Assessing Exposure to Radiation

Lawrence Livermore National Laboratory is a world leader in evaluating exposures to radiation, monitoring releases of radionuclides to the environment, and managing the environmental and health risks of such releases.
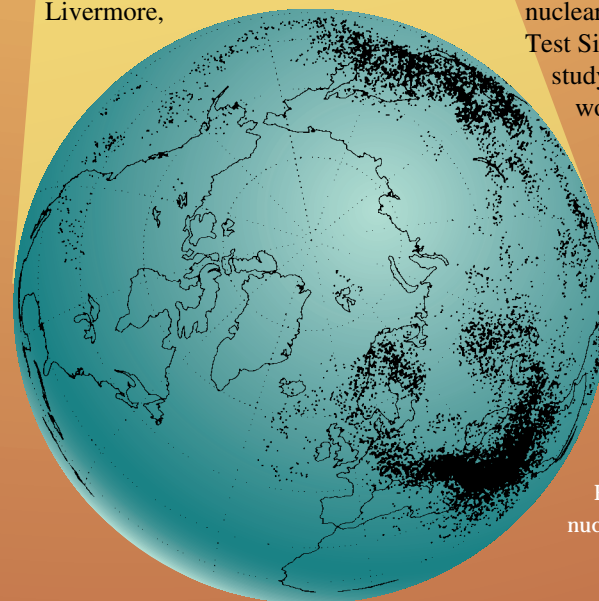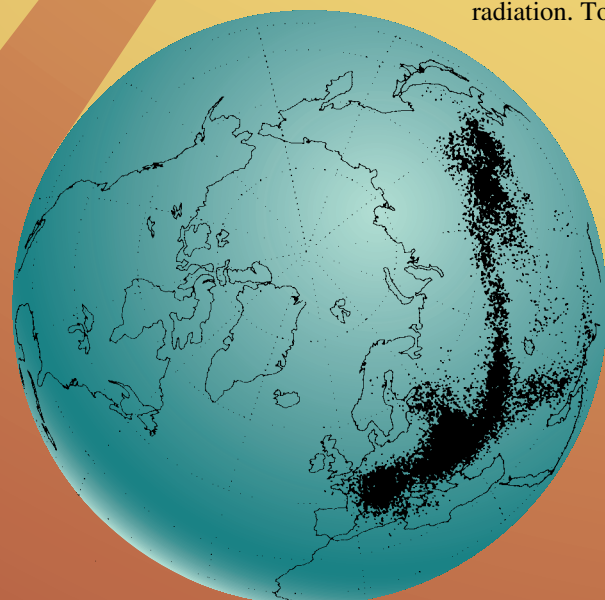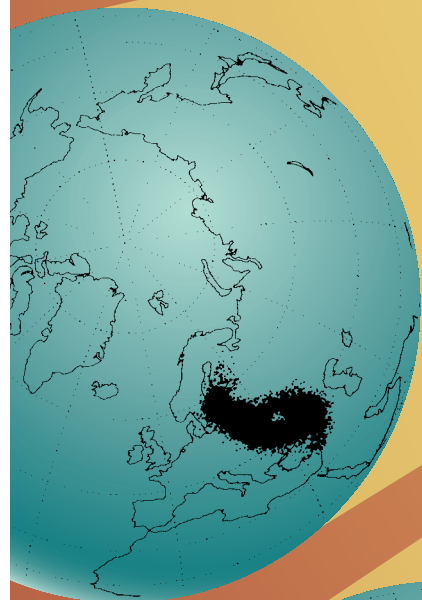
DAILY, every person receives "background" radiation from a variety of natural sources: from cosmic rays and radioactive materials in the Earth, from naturally occurring radionuclides in food, and from inhaling particulate decay products of radon gas. But as we have harnessed radioactivity and the power of the atom, we have created situations where we may also be exposed to manmade radiation.

