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METHODS IN ENVIRONMENTAL SAMPLING FOR RADIONUCLIDES

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Richard C. Ragaini[†]Summary

This paper reviews methods of environmental sampling for radionuclides around operational and preoperational nuclear power plants. We examine in detail the implications of the established radiation standards and their effect on sampling procedures. Transport mechanisms of radionuclides in liquid effluent, and the deposition of airborne radionuclides onto soil and vegetation are discussed. We evaluate water- and soil-sampling procedures. The Lawrence Livermore Laboratory program of terrestrial gamma-ray surveys at preoperational nuclear power plants is described.

Introduction

The National Environmental Policy Act of 1969 requires preparation of an environmental impact statement for each nuclear plant by the Nuclear Regulatory Commission (NRC). There are many similar state and local regulations. To satisfy the regulations, the electric utilities must: (1) study the ecosystems surrounding the plant before operation so as to predict the impact and to minimize adverse effects, and (2) monitor the plant vicinity during operation so as to determine the actual impact.

In this paper a general overview is given of the methodology for monitoring radioactivity in the soil, vegetation, and water around nuclear power plants. Specific sampling methods, equipment, and procedures are discussed, as is general sampling design.

Radiation Standards

Rules governing environmental monitoring have been published by the NRC and other agencies.¹⁻⁵ The NRC has stated that releases of radioactivity in gaseous and liquid effluents from light-water-cooled power reactors must be kept "as low as practicable," so as to keep additional radiation exposure to persons living near nuclear power plants at small fractions of both natural background radiation levels and existing radiation protection limits. But the specific design objectives of the regulations⁶ are: (1) to limit the amount of radioactivity released in liquid effluents from any light-water-cooled power reactor to levels that would keep the annual exposure to an individual in an unrestricted area to not more than 3 millirems (mr) for the whole body and not more than 10 mr to any organ; (2) to limit releases of radioactivity in gaseous effluents from any light-water-cooled power reactor to keep annual exposures to an individual in an unrestricted area to a maximum of 5 mr to the whole body, and not more than 15 mr to the skin; and (3) to limit releases of radioactive iodine and other radioactivity from any light-water-cooled power reactor to keep annual exposures to the thyroid of an individual in an unrestricted area to no more than 15 mr.

Existing radiation protection standards are designed in terms of permissible human exposure levels rather than lowest practicable emission levels. They limit to 500 mr, the total annual exposure to an indi-

vidual in the general population from all sources of radiation except natural background and medical sources. Because of the time-lag involved in calculating doses, operating limits have been derived for application at the sources. For example, concentration guides for air and water have been provided.⁶ The dose is calculated from measured concentrations to demonstrate compliance with regulations. Standardized human parameters are normally used in these calculations,⁷ however care must be taken, because local dietary and recreational habits may have significant effects on the actual doses, and because these concentration guides do not take into account exposure which might result from multiple sources or from enrichment mechanisms in the environment.

The EPA Surveillance Guide⁸ recommends methods for conducting a minimum-level off-site environmental radiation monitoring program. However, this program addresses off-site measurements only and must be supplemented by on-site measurements in the stack and in the liquid effluent stream. The number, the source media, and the source location of samples may vary from site to site. At times it may not be possible or practical to obtain samples of the media of choice at the most desired location or time. In these instances, suitable alternates may be chosen for the particular pathway in question and submitted for acceptance. The suspected presence of fresh fallout from weapons testing or other sources may warrant more frequent sampling and more detailed analyses. The recommended program includes monitoring the four basic exposure pathways: air, water, food, and external radiation. We will discuss here the monitoring of water and sediments as well as soils and vegetation.

Monitoring Programs

The purpose of environmental monitoring is to describe the impact of a given nuclear power plant at a specific time and location and to obtain data that can be used as a basis for future action. The monitoring must be carried out with an understanding of the potential impacts and the existing physical conditions around each plant.

Monitoring takes place in three stages: initial studies, preoperational monitoring, and operational monitoring. Initial studies are carried out in the vicinity of the plant site to identify possible impacts and to predict the magnitude of the expected changes. Preoperational monitoring is carried out to determine the characteristics of the ecosystems at the plant site before operation.

