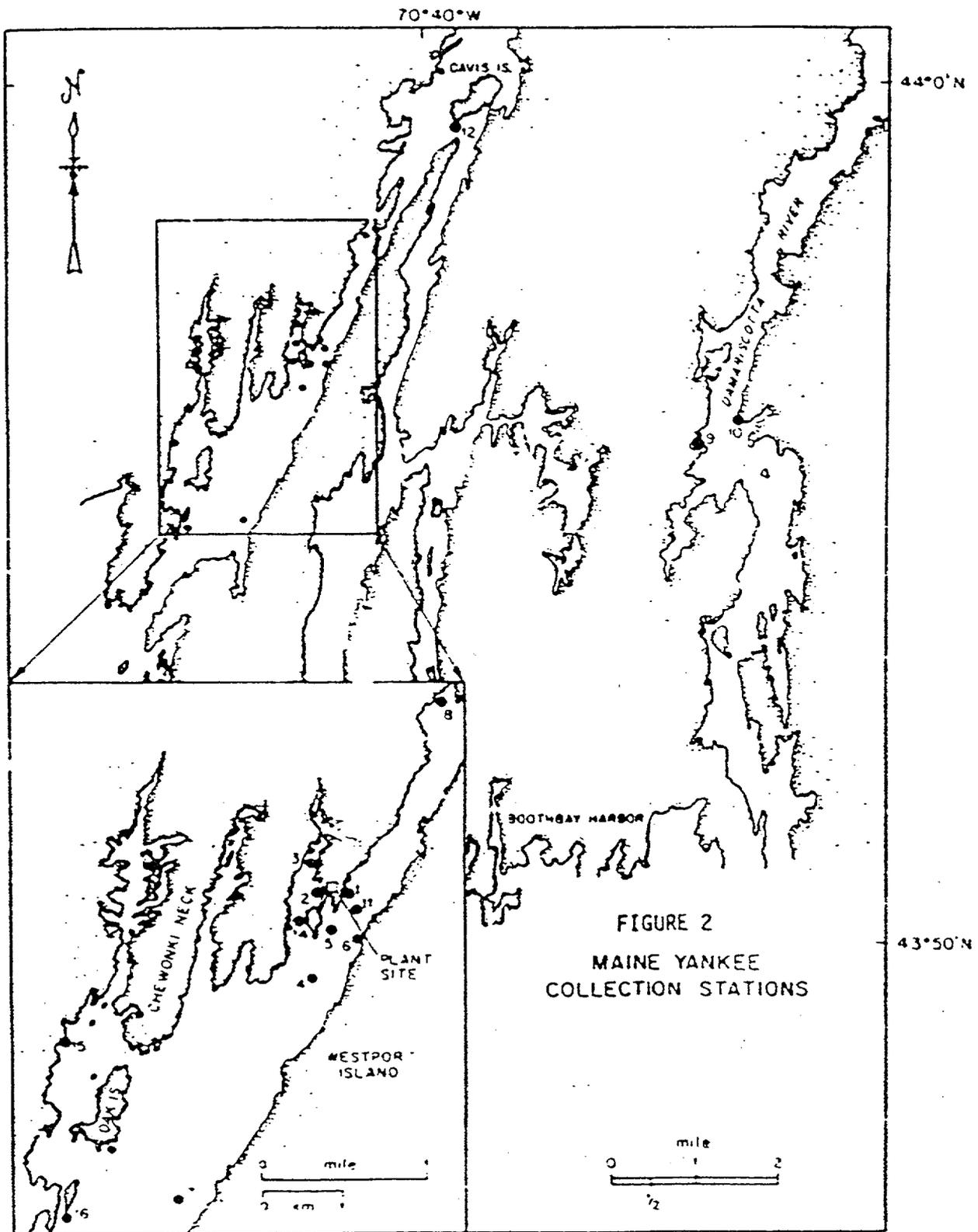


TABLE 8

Wiscasset, Maine. Area

## Surface Water from Montsweag Bay\*

(Radionuclides in disintegrations per minute per 100 kg water)

<u>Station</u>	<u><math>^{137}\text{Cs}</math></u>	<u><math>^{90}\text{Sr}</math></u>	<u><math>^{238}\text{Pu}</math></u>	<u><math>^{239,240}\text{Pu}</math></u>	<u><math>^{55}\text{Fe}^{\dagger}</math></u>
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
i (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.

