Appendix C. Analytical Procedures

This appendix summarizes selected procedures that the Analytical Services Laboratory (Lab) uses to analyze indirect bioassay samples. The procedures used by the In Vivo Monitoring Program (IVMP) to perform direct bioassay measurements are also summarized here.

C.1 Indirect Bioassay Samples

All indirect bioassay samples are analyzed to determine their content of various radionuclides, according to detailed procedures written and maintained by the Lab. A brief description of most frequently used procedures follows.

C.1.1 Tritium in Urine

A 1-mL sample aliquot is mixed with scintillator solution and counted directly in a liquid scintillation spectrometer.

C.1.2 Emergency and Expedite Processing of Urine

Total Radiostrontium

Up to 100 mL of the sample is precipitated as a carbonate, dissolved and re-precipitated as a nitrate, scavenged with barium chromate, converted to carbonate, and counted in a low-background gas-flow proportional counter.

Plutonium

Plutonium is converted to (+IV) valence state using sodium nitrite and adsorbed on an anion exchange column. Iron and thorium are removed by nitric and hydrochloric acid. The plutonium is desorbed from the column, using ammonium iodide, and electrodeposited on a planchet. Alpha-emitting isotopes are then counted, using alpha spectrometry. Plutonium-241 activity is determined by dissolving the material from the planchet with nitric and hydrochloric acid and then counting the beta emissions in a liquid scintillation spectrometer.

Americium and Curium

The sample is wet-ashed using nitric acid and mixed with anion exchange resin and/or Eichrom resin to remove plutonium and other heavy metals. The residual solution is coprecipitated with calcium oxalate and re-dissolved with nitric acid. The americium and curium are concentrated with neodymium fluoride, electrodeposited on a planchet, and counted using alpha spectrometry.

Uranium

A 10-mL aliquot of the sample is wet-ashed with acid and passed through an ion exchange column. The uranium is eluted with a weak acid and brought to a fixed volume with acid. The final sample is submitted for laser-induced phosphorescence uranium analysis.

C.1.3 Emergency and Expedite Processing of Fecal Samples

Total Radiostrontium

The sample is wet- and dry-ashed. Strontium is precipitated as the nitrate, scavenged with barium chromate, converted to carbonate, and counted in a low-background gas-flow proportional counter.

Plutonium The sample is wet- and dry-ashed. Enhanced oxidation steps using hydrofluoric acid and hydrogen peroxide can be requested. Then the procedure is the same as that for urine samples (see Section C.1.2).

Americium and Curium The procedure is the same as that for urine samples (see Section C.1.2).

Uranium The sample is wet-ashed only. Then the procedure is the same as that for urine samples (see Section C.1.2).

C.1.4 Priority and Routine Processing of Urine and Fecal Samples

For all analyses except tritium, the samples are first wet- and dry-ashed. Entire samples are used except for the low-sensitivity uranium analysis. Additional wet- and dry-ashing, sometimes using perchloric and hydrofluoric acids, may be needed on fecal samples.

Tritium analysis and the low-sensitivity uranium analysis are not available for fecal samples.

Strontium-90 Strontium is precipitated as the carbonate and then as the sulfate. The re-dissolved material is scavenged with hydroxide, followed by two barium chromate precipitations. This is followed by carbonate, nitrate, and a final carbonate precipitation. The carbonate is mounted on a planchet and counted with a low-background gas-flow proportional counter. If this count is less than 1 dpm, the procedure is terminated. If the first count is greater than 1 dpm, ^{90}Y is allowed to grow into equilibrium with the ^{90}Sr. The ^{90}Y is then separated by hydroxide and oxalate precipitations, and ignited to yttrium oxide. The beta emissions are then counted. The first count gives the total strontium activity, and the second (yttrium) count gives the ^{90}Sr activity.

Plutonium Two methods are used to process plutonium. These methods are essentially the same as the procedure for emergency and expedite processing, except that counting times are increased to achieve the desired sensitivity. Priority and routine processing use a 2,500 minute count time. The low-level plutonium procedure (IPUL) uses an approximate 48-hour urine sample and a 10,000 minute count time.

Americium and Curium The ash is dissolved in nitric acid and successively coprecipitated with oxalate and ferric hydroxide. The final precipitate is redissolved with nitric acid and oxidized with sodium nitrite. Plutonium and other metals are separated using anion exchange resin. Americium and curium are adsorbed on a caution exchange column, using ammonium thiocyanate as a complexing agent to remove iron and thorium or an Eichrom resin column is used. The americium and curium are removed from the column, electrodeposited on a planchet or coprecipitated on a filter, and counted by alpha spectrometry.

Uranium A 50-mL aliquot of the sample is wet-ashed with acid and passed through an ion exchange column. The uranium is eluted with a weak acid and brought to a fixed volume with acid. The final sample is submitted for laser-induced phosphorescence uranium analysis.

