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BNWL-2267
UC-41

Sequim Marine Research Laboratory Routine Environmental Measurements During CY -1976

J. J. Fix
P. J. Blumer

May 1977

Prepared for the Energy Research and Development Administration under Contract EY-76-C-06-1830

Battelle
Pacific Northwest Laboratories

BNWL-2267

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Price: Printed Copy $_____*; Microfiche $3.00

*Pages	NTIS Selling Price
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SEQUIM MARINE RESEARCH LABORATORY ROUTINE ENVIRONMENTAL MEASUREMENTS DURING CY-1976

by

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May 1977

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SEQUIM MARINE RESEARCH LABORATORY ROUTINE ENVIRONMENTAL MEASUREMENTS DURING CY-1976

INTRODUCTION

Beginning in 1976, a routine environmental program was established at the Marine Research Laboratory (MRL) at Sequim, Washington. The program is intended to demonstrate the negligible impact of current MRL operations on the surrounding environs and to provide baseline data through which any cumulative impact could be detected. The sampling frequency of the program, as shown in Table 1, is much greater during the first two years of the program to provide sufficient data to provide reliable estimates of current conditions.

The program is designed, primarily, to determine levels of radioactivity present in selected biota in Sequim Bay. The biota were selected because of their presence near the laboratory and their capacity to concentrate trace elements. Other samples were obtained to determine the radionuclides in Sequim Bay and laboratory drinking water, as well as the ambient radiation exposure levels and surface deposition of fallout radionuclides for the laboratory area. Appendix A provides a summary of the analytical methods used.

The present document includes data obtained during CY 1976, the first year of the program. It is intended that a document be written each year providing a summary of the data collected and an evaluation of the data based on either historical data or regulatory requirements promulgated by the State of Washington or the U.S. Environmental Protection Agency.

SAMPLE COLLECTION AND ANALYSIS

Since initiation of the environmental program at Sequim MRL during 1976, only a few results are available for each sample type collected. As such, estimates of the existing conditions are preliminary. As additional data become available during 1977, more reliable estimates can be made.

DRINKING WATER

Results of radiological analyses for gross beta activity, gross alpha activity, ^{3}H and gamma-emitting radionuclides are shown in Table 2. All of the results are less than detectable with the exception of the gross beta and ^{3}H results for 12/2. These results are very near the detection level and may simply be due to counting statistics. As additional data become available, it will be possible to determine whether the positive results are due to the presence of low level radioactivity or to the statistical uncertainty associated with background counting rates.

Table 3 presents the results of total coliform analyses of MRL drinking water. The results are all <2 per 100 ml or within the standard of no detectible coliform organisms in drinking water.

BAY WATER

Results of radiochemical analyses of Sequim Bay water samples are shown in Table 4. All of the results are below the detection level with the exception of gross beta activity, gross alpha, and ^{40}K. The ^{40}K activity is responsible for the positive gross beta counts. The positive gross alpha activity is likely attributable to naturally occurring uranium. A specific analysis for uranium will be done during 1977.

Table 5 is a summary of the copper as well as oil and grease analyses of Sequim Bay water during 1976. Additional data will be collected during 1977 to better estimate the concentrations over a longer time period.

CLAMS

Three varieties of clams were collected during 1976. Results of analyses for gamma-emitting radionuclides, ^{90}Sr and total Plutonium are shown in Table 6. All of the results are near the detection limit. Additional data will be necessary to determine if there is any detectible radioactivity with the current radiochemical procedures.

SEAWEED

Samples of Kelp and Eel grass were collected from Sequim Bay near the MRL dock. With the exception of ^{40}K and total Plutonium, all of the results shown in Table 7 were less than detectible. There is an apparent large difference in the amount of potassium present in Kelp as opposed to Eel grass.

AMBIENT RADIATION EXPOSURE

Table 8 is a summary of the external radiation level measured by thermoluminescent dosimeters placed one meter above ground level. The locations of the dosimeters are shown in Figure 1. Each dosimeter includes 3 chips of CaF_2:Dy (Harshaw TLD-200) encased in an opaque capsule lined with 2 mils of lead and 10 mils of tantalum.

The data have been very consistent from month to month as shown in Table 8. The relatively low rate of exposure is somewhat surprising. The estimated annual exposure rate would be approximately 40 mR. It is difficult to coordinate the placement and prompt return of the dosimeters. Any time spent in a building awaiting deployment or their return will lower the average daily exposure because of the shielding provided by the building.