† = Decay correction to 1 January 1975.

TABLE 9

Miscasset, Maine Area

Surficial Sediment Radiochemistry. I

(Radionuclides in disintegrations per minute per kg dry sediment)

Sample Location	Date	$^{134}\text{Cs}$	$^{137}\text{Cs}$	$^{134}\text{Cs}/^{137}\text{Cs}$	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$
2	14 Aug 1974	<20	885±30	<0.02	4.04±0.73	95±2.7
2	25 June 1975	16,000±600	27,700±1,000	0.58	3.03±0.27	71±2.2
2	8 Dec. 1975	3840±230	8,260±110	0.46	3.55±0.41	98±4
1	30 May 1975	340±50	1,030±40	0.33	2.17±0.23	63±2.5
3	30 June 1975	10,600±500	19,200±40	0.55	7.6±0.5	174±5
15	-	500±50	1,620±30	0.31	3.13±0.24	87±2.8
16	-	lost	lost	--	0.80±0.08	22.4±0.6

Notes: 1 - For locations see Figure 2.

2 - Decay corrected to date of collection

TABLE 10

## Miscasset, Maine, Area

Sediment Cores<sup>1</sup> Radiochemistry, II

(Radionuclides in disintegrations per minute per kg dry sediment)

A. Core #3 (Location 6)						
Section cm	Wet/ Dry	<sup>55</sup> Fe <sup>2</sup>	<sup>134,137</sup> Cs <sup>3</sup>	<sup>134</sup> Cs/ <sup>137</sup> Cs <sup>3</sup>	<sup>238</sup> Pu	<sup>239,240</sup> Pu
0-1	2.26		534±5	0.09±0.10	6.4±0.4	98±3
1-2	1.72		934±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 <sup>2</sup> :		61.7	102	---	---	0.605
% estimated delivery:		250%	83%			29%

Notes: 1 = For locations see Figure 2; samples collected 5 January 1977.  
 2 = <sup>55</sup>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	<sup>55</sup> Fe	<sup>137</sup> Cs	<sup>238</sup> Pu	<sup>239,240</sup> Pu	<sup>241</sup> Am
1	<i>C. virginiana</i>	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	<i>A. nodosum</i>	Whole	Aug. 1976		40±0.4	0.07±0.03	1.53±0.17	
Boothbay	"	"	Sept. 1976		27.6±0.3	0.11±0.03	4.16±0.29	0.690±0.072
14	<i>F. vesicularus</i>	"	Aug. 1976		53±0.5	0.05±0.02	2.7±0.2	
Boothbay	"	"	Sept. 1976		14±0.3	0.02±0.01	1.4±0.1	
"	Green Urchin	"	" 1976		4.1±0.2	0.35±0.007	0.60±0.03	
6	<i>M. arenaria</i>	Soft Parts	May 1977		21±0.5	0.043±0.012	0.57±0.05	
11	<i>M. edulis</i>	" "	Dec. 1977		5.2±0.3	bd1	0.25±0.02	
?	"	" "	Nov. 1977	13.1±1.2	3.7±0.5	0.002±0.004	0.14±0.02	
<u>Fallout contaminated samples, for comparison:</u>								
Sears Isl.	<i>M. edulis</i>	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
Walley 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
35 mi SW	"	" "	1977		9.1±0.24	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.  
 † Result of replicated counting.  
 bd1 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 <sup>1</sup>	Sample	Date	<sup>137</sup> Cs	<sup>238</sup> Pu/ <sup>239</sup> Pu	<sup>239,240</sup> Pu
Wiscasset (Area 1)	Whole Worms	1-2 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 4	"	1524±10 (508)	0.041±0.005	62.5±1.6 (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 4	"	111±3 (37)	0.028±0.013	9.57±0.82 (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.