Since its inception, Lawrence Livermore National Laboratory has been a world leader in evaluating exposures to radiation and their resultant risks. We use dosimetry to infer the "dose," or energy deposited in tissue, that a person has received as a result of exposure to radiation. To study past events, we have found ways to reconstruct the dose that individuals received many years earlier. We are also able to project the radionuclide retention in an individual 50 years after exposure. We can run a sampling program, do all of the analytical work, and produce assessments of the dose received. Few other facilities possess this range of capabilities.

Ultrasensitive measuring devices and sophisticated modeling techniques give us the means to quantify and extrapolate radiation exposures. We use fluorescence *in situ* hybridization (FISH), developed at Livermore, to study the effects of radiation at the cellular level. This tool gives us a better understanding of the risks associated with radiation and provides the basis for radiation safety standards worldwide.

The Laboratory has been called upon to investigate many significant exposures. We began studying the Chernobyl accident almost as soon as it happened. Dose reconstruction work related to the bombing of Hiroshima continues today with studies of nickel-63 in copper to refine the estimate of the neutron dose. We participated in a huge project to reconstruct the dose received by populations residing downwind from nuclear tests performed at the Nevada Test Site from 1951 to 1981. Another study will analyze the exposures to workers, children, and area residents at Mayak in the former Soviet Union, where plutonium and other nuclear weapons materials were produced.

Workers, including those at Livermore, who handle significant quantities of radioactive material as part of their jobs are routinely monitored. We have combined those data with information on monitored workers extending back to the Manhattan Project to update and revise the models that dosimetrists throughout the world use to describe the retention of radioactive substances in the body. In another project, our dose assessment and projection efforts will help the residents of Bikini Atoll, whose homeland was contaminated by atmospheric nuclear testing, return home soon.

We also work with numerous federal agencies to avoid nuclear accidents and to be prepared for them when they do occur. Our Atmospheric Release Advisory Capability (ARAC—see pp. 18–20) is pre-eminent in assisting with advanced planning for potentially hazardous activities, staff training, and emergency response.

Figure 1. Plots prepared by our Atmospheric Release Advisory Capability showing how the clouds of radioactive material spread around the Northern Hemisphere at (from upper left) 2 days, 4 days, 6 days, and 10 days after the initial nuclear accident at Chernobyl.

The Chernobyl Accident

Shortly after the April 1986 Chernobyl nuclear power plant accident, the Department of Energy assembled a task force, which included a Livermore team led by Lynn Anspaugh, to assess the accident's biological effects. Reconstructing the dose posed special problems. For many months after the accident, the source term—the amount and kinds of radionuclides released—was not revealed by the Soviet Union and so had to be inferred.

One of the earliest samples of Chernobyl's radioactive effluent was an air sample taken in Finland nearly two days after the release. From the ratio of the activities of the dozens of radionuclides in this sample, the known half-lives of the nuclides, and the known radioactive products ("daughters") of these nuclides and their activities and half-lives, we could calculate what the radionuclide mixture in the air would be at any subsequent (or earlier) time.

Then, by adding measurements of the external gamma- exposure activity rates or radionuclide deposition on the ground, we could calculate what the external exposure rate and dose from each radionuclide would be to individuals, both at the time of the sampling and well into the future. Once we reconstructed the major pathways by which these radioisotopes could enter individuals via the ingestion of contaminated food, we could calculate what their internal doses would be. Finally, from the external and internal dose, we estimated the possible biological effects on individuals and populations of the radioactive release using health-risk models (see box pp. 18–20).

Calculating the internal dose is generally the most difficult part of a dose reconstruction problem because of the many available mechanisms by which radiation can enter the human body. We

used PATHWAY,[1] the model that was developed for our dose reconstruction work at the Nevada Test Site, to calculate radiation dose resulting from ingested radionuclides.

