SAMPLING PROCEDURES

Groundwater: Domestic and/or irrigation wells were sampled at their highest and lowest levels in the spring and fall of each year.

Milk: Milk was collected monthly by the Oregon Department of Agriculture from production sources at the eight milk districts statewide. Also, increased sampling was conducted where needed to evaluate trends.

Ambient: Air particulate samples were obtained using continuous low-volume air monitors. Filters were exchanged and counted biweekly in Portland and bimonthly at the other locations. Vegetation samples consisting of various native grasses were collected when available at the various locations. Precipitation samples representing quarterly composites were collected using a rain collector. Soil samples representing an area of one square foot wide and one inch deep were obtained yearly. Ambient dose rates were determined using thermoluminescent dosimeters (TLD's), specifically, Harshaw TLD 200's encased in a copper energy-compensating capsule and exchanged on a quarterly schedule. Yearly measurements were made at all ambient sampling sites using a Reuter-Stokes Pressurized Ionization Chamber.

ANALYSIS PROGRAM

Gamma Spectrometry

Six gamma spectrometry systems were used during the 32-year period covered by this report:

1. Nuclear Data-120
2. Nuclear Data-130
3. Northern 660
4. Tracor-Northern TN-11
5. Nuclear Data 6610
6. Nuclear Data 9900

The Nuclear Data 120 and 130 and the Northern systems were used with sodium iodide detector assemblies. A 3" x 3" solid integral line assembly was used initially, then replaced with a 4" x 4" unit with a matching photo multiplier tube assembly. The Nuclear Data 6610 system was used with a germanium-lithium drifted (GeLi) detector. The Nuclear Data 9900 system interfaced with both the GeLi and the intrinsic germanium detector (INGe).

The sodium iodide detector assemblies used with the first four analyzers were housed in a unique shield consisting of an old money safe with 1-inch thick steel walls; the interior dimensions were 22" x 22" x 20". Exterior shielding was provided by 2 inches of lead on the bottom, 4 inches on the sides and back, and 4-3/8 inches on the top. The four-inch thick, swing-open, steel door was used with no additional shielding.

The shield used for the GeLi detector, and subsequently, the higher efficiency INGe detector, had interior dimensions of 16" x 16" x 16" with 6-inch-thick steel walls. The GeLi detector was relocated to a smaller shield with dimensions of 7½" x 7½" x 14", with a 4-inch lead sliding top cover. The detector was surrounded by 2 inches of lead shielding. All the shields had thin copper liners to reduce Compton scatter.

Calibration Procedures

Until 1975, a Marinelli beaker was the standard counting geometry for water and other low activity samples. The sample surrounded the detector at a constant depth of 1½ inches on the sides and top, with a total volume of 1,500 ml for the 3" x 3" sodium iodide detector and 1,750 ml for the 4" x 4" sodium iodide detector. After 1975, the standard counting geometry was a 500-ml capacity disposable plastic container.

Disposable plastic pint containers were used for counting sediment and algae samples: a 400-ml geometry in most cases and a 200-ml geometry for samples of limited availability.

Originally, the analyses were performed using a library of standards developed from solutions traceable to the National Bureau of Standards (NBS). After 1980, a library was created using a mixed radionuclide standard from Amersham. Counting efficiencies were determined by counting the radionuclides (along with appropriate stable carriers) in distilled water. The accumulated counts, in the photopeak of interest, were used to determine counting efficiencies and Compton interferences in lower-energy photopeaks. With the TN-11 system, radionuclide spectra were stored on cassette tape and used as a standards library.

Water, soil and vegetation samples were normally counted with little sample preparation. Water samples, stored in two-quart polyethylene bottles, were agitated with a rubber-tipped stirring rod to displace deposited material before transfer to a Marinelli beaker or plastic container for counting. Soil and vegetation samples were analyzed in the same plastic containers used for collection.

Interferences from the presence of higher-energy photopeaks in the same spectrum (Compton scatter) were compensated for initially by manually subtracting the interfering photopeaks. When this technique could no longer be applied due to closely adjacent overlapping photopeaks, a step-wise multiple regression (spectrum stripping) method was applied using the analyzer computer.

In May 1975, data reduction methods changed to incorporate the use of a multiple-least-squares fitting program by the TN-11 data acquisition system – a system with the ability to resolve 15 gamma-emitting radionuclides in the spectrum.

In 1980, the Nuclear Data 6610 "MIDAS" GeLi System was applied to routine operations. Used with GeLi gamma-spectrometry application software, the system processed data by identifying the nuclides through a peak search. The activity of the sample was calculated using a quadratic curve-fitting routine that interfaced the photopeaks in the sample spectrum with the calibration curves and nuclide parameters.

In 1986, the MIDAS system was replaced by the more advanced Nuclear Data 9900 (GENIE), a VAX-based system used with two detectors, the INGe and the GeLi.

All values are reported in picocuries per gram, wet or drained weight for biological and sediment samples, and in picocuries per liter for water samples. All activities reported were decay corrected to the date of collection.

Gross Alpha and Beta

Total gross alpha and beta activities in water were determined by evaporating a 500-ml aliquot and transferring the residue directly onto the planchet.

Three gas-flow proportional counters were used for gross alpha and beta analyses. A Nuclear Measurements PC-3A internal proportional gas-flow counter with a windowless 2-inch hemisphere detector was used until July 1966. This instrument was operated in the proportional region using P-10 counting gas. Beta background for this system, with extra lead shielding, was 40 counts per minute. The minimum detectable activity, based on three standard deviations of the background, was 10 pCi/liter for gross beta activity and 1 pCi/liter for gross alpha activity in water.