Fatics:

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 0% 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  $^{137}\text{Cs}$  or of Pu; its ratio  $^{137}\text{Cs}$  to  $^{90}\text{Sr}$  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:

	<u>Chondrus</u>	25.2	<sup>137</sup> Cs;	1.07	239,240 <sub>Pu</sub>
Fall	<u>Fucus</u>	13.4	" ;	1.9	"
	<u>Red Algae</u>	47.9	" ;	2	"
	<u>Chondrus</u>	38.1	" ;	0.67	"
Spring	<u>Fucus</u>	12.7	" ;	1.33	"
	<u>Red Algae</u>	21.9	" ;	0.66	"

It appears that the seasonal change has led to increased <sup>137</sup>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 <sup>137</sup>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 <sup>137</sup>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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$ , 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 or 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  $^{137}\text{Cs}$

Site 1: 5/28/78  $^{55}\text{Fe}$  (ratio to Pu is 390, vs 30 to 50 elsewhere)

Site 2: None

Site 3: 6/15/71  $^{137}\text{Cs}$ , 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  $^{55}\text{Fe}$  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 ( $^{55}\text{Fe}$ ) effects, and the site 7 high Pu was accompanied by the lowest  $^{241}\text{Am}$  we have seen in a mussel (or other environmental sample); a ratio  $^{241}\text{Am}$  to  $^{239,240}\text{Pu}$  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 contra-indicated) by the relatively low ratio  $^{238}\text{Pu}$  to  $^{239,240}\text{Pu}$ . Unfortunately the  $^{241}\text{Am}$  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  $^{137}\text{Cs}$  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.