Another consideration in this work was the sheer size of the Chernobyl release and the complex global circulation pattern the radioactive plume followed. ARAC's modeling efforts showed that Chernobyl produced three separate clouds of radioactivity that went in different directions. The radiation released in the initial explosion rose to an altitude of 1.5 to 7.5 kilometers (0.9 to 4.7 miles) and went east and southeast, whereas the radiation released from the resulting fire stayed below 1.5 kilometers and headed northwest (see Figure 1). Our work showed that the dose to individuals and the collective dose to populations varied considerably from one country to another. Only within the immediate vicinity (30 kilometers, or 18.6 miles) of Chernobyl were the biological effects expected to be large enough to be detected epidemiologically.[2]

More recently, the incidence of childhood thyroid cancer has risen dramatically in the areas of Belarus, Ukraine, and Russia nearest Chernobyl, prompting a renewed interest in reconstructing the dose because of iodine-131, which collects in the thyroid gland. However, iodine-131 has a half-life of only eight days and thus had decayed by the time most soil measurements were taken. Ordinarily, iodine-131 deposition can be estimated from the cesium-137 level in soil. But the explosion and subsequent fire at Chernobyl produced releases with different relative quantities of the two radionuclides, and the debris then traveled in different directions. The ratio of cesium-137 to iodine-131 was thus not the same at different locations.

In a project for Belarus, Livermore's Tore Straume, Lynn Anspaugh, and

others developed a method using accelerator mass spectrometry (AMS—see box pp.18–20) to measure the deposition in soil of the isotope iodine-129, which is much longer lived but more difficult to measure than iodine-131. Initial measurements of the ratio of iodine-129 to iodine-131 indicated that the ratio is constant over different locations, making it possible to reconstruct the iodine-131 dose from iodine-129 soil measurements.[3]

The next step will be to take enough soil samples to develop an "iodine map" for all of Belarus to determine the correlation between thyroid cancer and iodine deposition. This information will be enormously useful for future epidemiological studies, for establishing thyroid cancer screening procedures, and for dose reconstruction in other areas of the country.

Assessing Worker Exposure

When x rays were first used in the late 19th century, skin burns were observed, and in the 1920s, scientists discovered that radiation exposure caused genetic mutations on a cellular level. When the Manhattan Project began in the 1940s, radiation protection was considered important, but standards and established practices did not exist. Scientists did not know how to evaluate the dose to workers from internal depositions of radioactive materials because there was limited information on the biological effects of radiation and how the body retains radioactive substances.

Since then, research has provided better information about biological effects and retention of radionuclides, with a consequent improvement in radiation protection standards and practices. Today, for example, workers at Livermore's Plutonium Facility follow strict procedures.

Since the 1940s, radiation workers have been routinely monitored using

bioassay techniques—both *in vivo* (e.g., whole-body and lung counters) and *in vitro* (e.g., urine, feces, hair, saliva, and blood samples). (See Figure 2.) From these bioassay results, internal doses are derived using biokinetic models (see box pp. 18–20) and dose-estimation techniques. From the resulting doses, the worker's level of exposure is estimated and compared with radiation protection standards.