However, the consistency of the data and the known prompt delivery and return for most of the crystals would tend to support the validity of the tabled values. During 1977, measurement with a pressurized ion chamber will be used to further check the exposure rate expected.

SOIL

Results of radiochemical analyses of soil samples collected from the immediate vicinity of the MRL are shown in Table 9. The sample locations are shown in Figure 1. The low concentrations of naturally occurring ^{40}K, ^{224}Ra, and ^{226}Ra further confirm the relatively low exposure rates measured in the environs of the MRL. These concentrations are about 50% of the concentrations observed in the Hanford environs.

The only statistically-positive measurement of artificially produced radionuclides was for ^{90}Sr, $^{95}ZrNb$, ^{137}Cs and $^{239-240}Pu$. All of the observed activity is attributed to world-wide fallout. Additional data collected during 1977 will allow a better estimate of the expected range of activity in soil for the MRL environs.

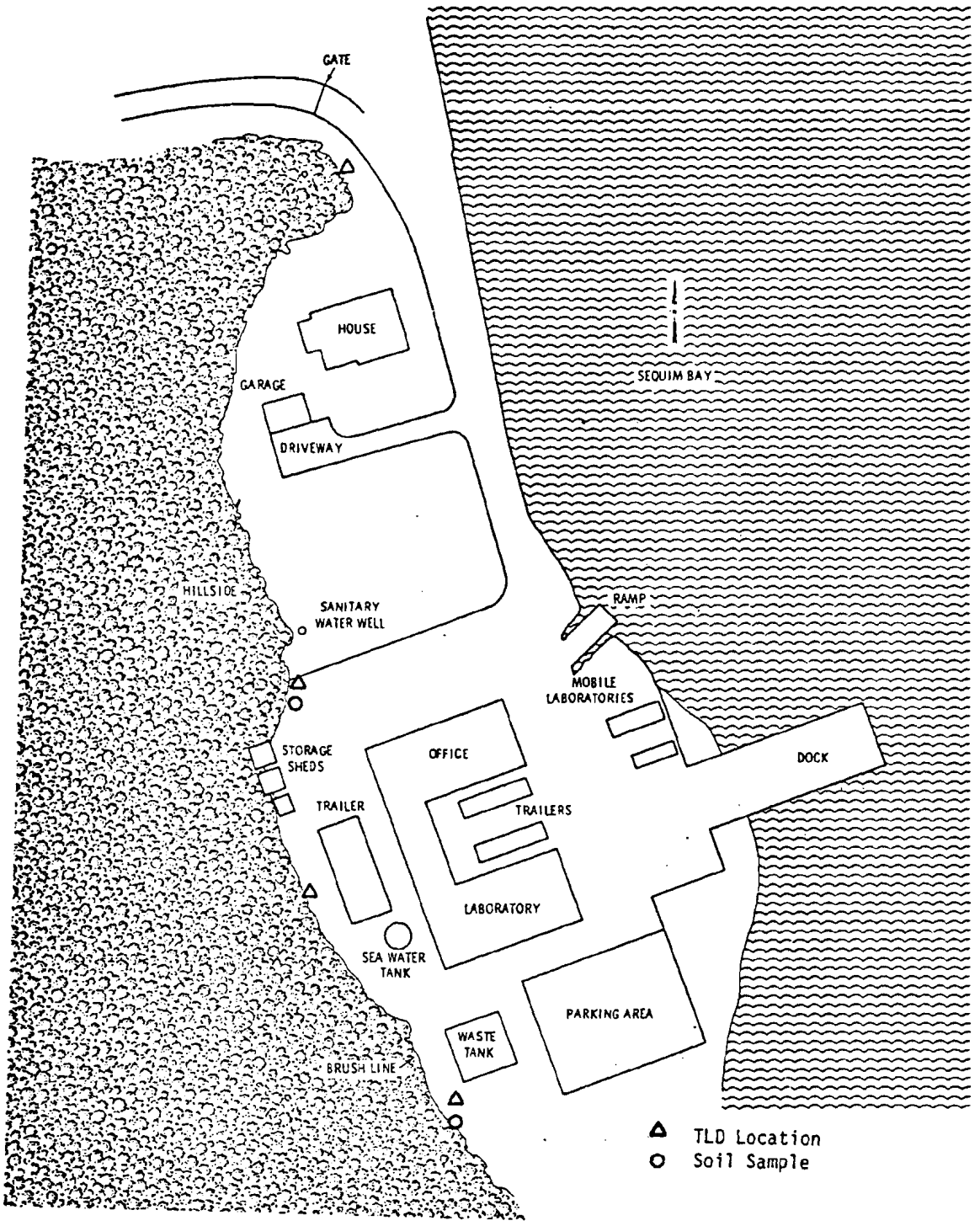

FIGURE 1. Sample Collection Locations at the Sequim MRL

TABLE 1. Sequim Marine Research Laboratory Sampling Schedule

Sample Type	Locations	Frequency		Amount Sampled	Analyses	
		1st 2 Years	Thereafter		Radiological	Other
TLD's[a]	4	Monthly	Monthly	--	External Radiation	
Bay Water	1	Monthly	Semiannually	4ℓ	γ-scan Gross beta Gross alpha Tritium C-14	Oil and grease[b] Heavy metals
Drinking Water	1	Quarterly	Semiannually	1ℓ	γ-scan Gross beta Gross alpha Tritium	Coliforms
Plankton	1	When Available[c]		2Kg	γ-scan Sr-90 Pu-total	
Kelp	1	Quarterly	Semiannually	2Kg	γ-scan Sr-90 Pu-total	
Eel Grass	1	Quarterly	Semiannually	2Kg	γ-scan Sr-90 Pu-total	
Clams	1	When Available[c]		2Kg	γ-scan Sr-90 Pu-total	
Soil	2	Annually	Annually	2Kg	γ-scan Sr-90 Pu-total	

(a) Three chips of CaF_2:Mn (Harshaw type TLD-400) encased in an opaque capsule lined with 2 mils of lead and 10 mils of tantalum.