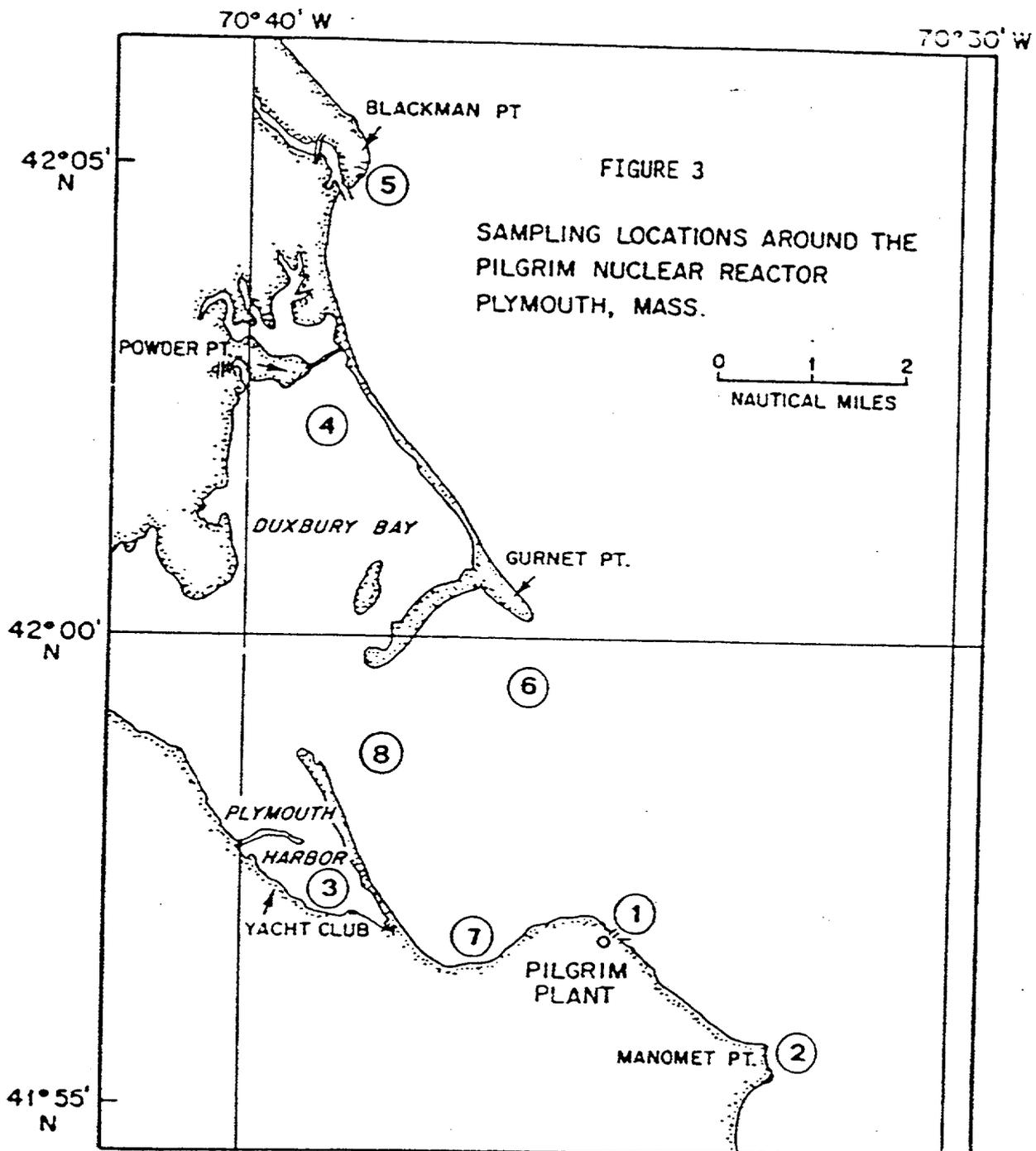


TABLE 13

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	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{238}\text{Pu}$	$^{239,240}\text{Pu}$	$^{238}\text{Pu}/^{239}\text{Pu}$
1*	Oct.	surface	$36.2 \pm 0.6$	$32.0 \pm 0.5$	N.M.	$0.15 \pm 0.06$	---

Part B - Sediment Analyses:

1 <sup>x</sup>	Oct. 20	0-3	Grab	$66.1 \pm 1.5$	$0.29 \pm 0.04$	$8.2 \pm 0.3$	0.035
2	Oct. 19	0-4	Core	$157 \pm 3$	$2.6 \pm 0.2$	$61.5 \pm 1.7$	0.042
4	Oct. 19	0-6	from Anchor	$59.7 \pm 1.3$	$0.68 \pm 0.07$	$21.3 \pm 0.6$	0.032
Cape Cod Bay	Aug.	0-1	Core	$187 \pm 6$	$2.5 \pm 0.3$	$101 \pm 3$	0.034
		1-2	Core	$181 \pm 3$	$2.8 \pm 0.2$	$75 \pm 2$	0.037
		3-4	Core	$176 \pm 2$	$2.6 \pm 0.2$	$69 \pm 2$	0.038
		5-6	Core	$181 \pm 3$	$2.7 \pm 0.3$	$62 \pm 3$	0.043
		8-10	Core	$174 \pm 3$	$2.5 \pm 0.3$	$69 \pm 3$	0.036
		12-14	Core	$198 \pm 3$	$2.4 \pm 0.2$	$69 \pm 2$	0.034
		16-18	Core	$107 \pm 3$	$1 \pm 0.1$	$33 \pm 1$	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 <sup>1</sup>	Date	Organism <sup>3</sup>	<sup>55</sup> Fe <sup>2</sup>	<sup>137</sup> Cs	<sup>238</sup> Pu	<sup>239,240</sup> Pu	<sup>241</sup> 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	Elderduck - 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 6.