Because the biokinetic models, dose-estimation techniques, regulatory standards, and reporting requirements have evolved over time, today we have a vast number of dose estimates that are difficult to compare from one time period to another. One task of our Internal Dosimetry Assistance Group, led by David Hickman, is to evaluate and reconcile these past data. This is a huge task involving more than 50 years of bioassay data, sometimes with daily sample frequencies.
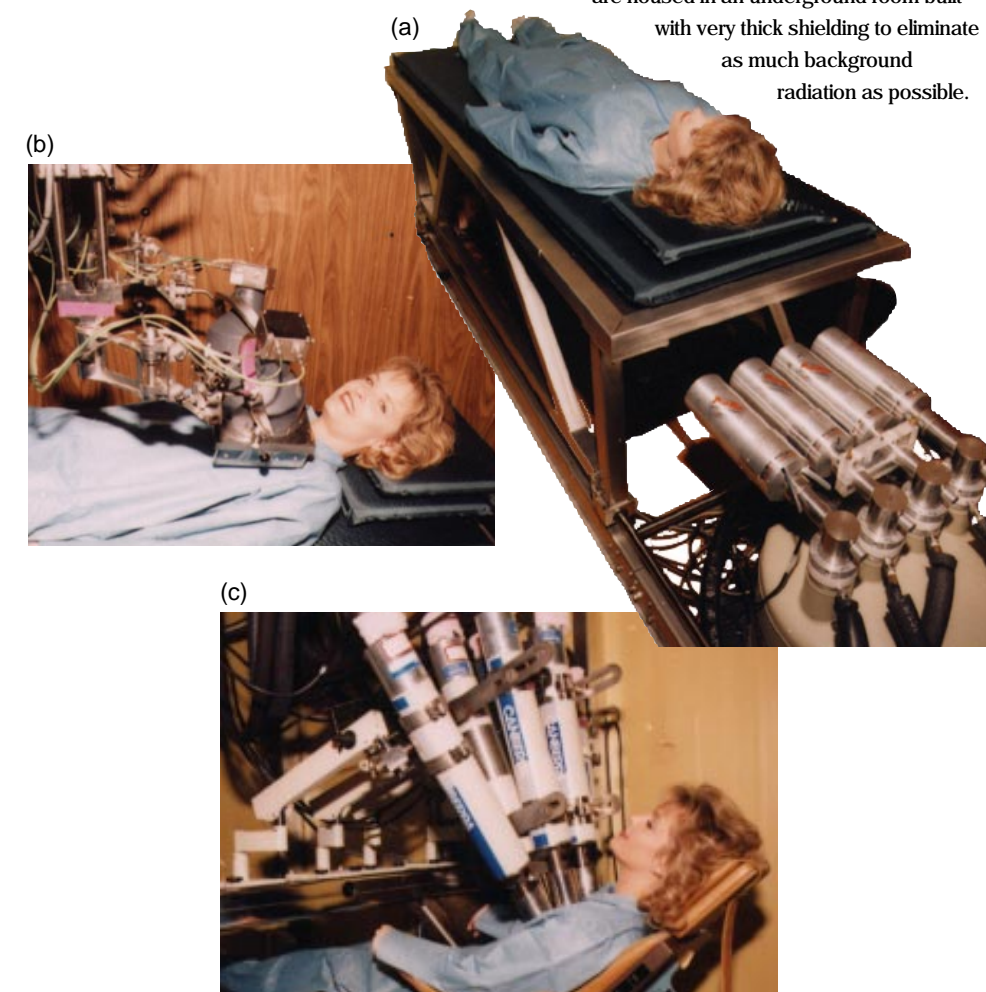
The Internal Dosimetry Assistance Group also performs work for outside organizations. One recent study of workers at another facility involved employees who had unintentionally received intakes of phosphorus-32, an isotope commonly used in biomedical research. Large amounts of *in vivo* and *in vitro* bioassay data were collected to make a determination of the internal dose. This project also supplied enough data to derive a new biokinetic model for phosphorus-32 based on human metabolic data. Many biokinetic models are based on animal data because those are the only data available, but human data, when available, tend to be more valuable.

Using our accumulated information, we have also been able to produce a standard method of evaluating worker exposure to radionuclides independent of the time the exposure occurred. We have used this information to derive new human-based, rather than animal-based, biokinetic models that dosimetrists can

use as default radionuclide retention assumptions when bioassay data are not available.