Other purposes of preoperational monitoring are: (1) to design a monitoring program for ultimate use in the operational stage, (2) to take background measurements for comparison with operational data, (3) to develop and evaluate sampling measurement procedures and laboratory analysis capability, (4) to detect previously unevaluated impacts, (5) to measure radiation from other sources such as weapons test fallout, (6) to collect data that can be used in the required environmental impact statements, and (7) to identify critical pathways for radionuclide transport from the source to man. Operational monitoring is carried out to ensure that adverse environmental impacts are minimized and that regulations are complied with.

Environmental measurements fall into two categories, *in-situ* field measurements using mobile or

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fixed instruments and measurements made on samples taken in the field and brought back to the laboratory. Either continuous (composite) or grab samplings may be taken.

Grab samples and laboratory analysis are usually adequate for routine environmental surveillance conducted to evaluate contamination rather than to detect radionuclide releases. The magnitude of any release, however, should be determined at the release point. Reactor effluent measurements made there may in addition give reasonable estimates of environmental concentrations.

Specific monitoring methods should be developed individually for each reactor site. Surveillance programs will vary slightly for the different reactor types because of the differences in radionuclide releases. Pressurized-water reactors (PWR) release more tritium in the liquid effluents, while boiling-water reactors (BWR) release more short-lived halogens and noble gases in the stack airborne effluents.

Preoperational and operational environmental measurements around power reactors and other nuclear operations raise specific questions with regard to radiation standards, measurement problems, sampling procedures, instrumentation, and analysis methods. Radiation standards, measurement problems, and sampling procedures are discussed here. For detailed evaluations of commercially available instrumentation and government laboratories' instrumentation, see the survey conducted recently by the Lawrence Berkeley Laboratory.¹¹ Analysis methods are discussed in another paper in the symposium.

Water Sampling and Analysis

Figure 1 illustrates the pathways of released radionuclides through ground and surface waters to man.⁶ The drinking water pathway is considered most significant. Table I, taken from the EPA Surveillance Guide⁶ and from Ref. 11, lists the recommended pro-

cedure for procuring samples for laboratory analysis from surface, ground, and drinking water, as well as from sediments. On-site water measurements involving direct in-stream instrumentation will not be discussed here.

An important distinction is that between composite and grab sampling. In determining what type of sampling is needed, the following factors must be considered: (1) the frequency of concentration fluctuations, (2) the purpose of the survey (whether it is designed for trend evaluation or for human dose calculations), and (3) the half-life of the particular radionuclides in question.

A minimum sample volume of 1 l is recommended. A smaller volume is acceptable if the sample can be considered representative. In any case, this procedure should be evaluated in the preoperational survey. Sample containers and samples to be stored for more than 24 hours should be pretreated to minimize wall absorption. Used containers should be discarded so as to avoid cross contamination.

Proper water sampling is difficult. Streams containing settleable solids require special techniques. Also, extremes in water flow may bias sampling results. When improper sampling techniques are used, changes in stream flow patterns may produce variations that appear to be seasonal. If the chemical characteristics of the stream change, precipitation or redissolution of radionuclides may occur. In large bodies of water, stratification and thermal inversions are common. All of these conditions must be taken into account in a water sampling program.

Surface Waters and Sediments

Liquid effluents will disperse immediately on contact with surface water, and the rate of dilution will depend on the relative volumes and flow rates of the liquid effluent and the receiving water. In a river the dilution will be proportional to the ratio of flow rates of the effluent and the river.¹¹

The sediments of the receiving waters will contain some of the discharged radionuclides. The concentration in the sediments is governed by the effluent release rate, the stream velocity, and the presence of organic debris. Mixing processes in eddies, confluences, or impoundments may also increase sediment concentrations.

The most dangerous radionuclides in surface water and sediments are ¹³⁷Cs and ¹³⁴Cs.¹² Tritium may be present in larger concentrations, but it has less dose significance.