TABLE 14B  
Plymouth, Mass. Area Environmental Samples - Biota  
(Radionuclides in disintegrations per minute per kg fresh weight)

Collecting Site <sup>1</sup>	Date	Organism	<sup>55</sup> Fe <sup>2</sup>	<sup>137</sup> Cs	<sup>238</sup> Pu	<sup>239,240</sup> Pu	<sup>241</sup> 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 undatum	N.D.	4.6±0.3	0.03±0.01	0.75±0.04	N.D.
3	" "	Lunatia heros	N.D.	39±3	0.05±0.01	0.69±0.04	N.D.
3	Aug 28 75	Mytilus edulis	N.D.	3±0.2	0.02±0.01	0.35±0.05	N.D.
3	" "	Balanus balanoides	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 <sup>d</sup>	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 <sup>D</sup>	0.032±0.004 <sup>D</sup>
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<sup>1</sup> to Plymouth Area (Tables 14A + 14B)  
 (Radionuclides in Disintegrations per Minute per Kg Wet Weight)

Collection Site	Date	Organism <sup>2</sup>	<sup>137</sup> Cs	<sup>238</sup> Pu	<sup>239,240</sup> Pu	<sup>241</sup> Am
Boston, MA <sup>3</sup>	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 <sup>4</sup>	9-25-76	" "	1.9±0.3	0.012±0.006	0.13±0.02	0.020±0.008
	8-19-77	" "	1.9±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 liaison 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  $^{55}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  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

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

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

$^{238}\text{Pu}$  is released at very high ratios to  $^{239,240}\text{Pu}$ , 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  $^{55}\text{Fe}$  or  $^{60}\text{Co}$ , 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 radio-nuclide tracers.

#### Environmental Samples

Millstone Point sampling has shown the following: All  $^{55}\text{Fe}$  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

## Appendix Table

Common Names and Taxonomic Details for  
Organisms listed in Tables 7, 11, 12, 14A-8, 15

<u>Plants</u> <sup>1</sup>	<u>Common Names</u>
A. <u>Brown Algae</u>	
<u>Fucus vesiculosus</u>	Rock Weed
<u>Ascophyllum nodosum</u>	" "
<u>Laminaria Tongicruris</u>	Kelp
B. <u>Red Algae</u>	
<u>Chondrus crispus</u>	Irish Moss
Mixture: <u>Ceramium rubrum</u>	None
<u>Soeremothamnion Turner</u> } chiefly	"
<u>Phycodrus rubens</u> }	"
<u>Phyllophora brodiaei</u> } some	"
 <u>Animals</u>	
A. <u>Annelida</u> <sup>2</sup>	<u>Worms</u>
<u>Nereis virens</u>	Sandworm; Clamworm
B. <u>Molluscs</u> <sup>2</sup>	<u>Shellfish</u>
<u>Bivalves</u> <sup>2</sup>	
<u>Crassostrea virginiana</u>	Oyster
<u>Spisula solidissima</u>	Surf Clam
<u>Mytilus edulis</u>	Blue Mussel
<u>Gastropods</u> <sup>2</sup>	<u>Snails</u>
<u>Lunatia heros</u>	Moon snail
<u>Succinum undatum</u>	Waved Whelk
<u>Littorina littorea</u>	Common Periwinkle
C. <u>Crustacea</u> <sup>2</sup>	
<u>Balanus balanoides</u>	Barnacle
<u>Homarus americanus</u>	Lobster
D. <u>Echinodermata</u> <sup>2</sup>	
<u>Strongylocentrotus drobachiensis</u>	Green Sea-urchin
E. <u>Birds</u> <sup>3</sup>	
<u>Somateria mollissima</u>	Common Eider duck
 <u>Notes:</u>	
<sup>1</sup> - Taxonomy from Kingsbury, 1969.	
<sup>2</sup> - Taxonomy from Smith, 1964.	
<sup>3</sup> - Taxonomy from Robbins, et al, 1966.	

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