The Marshall Islanders

Fallout from nuclear tests between 1946 and 1958 at Bikini and Enewetak atolls contaminated several islands of the Northern Marshall Islands in the western Pacific Ocean. In particular, the BRAVO Test on March 1, 1954, contributed heavily to the contamination at Bikini Atoll and at other atolls to the east. Relocated to other islands prior to the tests, the people of Bikini and Enewetak atolls have wanted to return home ever since. As part of an evaluation of resettlement options, our team led by

Figure 2. A variety of detectors are used for *in vivo* measurements of radionuclide levels. (a) shows Nancy Allen, a training instructor in the Hazards Control Department, lying on a scanning-bed, whole-body counter. Its four germanium detectors measure the distribution of high-energy, photon-emitting radionuclides. In (b), 15.2-centimeter- (6-inch-) square Phoswich detectors are positioned over her lungs, and in (c), low-energy germanium (LEGe) detectors also cover her lungs. Both the Phoswich and LEGe detectors measure low-energy, photon-emitting radionuclides in such organs as the lungs, thyroid, liver, kidney, bone, and lymph nodes. The LEGe detector best identifies specific radionuclides, while the Phoswich measures a larger area and provides more accurate readings of nuclide activity levels. These detectors are housed in an underground room built with very thick shielding to eliminate as much background radiation as possible.

Tools for Assessment

Lawrence Livermore comes to the task of dose assessment with a full complement of time-tested tools. Many have been evolving since the days of the Manhattan Project, while others, like computer modeling, are much newer. In addition to the tools described below, we also apply industry-standard radiation detection and bioassay techniques as needed.

Biodosimetry

Biological dosimetry—or biodosimetry—measures the effects of radiation exposure on biological organisms. The goal of biodosimetry is to quantify how an exposure is distributed within an organism when the exposure is known or, when the exposure is not known, to "back in" to the dose from observation of the organism.

Biodosimetry involves establishing a dose-response relationship or following the dose as it is distributed throughout an organism. Dose measurements can be made directly, such as by measuring the radiant energy emitted with a whole-body counter, or indirectly by measuring the dose's biological effects. The biological parameters most often relied on are survival, birth defects, chromosomal abnormalities, chromosome breakage, and chemical changes.

To help us understand the effects of low doses of radiation on people and animals, at Livermore we use the sensitive techniques of chromosome painting and accelerator mass spectrometry.

Chromosome painting using fluorescence *in situ* hybridization (FISH) is a technique developed at Livermore for studying chromosomal changes in cells. Chromosomal DNA is labeled with chemicals that can be stained to produce vivid colors; the colors allow the different chromosomes to be readily distinguished so that chromosomal rearrangements (such as translocations) can be identified. (See the images below.) Studies have shown that these translocations are stable over time,* which makes FISH particularly useful for dose reconstruction. For example, a worker who accidentally inhaled tritium oxide in 1986 was re-evaluated six years later using translocations measured by FISH. The dose, estimated biodosimetrically, confirmed the dosimetry results obtained immediately after the accident.

Accelerator Mass Spectrometry

Accelerator mass spectrometry (AMS) is used to detect a variety of isotopes at very low concentrations. Its sensitivity is typically a million times greater than that of conventional mass spectrometry. AMS was first used for dose reconstruction in 1979 to analyze concrete and other mineral materials from Hiroshima for chlorine-36 produced by the bomb. Today, Livermore's Center for Accelerator Mass Spectrometry, under the leadership of Acting Director Ivan Proctor, is one of the few in the world used for dose reconstruction purposes. (See the photo p. 19.)

Mass spectrometry is used to determine the mass of an atomic species or molecular compound. AMS, as it is applied at Livermore, adds three steps to mass spectrometry. After the initial acceleration to kilovolt energies and the separation of the ion beam by mass to electrical charge, a second acceleration of millions of volts is applied. Then the ion beam is stripped to a charge state where at least three electrons are removed from the atoms of interest, thus destroying all molecular species. The resulting positive ion beam is further accelerated through a third stage of millions of volts. Finally, the isotope has its mass, energy, velocity, and charge redundantly determined to remove background events associated with the instrument.