Surface water should be collected routinely at a background location, at the edge of the mixing zone of the effluent, and at the nearest point of use downstream. Automated continuous sampling is recommended for downstream samples.¹¹ Fifty ml should be taken about every half-hour, and the samples composited for weekly analysis. It is desirable, but not required by the regulations, to take samples with volume proportional to the flow rate. The samples are then self-weighting and they provide a statistically unbiased estimate of the time-averaged weekly concentrations. Grab or constant-volume samples may miss peaks and valleys in radionuclide concentrations.

Background concentrations are estimated by samples up-stream on a river or up-current in lakes or oceans. Monthly grab samples may be adequate to estimate annual average background levels, but it may be

Fig. 1. Pathways of radioactive materials released to ground water, surface water, and oceans (from Ref. 5)

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Table 1. Offsite radiological surveillance for an operating light-water-cooled nuclear power reactor (taken from Refs. 4 and 11).

Sample	Number and locations of samples ^a	Sampling frequency	Type and frequency of analysis
Surface water	1 sample in immediate area of discharge 1 sample upstream 1 sample downstream after dilution ^b	Collect immediately following liquid discharge, allowing sufficient time for movement downstream with current to verify dilution. Monthly for continuous discharge ^c	Tritium and gamma isotopic analysis upon collection, composite for tritium and Sr-90,90 analyses quarterly
Ground water	1 or 2 samples from sources most likely to be affected ^d 1 sample from groundwater source upgradient	Quarterly	Gamma isotopic and tritium
Drinking water	1 sample at each of 1 to 5 supplies obtained within 10 miles of the facility which could be affected by its discharge, or the first supply within 100 miles if none exist within 10 miles	Continuous sampler operation, proportional composite sample ^e	Gamma isotopic weekly, gamma isotopic analysis monthly, composite for tritium and Sr-90,90 analyses quarterly
Sediment	1 sample in area immediately downstream from discharge point ^f 1 sample upstream from discharge point 1 sample at first downstream impoundment	Semiannually	Tritium, gamma isotopic, and Sr-90,90 analyses semiannually

^aThe number, media, and location of sampling may vary from site to site. It is recognized that, at times, it may not be possible or practical to obtain samples of the media of choice at the most desired location or time. In these instances suitable alternate media and locations may be chosen for the particular pathway in question and submitted for acceptance. In addition, the suspected presence of fresh fallout from weapons testing at other sources may warrant more frequent sampling and more detailed analyses.

^bFor facilities not located on a stream, the upstream sample should be taken at a distance beyond significant influence of the discharge. The downstream sample should be taken in an area beyond the outfall which would allow for mixing and dilution. Upstream samples in a tidal area must be taken far enough upstream to be beyond the plant influence when the effluent is actually flowing upstream during incoming tides.

^cGenerally, seawater measurements are not required, except when the vicinity of the plant is utilized for recreational activities.

^dGroundwater samples should be taken when this source is tapped for drinking and/or irrigation purposes in areas where the hydraulic gradient or recharge properties are suitable for contamination.

^eIf a reservoir is used, drinking water samples should be taken from the reservoir monthly. If the holding time for the reservoir is less than 1 month, then the sampling frequency should equal this holdup time. Increases in concentration of activation and/or fission products at these sources will necessitate the analysis of tap water for the purpose of dose calculations.

^fOther sampling locations may be required on specific sites. For further guidance, see pertinent footnotes in Refs. 4 and 11.

desirable to take samples weekly to compare with the downstream samples.

If the sampling location is near the line of homogeneity of the mixing zone, grab sampling is adequate. Otherwise, multiple sampling based on diffusion and transport studies of the mixing zone is necessary.

Stream sediments provide a record of deposition from liquid releases just as soil samples provide a record of deposition from airborne releases. Figure 2 suggests sampling locations^g for routine stream sampling. Samples near the edge of the mixing zone are used to estimate the initial contaminant concentra-

tions in the sediments. The downstream sample should be taken at the point of fastest flow. To estimate the maximum accumulation, samples should be taken at the first impoundment or bend. If the plant is located on a lake or ocean, the sediment sample should be taken near the outfall, but beyond the turbulent area.

Sediment samples can be taken in a bottle or pail in shallow, slow-moving streams. In deep streams a dredge must be used, but some of the sediment may be washed out while bringing it to the surface. Sediment samples should weigh at least 1 kg and should consist of the most recent sediment. Core samplers may be used for compacted sediment.