In August 1966, routine counting was shifted to a Sharp Wide Beta I automatic low-background proportional counting system incorporating a 2 1/4-inch gas-flow detector with an anti-coincidence guard detector. It had a 500 microgram per-square-centimeter window and was operated with methane counting gas. Under normal operation, the beta background was less than one count per minute. The minimum detectable activity for water was 1 pCi/liter for gross beta activity and 1 pCi/liter for gross alpha activity..

In January 1979, a low-background Tennelec LB 5100 system was installed. This system, which also incorporated an anti-coincidence guard detector, had a beta background of one count per minute.

In all three systems, the intrinsic beta counting efficiencies were determined using cesium-137, strontium-yttrium-90 and phosphorus-32. Alpha efficiencies, determined using plutonium and uranium standards, were comparable to values reported in the literature and by the equipment manufacturers. The plateaus to ascertain the best operating voltages for alpha and beta counting, were determined using the standard alpha and beta counting sources listed above.

Thermoluminescent Dosimetry (TLD), for Ambient Radiation Measurements

TLD's were annealed first by heating for one hour at 400þ and then for two hours at 100þ. After the TLD's had been returned from the field, they were placed in a lead shield for approximately two weeks before readout.

The pre-read annealing process consists of heating the TLD's at 100þC for 30 minutes and then allowing the them to cool to room temperature. The TLD's were readout using a Harshaw 2000-B with a readout time of 30 seconds.

QUALITY ASSURANCE

The goal of quality assurance (QA) is to ensure the defensibility of data and measurements. QA procedures in this surveillance program consisted of independent checks on sample handling procedures, radiation counting equipment performance, radiochemical procedures, and data management. The procedures included:

1. Participation in Environmental Protection Agency Radiation Quality Assurance Intercomparison Studies.
2. Split sampling and analysis with similar facilities, including the State of Washington Environmental Radiation Program, University of Washington Laboratory of Radiation Ecology, and Oregon State University Department of Oceanography.
3. Comparisons made through duplicate analyses performed on a periodic basis.

There are two considerations in quantifying the presence of radioactivity in a sample:

1. The uncertainty in measuring the background activity.
2. Determining net sample activity above the background level.

The following sections outline statistical methods used to help ensure the defensibility of data in this report. Readers should also note that throughout the report:

- þ Any analysis reported as "less than" (<) a certain activity, is one where there was no activity above the stated lower limit of detection.
- þ Any summary average followed by an asterisk (*) is one where the indicated activity may be less than the amount indicated.

Minimum Detectable Activity

Since radioactive disintegration is a random event, repetitive counts on the same sample may not be identical. These counts, accumulated during a predetermined counting period, will vary within a predictable range, clustering near the center. Deviations from the center or mean value can be assessed through applied statistics. Small deviations are more probable than large ones.

Standard statistical analyses can be used to calculate the probability of a value falling within a predetermined confidence level. ("Confidence level" is the frequency with which the counts will fall within predetermined limits of error.) In this report, that confidence level has been calculated to be 95%, indicating that the specific result or value will fall within these stated limits of error in 95 out of 100 measurements.

The highest uncertainty exists when measuring sample activity near the minimum detectable level, i.e., close to background. Therefore, a high confidence level is desirable to ensure that the counts reflect sample activity and not background fluctuations.

The minimum reportable activity limits in this report were in accordance with the minimum detectable activity definition from the National Bureau of Standards Handbook 80, *A Manual of Radioactivity Procedures*:

"Minimum detectable activity is the amount of activity which, for the same counting time, gives a gross count that exceeds the gross background count by three times the standard deviation of the background count." (Commonly referred to as a 3 sigma.)

Based on this definition, there is a 99.7% probability that the count at the minimum reportable level represents sample activity above background. The standard deviation can be defined as the square root of the observed events. Background counting rate, counting time, and intrinsic counting efficiency all influence the minimum detectable activity.

Count Rate Error

When activity above the minimum detectable level is observed, it creates another uncertainty. It is defined as the count rate error or "counting error," which encompasses both the background and gross sample counting error. The 95% confidence level (2-sigma error) is defined by the following equation:

$$E = \pm 1.96 \sqrt{\frac{R_t}{T_s} \quad \frac{R_b}{T_b}}$$

E = count rate error at the 95% confidence level.
R_t = sample counting rate (gross CPM).
R_b = background counting rate (CPM).
T_s = sample counting time (minutes).
T_b = background counting time (minutes)

Detection Capabilities for Environmental Sample Analyses

Any measurement system contains a degree of uncertainty. In the field of nuclear radiation detection, much statistical work has been done to define the uncertainty associated with a counting determination. The highest degree of uncertainty exists when measuring the same radioactivity near the commonly found natural levels (background levels). The lower limit of detection (LLD) of sample activity over background is stated in HASL-300[2] by Pasternack and Harley:

"The smallest amount of sample activity that will yield a net count for which there is confidence at a predetermined level that activity is present."

Therefore, the LLD in cpm at the 95% confidence level is defined by the following equation:

$$LLD\ 4.66\sqrt{\frac{Bkg\ counts}{Time\ counted}}$$

In this report, any analysis reported as "less than" (<) a certain activity is one in which

[2]HASL 300 p. D-08-01 ff.

there was no activity observed above the stated LLD.

Counting Statistics

In low-level environmental radiation counting, the Poisson distribution is used to describe a discrete variable (detectable counts). The standard deviation (of the mean), s, of this distribution is expressed in HASL-300[3].

$$s \; \sqrt{N}$$

Where N is the number of counts obtained, the standard deviation, s, for a net sample counting rate is:

$$\sqrt{\frac{\text{Bkg rate}}{\text{Bkg time counted}} \quad \frac{\text{sample gross counting rat}}{\text{sample gross time counte}}}$$

[3]HASL 300 p. A-06-02 ff.