Livermore's pioneering use of AMS in carbon-14 measurements for biomedical applications led to applications involving other radioisotopes of biological interest, such as tritium (hydrogen-3), aluminum-26, calcium-41, and iodine-129. Today, for example, tritium can be detected at the level of 0.1 parts per quadrillion (10^{15}) and iodine-129 at 40 parts per quadrillion.

A project led by Laboratory scientist Tore Straume is using AMS's advanced separation techniques to measure levels of nickel-63 in copper samples from Hiroshima. These measurements, which sample a different energy slice of the bomb's neutron output, will help resolve discrepancies in earlier, less sensitive measurements of neutrons near the bomb's hypocenter and lead to a more accurate dose assessment.

Biokinetic Models

Livermore was a pioneer in the development of biokinetic models to describe the retention of a radioactive substance in the body or an organ as a function of time. (Biological kinetics, or biokinetics, describes the dynamics of the body—e.g., inhalation, exhalation, absorption, adsorption, metabolism, excretion—any of which may affect the retention of a substance.) Some biokinetic models are specific for a particular organ while others describe whole-body systemic retention.

Biokinetic models are published as internationally accepted default models for use by laboratories. Scientists use the fractional retention from the model coupled with bioassay measurements to derive an estimate of intake. Biokinetic models are also used to project a worker's internal dose for up to 50 years after an intake.

Livermore's Internal Dosimetry Assistance Group has been responsible for updating many biokinetic models. Workers who handle radioactive materials have been regularly monitored since the inception of the Manhattan Project, supplying a wealth of information about individuals and groups of people. The accuracy of internal dose projections 50 years after an intake depends on the adequacy of the biokinetic models used for the projection. We use biokinetic models to calculate the expected results from bioassays over 50 years, which are then compared with measured bioassay levels in the years following an intake. With these data and within the accuracy of the models, we can determine whether additional intakes occurred during the working career of the employee.
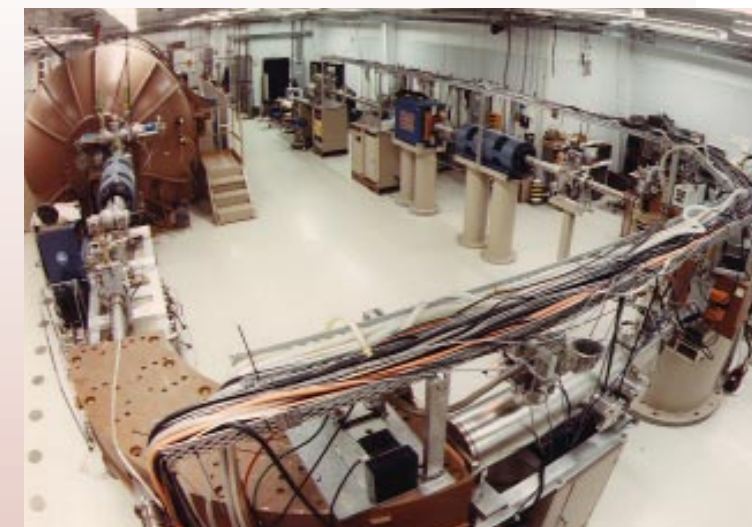
The high-energy spectrometer and particle counter at Livermore's Center for Accelerator Mass Spectrometry.

Likewise, the projection of bioassay levels makes it possible to determine the adequacy of the biokinetic models for humans and to refine these models as data become available, thus improving internal dose projections. Our model-development work provides realistic biokinetic parameters that are used by dosimetrists all over the world to estimate doses to anyone who has been exposed to radioactive materials.

ARAC

Livermore's Atmospheric Release Advisory Capability (ARAC), under Thomas Sullivan, has a very clear charter: to address any kind of radiological release anywhere in the world that could affect a U.S. citizen. Over the years, ARAC has added toxic material modeling and analysis to its repertoire and has helped to assess potential problems arising from chemical and biological warfare conditions.