Analyses for other radionuclides are available, as indicated in Tables 6.2 through 6.6.

C.1.5 Combinations

Usually, more than one procedure can be performed on one sample. For instance, 1 mL of a urine sample can be extracted for tritium analysis before any of the other analyses are begun. Some of the other more common combinations are

- plutonium and strontium
- plutonium and americium
- plutonium and uranium
- plutonium, strontium, and americium.

C.2 Direct Bioassay Measurements

Details concerning procedures, equipment, and data processing for direct bioassay measurements are provided in the *In-Vivo Monitoring Program Manual*.[(a)] Pertinent information is provided as follows.

C.2.1 Whole Body Counts

Whole body counts are performed using the stand-up counter (a five-detector NaI system in a stand-up position within a shielded booth) or a high resolution germanium detector system involving an array of coaxial high-purity germanium (HPGe) detectors in a shielded counting room. Most radionuclides with gamma-ray energies from about 200 to 3000 keV can be quantified (e.g., ^{137}Cs, ^{60}Co). The germanium detectors have much better photopeak resolution, which generally eliminates interferences from medical radionuclides and natural radon progeny. The stand-up counter uses a 200-second count and can make some determination of spatial distribution of radioactivity as being in the head, chest, abdomen, or legs by identifying the detector with the most counts. The coaxial system uses a 600-second count time, with the option of longer counts, and can be used in either a moving scan or static position mode.

If a radionuclide other than ^{40}K is detected, the person is asked to shower, change into clean coveralls, and be recounted. The recount may be performed either on the same system as the initial measurement, or on a more sensitive system.

(a) Internal manual, PNL-MA-574, Pacific Northwest Laboratory, Richland, Washington.

C.2.2 Chest Counts for Lung Activity

The presence of high-energy gamma-emitting radionuclides in the chest is determined by whole body counting or stationary counting using the coaxial germanium system. The presence in the chest of gamma- or x-ray-emitting radionuclides with energies in the range of a few tens of keV to 200 keV is determined by chest counting. The chest counter routinely reports ^{241}Am, ^{235}U, and ^{234}Th (as an indicator of ^{238}U). A peak search program is used to identify the presence of other significant photon energies.

The two chest counting systems used to detect nuclides that emit low energy photons each consist of an array of four planar HPGe detectors and associated electronics. The detectors are positioned anteriorly over the lungs in light contact with the chest. The subject is seated in a slightly reclining position. The routine counting time is 3000 seconds. Longer count times are employed where a lower detection level is required.

If a radionuclide is detected, the person is asked to shower, change into clean coveralls, and be recounted. The recount will usually be slightly longer in duration to improve sensitivity.

If material is detected in the chest, then an ultrasound measurement of the thickness of the chest is made, and the calculated activity in the lung is corrected for the absorption of the low-energy rays in the chest wall.

Additional corrections can be performed when activity such as ^{241}Am can exist simultaneously in the lung, liver, and bone. Such correction will usually be made based on additional measurements, notably head counts and liver counts. The final corrected activity represents a best estimate of the activity actually in the lung.

C.2.3 Head Counts for Skeleton Activity

Head counts are performed to quantify the skeletal activity of low-energy x-or gamma-ray-emitting radionuclides, such as ^{241}Am. The head count consists of planar germanium detectors placed on the forehead. The typical count time is 3000 seconds. The results of the head count are converted to activity in the total skeleton based on the distribution of ^{241}Am observed in the skeleton of a total body donation to the U.S. Transuranium Registry.

C.2.4 Thyroid Counts

Thyroid measurements are routinely performed using a single HPGe detector positioned 10 cm above the thyroid. The routine counting time is 600 seconds.

C.2.5 Liver Counts

Liver measurements are performed using arrays of HPGe detectors positioned anteriorly over the liver in light contact with the subject. The routine counting time is 3000 seconds. Routine calibrations are performed for Am-241. If a significant amount of activity is also present in the skeleton, then the measurement count rate over the liver is corrected for

contribution from the skeleton. The liver calibration factors are determined based on the thickness of the tissue over the liver.

C.2.6 Wound Counts

Wound counts may be performed at either the Emergency Decontamination Facility (EDF) or at the 747-A Building (IVRRF), depending on the circumstances. For low-energy x- or gamma rays, a single germanium detector is used. A portable germanium spectroscopy system is also available for making low energy wound counts. For small, localized puncture wounds, a large volume coaxial HPGe detector is usually used at IVRRF to estimate the activity for nuclides that emit high-energy photons. A sodium iodide detector is also available at the EDF for this purpose. The typical count time is 10 minutes. The activity of plutonium isotopes should be considered approximate, unless the depth of the activity in the tissue and relative abundance of each plutonium isotope are known.