(b) Analysis for heavy metals dependent on their use in laboratory research projects.

(c) Samples obtained when they are available. Frequency is dependent on their availability.

TABLE 2. Concentrations of Radionuclides in Drinking Water Samples from Sequim (Grab Samples) - 1976[a]

(Units of 10^{-9} μCi/ml of Water)

Date	Beta	Alpha	^{3}H	^{46}Sc	^{51}Cr	^{60}Co	^{65}Zn	^{137}Cs
8/2	3.9 ± 5.5	0.25 ± 0.39	-0.02 ± 140			0.93 ± 28	-0.32 ± 55	0.55 ± 31
10/6	2.4 ± 5.9	0.34 ± 0.35	-170 ± 110			2.5 ± 28	13 ± 57	4.0 ± 31
12/2	7.5 ± 6.1	0.34 ± 0.35	280 ± 200	0.09 ± 36	0.38 ± 490	0.16 ± 28	-0.003 ± 56	0.05 ± 31

(a) Analytical results ± 2 sigma counting uncertainty. Negative results do not reflect a physical reality but result from the statistical uncertainty of discriminating between the sample and instrument background count rate.

TABLE 3. Total Coliform Analyses of Sequim MRL Drinking Water-1976

Date	Total Coliform (per 100 ml)
8/2	<2.0
10/6	<2.0

TABLE 4. Concentrations of Radionuclides in Bay Water Samples from Sequim (Grab Samples) - 1976(a)

(Units of 10^{-9} μCi/ml of Water)

Date	Beta	Alpha	^{3}H	^{14}C	^{40}K	^{46}Sc	^{51}Cr	^{60}Co	^{65}Zn	^{137}Cs
8/2	120 ± 28	0.35 ± 2.6	260 ± 110	19 ± 200	160 ± 240			0.06 ± 14	1.6 ± 28	-1.1 ± 15
9/2	130 ± 29	2.4 ± 1.9	550 ± 200	-9.9 ± 2.6	210 ± 170			11 ± 11	-1.9 ± 20	-0.89± 10
10/6	220 ± 34	2.7 ± 2.2	80 ± 100	7.2 ± 8.8	180 ± 170			0.06 ± 11	-0.35± 20	2.0 ± 10
11/15	440 ± 44	2.3 ± 1.4	-47 ± 180	5.6 ± 6.9	190 ± 170	-0.16 ± 17	-19 ± 230	0.63 ± 11	-3.8 ± 20	-0.75± 10
12/2	450 ± 45	2.5 ± 2.0	200 ± 540	3.3 ± 7.3	190 ± 170	0.03 ± 14	-12 ± 150	0.06 ± 11	-2.7 ± 19	-1.3 ± 10

(a) Analytical results ± 2 sigma counting uncertainty. Negative results do not reflect a physical reality but result from the statistical uncertainty of discriminating between the sample and instrument background count rate.