ARAC's physical resources are built around the AEROS (ARAC Emergency Response Operating System) computer network. Central computers at Livermore support user workstations at 40 designated facilities, continuously exchanging real-time, site-specific meteorological information, local data, and central system–prepared advanced model calculations.

To produce simulation models that show the movement, extent, and magnitude of atmospheric releases, ARAC's experts
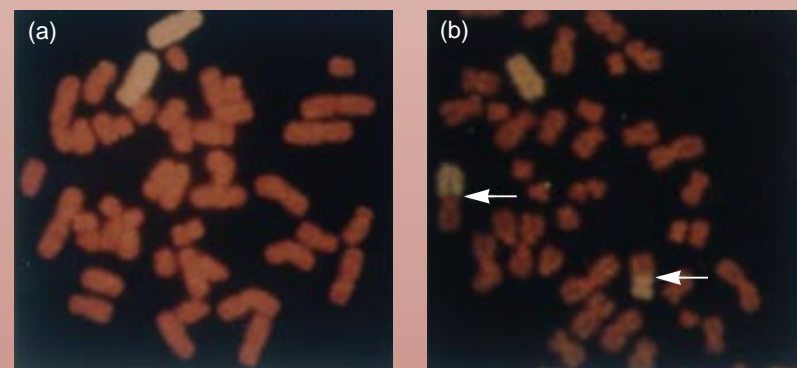
(a) Two chromosome 4s from a normal cell have been stained yellow using FISH (fluorescence *in situ* hybridization), while the other chromosomes have been counterstained red. (b) After irradiation, one normal chromosome 4 is visible, while the other has broken into two pieces and translocated to the corresponding pieces from a broken red chromosome (arrows).

* J. N. Lucas *et al.* "The Persistence of Chromosome Translocations in a Radiation Worker Accidentally Exposed to Tritium," *Cytogenetics and Cell Genetics* **60**, 255–256 (1992); and J. N. Lucas *et al.*, "Stability of the Translocation Frequency Following Whole-Body Irradiation Measured in Rhesus Monkeys," *International Journal of Radiation Biology* **70**, 309–317 (1996).

also rely on three-dimensional atmospheric transport and dispersion models, graphical displays, and extensive databases (e.g., geophysical data, radionuclide and toxic-substance properties, dose conversion factors, topography for any part of the world, a detailed weather network). ARAC can model a radiological accident in the U.S. within 15 to 60 minutes. HOTSPOT, another atmospheric dispersion tool developed at Livermore, can be run for initial assessment in a few minutes on a palm-sized computer.

ARAC has received international recognition for its success in responding to and assessing radioactive atmospheric release accidents throughout the world. Some of ARAC's more notable responses include the re-entry of the U.S.S.R.'s nuclear-powered Cosmos-954 satellite into the atmosphere over Canada (1978), the Three Mile Island accident in Pennsylvania (1979), the accidental release of uranium hexafluoride from the Sequoyah Fuels Facility in Oklahoma (1986), as well as the Chernobyl

reactor accident in the former Soviet Union. Other responses involved nonradioactive releases, including the oil fires in Kuwait and the eruption of Mount Pinatubo in the Philippines, both in 1991.

In an effort to be prepared for accidents like Chernobyl, for nuclear terrorism, and for other radiological events, ARAC also provides an emergency response and assessment service to many federal agencies. One aspect of this service is preparing assessments of the atmospheric dispersal of radionuclides from likely accidents so that planners can put in place the resources to deal with them. ARAC has done many such studies for facilities at the Nevada Test Site, at LLNL, and at other DOE sites.

ARAC also provides input to and participates in a variety of training exercises. We can model accident scenarios provided by a client by using real-time meteorology at the client's site, thus enhancing the realism of the exercise. ARAC can also "create" special meteorology to test a particular aspect of a site's response.

(continued from page 17)

William Robison began a radioecology and dose assessment program in 1973 that continues today.

We have taken nearly 70,000 samples of edible food crops, vegetation, soil, water, marine life, and animals to evaluate the various exposure pathways for radiological dose. At several atolls, we have also completed measurements of external gamma exposure and studies of the extent to which the resumption of human activity would cause nuclides in the surface soil to be resuspended into the air and thus inhaled. Results indicate that the terrestrial food chain is the most significant exposure pathway, with external exposure to gamma radiation the next most significant.

Combining the radionuclide inventory from the samples, a diet model for the population, and biokinetic models (see box on pp. 18–20), we have determined that the dose from the ingestion pathway contributes 70 to 90% of the total estimated dose. Cesium-137 would produce 96% of the estimated dose for

returning residents, mostly through uptake from the soil to terrestrial food crops. Inhalation accounts for about 1% of the estimated dose, according to surface soil levels and resuspension studies.

Our dose assessments indicate an estimated maximum annual effective dose at Bikini Island of 4 millisieverts.* This is based on a resettlement date of 1999 and assumes that no remedial measures are taken and that imported foods, which are now an established part of the diet, continue to be available. (In comparison, the average annual U.S. dose from all forms of natural background radiation is about 3 millisieverts.)

To reduce cesium-137 in food crops, we recommended treating them with potassium fertilizer, which would reduce the uptake of cesium-137 and therefore the ingestion dose to about 5% of pretreatment levels. Removing the top 40 centimeters (15.7 inches) of soil over the entire island would also

effectively reduce the potential dose, but at a high environmental and dollar cost. If the top 40 centimeters of soil were removed in just the housing and village areas and the rest of the island treated with potassium fertilizer, the maximum annual effective dose would be reduced to 0.41 millisieverts, which is comparable to the effective dose from two chest x rays.

This information has been provided to the Marshall Islanders, but they have not yet decided how they wish their islands to be rehabilitated. In the meantime, sampling continues (see Figure 3) with a focus on further characterization of the sampled areas, more systematic sampling of the atolls, and countermeasures to reduce doses.[4]

The Job Ahead

In all of our dose assessment work at Livermore, we strive to understand not only the effects of high levels of exposure from atomic weapons and reactor accidents but also of very small

exposures. And as we develop new methods and tools, our dose estimates can be made increasingly accurate. For example, a revised biokinetic model for a particular organ may prompt a new retrospective study to update an internal dose that had been based on an old model. The more we know about the effects of radiation, the better we can prepare for future accidents, educate our workers and the public about radiation's risks, and manage those risks to mitigate them.

—Katie Walter

Key Words: accelerator mass spectrometry, Atmospheric Release Advisory Capability (ARAC), biodosimetry, biokinetic model, BRAVO test, Chernobyl accident, dose reconstruction, dose assessment, FISH (fluorescence *in situ* hybridization), Marshall Islands, prospective dosimetry, worker exposure.

Figure 3. Livermore environmental scientists in the field at Bikini Island are collecting coconut samples at one of the experimental potassium fertilizer plots.

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About the Scientists

Radiation dose assessment at Livermore is multidisciplinary in a broad and deep sense. The scientists whose work contributed to the Laboratory's dose assessment capability reported on in this article are pictured left to right: TORE STRAUME, WILLIAM ROBISON, JOE LUCAS, THOMAS SULLIVAN, and LYNN ANSPAUGH. (Not shown are IVAN PROCTOR, and DAVID HICKMAN.) Their expertise and that of the groups they lead span a variety of disciplines—biology and genetics, biodosimetry, dose reconstruction, accelerator mass spectrometry, radioecology, biokinetic modeling, atmospheric dispersion and transport modeling, and emergency preparedness and response. Their scientific training and experience represent a Laboratory capability in dose assessment recognized for its excellence worldwide.

* Millisievert is a Standard International Unit for measuring dose to humans; 1 millisievert equals 100 millirems.