TABLE 5. Concentrations of Oil and Copper in Water Samples from Sequim (Grab Samples) - 1976

Date	Bay Water		Incoming Lab Bay Water	
	Oil & Grease (mg/l)	Copper (µg/l)	Oil & Grease (mg/l)	Copper (µg/l)
8/2	4.3	2.0		
10/6	1.4	2.0	1.8	2.0

TABLE 6. Concentrations of Radionuclides in Clams Collected at Sequim - 1976[a]

Units of 10^{-6} μCi/g (Wet Weight)

Date	Sample	^{40}K	^{59}Fe	^{60}Co	^{90}Sr	^{106}Ru	^{137}Cs	Total Pu
1/15	Saxidomus Giganteus	0.89 ± 0.64		0.00008 ± 0.05	0.0006 ± 0.003	0.003 ± 0.87	0.004 ± 0.04	0.0008 ± 0.001
1/15	Prototheca Staminae	1.2 ± 0.82	1.2 ± 3.0	0.002 ± 0.06	0.003 ± 0.003		-0.01 ± 0.05	0.0007 ± 0.0001
1/15	Tresus Nuttallii	0.70 ± 0.76		-0.004 ± 0.06	0.003 ± 0.003		-0.003 ± 0.05	0.001 ± 0.0001
7/30	Saxidomus Giganteus	1.5 ± 0.68		-0.007 ± 0.05	0.007 ± 0.003		-0.02 ± 0.04	0.0009 ± 0.0001

(a) Analytical results ± 2 sigma counting uncertainty. Negative results do not reflect a physical reality but result from the statistical uncertainty of discriminating between the sample and instrument background count rate.

TABLE 7. Concentrations of Radionuclides in Kelp and Eel Grass Samples from Sequim - 1976[a]

Units of 10^{-6} μCi/g

Date	Sample	^{40}K	^{60}Co	^{65}Zn	^{90}Sr	^{137}Cs	Total Pu
1/19	Kelp	66 ± 2.4	0.0009 ± 0.16	0.31 ± 0.47	0.005 ± 0.004	0.09 ± 0.14	0.01 ± 0.006
7/30	Kelp	73 ± 2.3	0.020 ± 0.14	0.27 ± 0.25	0.002 ± 0.004	0.08 ± 0.13	0.006 ± 0.004
7/30	Eel Grass	9.8 ± 3.8	0.001 ± 0.26		0.002 ± 0.008	-0.07 ± 0.23	0.06 ± 0.02

(a) Analytical results ± 2 sigma counting uncertainty. Negative results do not reflect a physical reality but result from the statistical uncertainty of discriminating between the sample and instrument background count rate.

TABLE 8. Sequim Land TLD-1976
mR/day

Date		Near Tile Field	Near Back Trailer	Near Well	N. End Near Transformer
8/2	9/16	0.12	0.12	0.12	0.11
9/2	10/7	0.11	0.11	0.12	0.11
9/29	12/6	0.11	0.11	0.11	0.11
11/4	12/6	0.12	0.11	0.12	0.12
11/29	1/18	0.11	0.10	0.11	0.11

TABLE 9. Concentrations of Radionuclides in Sequim Soil Samples - 1976

Units of 10^{-6} μCi/g of Soil (Dry Weight)[a]

Sample Location	Naturally Occurring Radionuclides			Artificially Produced Radionuclides			
	^{40}K	^{224}Ra	^{226}Ra	^{54}Mn	^{58}Co	^{60}Co	^{90}Sr
West Side	7.4 ± 1.1	0.36 ± 0.06	0.26 ± 0.07	-0.008 ± 0.02	0.0009 ± 0.02	0.006 ± 0.02	0.01 ± 0.004
East Side	7.9 ± 1.1	0.33 ± 0.06	0.30 ± 0.08	-0.007 ± 0.02	0.005 ± 0.02	0.001 ± 0.02	0.004 ± 0.004

Sample Location	Artificially Produced Radionuclides						
	$^{95}ZrNb$	^{106}Ru	^{134}Cs	^{137}Cs	^{154}Eu	^{238}Pu	$^{239-240}Pu$
West Side	0.008 ± 0.03	-0.04 ± 0.15	-0.02 ± 0.02	0.07 ± 0.03		0.0003 ± 0.003	0.002 ± 0.001
East Side	0.04 ± 0.03	-0.05 ± 0.15	0.01 ± 0.02	0.04 ± 0.03	0.004 ± 0.17	0.0007 ± 0.003	0.0004 ± 0.001

(a) Analytical results ± 2 sigma counting uncertainty. Negative results do not reflect a physical reality but result from the statistical uncertainty of discriminating between the sample and instrument background count rate.

APPENDIX A

ANALYTICAL PROCEDURES

APPENDIX A

ANALYTICAL PROCEDURES

The environmental samples collected were analyzed for radionuclides by United States Testing Company of Richland, Washington and for coliforms, oil and grease, and copper by Hanford Environmental Health Foundation of Richland, Washington. The radiological data shown in Tables 2, 4, 6, 7 and 9 include the counting result plus or minus the 2-sigma counting uncertainty. Whenever the counting result is less than the uncertainty, the counting result is less than the detection limit (2-sigma uncertainty) of the analysis. Because of the statistical nature of radionuclide counting, an occasional counting result may be slightly greater than the 2-sigma uncertainty even if the radionuclide is not present. For this reason, several environmental samples must be analyzed to provide a reliable estimate of the radionuclides present. A brief description of the analytical procedures used follows.

RADIOLOGICAL ANALYSES

Water Samples

Gross Beta

Gross Beta activity measured by a direct count with a gas flow proportional counter of the dried residue. The counter is calibrated with a ^{90}Sr-Y equilibrium source.

Gross Alpha

Gross Alpha activity measured by a low level alpha counter following extraction into ether from a strong nitric acid. The ether phase is evaporated off and the residue plated on a stainless steel planchet. The planchet is counted with a gas flow proportional counter. The counter is calibrated with a ^{239}Pu source.

Gamma-Emitting Radionuclides

Gamma-emitting radionuclides are determined by a direct count of 500 mls. of sample in the well of a 9 inch x 9 inch NaI(TL) well detector with a multichannel gamma ray spectrometer.

Strontium-90

Strontium-90 in water samples is precipitated with fuming nitric acid, scavenged with barium chromate, precipitated as a carbonate, transferred to a stainless steel planchet and beta counted with a low level beta proportional counter. After a 15-day period, the Yttrium-90 daughter is separated and counted with a low level beta proportional counter.

Tritium

Tritium is measured in distilled water samples with a liquid scintillation spectrometer.

Clams, Eel Grass and Kelp Samples

Gamma-Emitting Radionuclides

Gamma-emitting radionuclides are determined by a direct count of the sample in the well of a 9 inch x 9 inch NaI(TL) well detector with a multichannel gamma ray spectrometer.

Plutonium

After ashing in a furnace and wet ashing with concentrated nitric acid, plutonium is extracted from the sample using cation exchange resin and thenoyl trifluoroacetone. Plutonium in the eluate is electrodeposited on a stainless steel disk, exposed to nuclear track film, and then counted

Soil Samples

Gamma-Emitting Radionuclides

Approximately 500 grams of sample are placed into a marinelli beaker and counted on a lithium drifted germanium detector, with a multichannel pulse height analyzer.

Plutonium and Strontium-90

Soil is dried, mixed thoroughly, leached with a mixture of nitric and hydrochloric acids, and then passed through an ion exchange resin in 8N nitric acid. The 8N nitric acid retains strontium and other metal ions. This phase is precipitated with fuming nitric acid, scavenged with barium chromate, precipitated as a carbonate, and transferred to a stainless steel planchet. Sample is counted with a low background beta proportional counter.

The plutonium is eluted from the resin column with 0.4N HNO_3-0.01 HF and electrodeposited on a stainless steel disk for alpha spectrometric analyses.

NON-RADIOLOGICAL ANALYSES

Coliform

Standard total coliform MPN technique as described in Standard Methods, 14th edition, p. 916 is used to determine coliform counts.

Copper

Atomic Absorption Spectrophotometric following concentration and extraction using ammonium pyrrolidine dithiocarbamate (APDC) and methyl isobutyl ketone (MIBK) as given in Standard Methods, 14th ed, pp. 148-152.

Oil and Grease

Partition-Gravimetric Method as given in Standard Methods, 14th edition, p. 515.

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