Fig. 2. Suggested river sediment sampling locations (from left, 1).

Drinking Water

The radionuclide concentrations in drinking water will depend on the dilution of the effluent and the radionuclide decay factors during transit to the sampling point. As in the case of surface water and sediments, tritium will generally have the highest concentration, but the most important radionuclides from a dose point of view will be ^{137}Cs and ^{134}Cs .

Chemical treatment of drinking water will affect radionuclide concentrations. Chlorination precipitates the ferric group and affects only a few radionuclides. Filtration removes radionuclides on suspended particles. Flocculation and filtration remove a fraction of the dissolved radionuclides. Sorption water treatment will not affect the tritium concentrations.

Samples should be taken from the first point of public withdrawal (usually the first town or farm downstream from the plant) and should establish the maximum amount of radioactivity in the water supply. The sampling location should be the users' inlet line. To improve the accuracy of the dose estimates, treated water samples may also be taken. Drinking water must meet U.S. Public Health Service standards¹¹ in addition to SRC and EPA standards.

Ground Water

Inadvertent contamination of ground water can occur by leakage from waste lines or storage basins, or by recharge from surface streams receiving liquid effluents. To evaluate potential effects, a minimum amount of ground-water monitoring is required. The nearest down-gradient well should be sampled. Three wells should be sampled if the direction of ground water travel is not well known.

In soils with exchange capacity, most radionuclides are chemically bound near the source, and decrease in concentration down gradient from the source. Several radionuclides, including tritium (as HTO), radiocesium, and radioruthenium, are only slightly soil bound, and therefore can be used as indicator radionuclides. If the liquid discharge to the soil is continuous, it is possible that the exchange capacity could be exceeded. A contamination front will then proceed in the direction of the ground water flow. Local ground water studies are almost always required to provide reliable quantitative estimates of travel times and down-gradient concentrations of liquid effluents. Most ground waters have detectable amounts of natural uranium and thorium (and their daughters) as well as tritium from weapons tests.

Systematic sampling of piezometric wells and specially drilled monitor wells provides information on rates of concentration levels useful for monitoring trends. The frequency of sampling in the monitor wells to be monitored must be determined from local ground water conditions. The nearest down-gradient well should be sampled.

Ground water sampling should be scheduled to provide grab samples. If the flow rates and concentrations in levels are suspect, it is recommended that samples be collected at regular intervals using a peristaltic pump.

Standard methods for water analysis for contamination substances are described in Ref. 12. Laboratory analysis for radioactive substances should include gross β measurements, tritium determinations, ^{89}Sr and ^{90}Sr determinations by radiochemical separation, and gamma spectroscopy techniques. For the regulatory code full recommendation for laboratory methods and analysis procedures, which include minimum activities: 10^{-2} $\mu\text{Ci/l}$ for ^{137}Cs , ^{134}Cs , ^{131}I , ^{132}I , ^{133}I , ^{134}I , ^{135}I , ^{136}I , ^{137}I , ^{138}I , ^{139}I , ^{140}I , ^{141}I , ^{142}I , ^{143}I , ^{144}I , ^{145}I , ^{146}I , ^{147}I , ^{148}I , ^{149}I , ^{150}I , ^{151}I , ^{152}I , ^{153}I , ^{154}I , ^{155}I , ^{156}I , ^{157}I , ^{158}I , ^{159}I , ^{160}I , ^{161}I , ^{162}I , ^{163}I , ^{164}I , ^{165}I , ^{166}I , ^{167}I , ^{168}I , ^{169}I , ^{170}I , ^{171}I , ^{172}I , ^{173}I , ^{174}I , ^{175}I , ^{176}I , ^{177}I , ^{178}I , ^{179}I , ^{180}I , ^{181}I , ^{182}I , ^{183}I , ^{184}I , ^{185}I , ^{186}I , ^{187}I , ^{188}I , ^{189}I , ^{190}I , ^{191}I , ^{192}I , ^{193}I , ^{194}I , ^{195}I , ^{196}I , ^{197}I , ^{198}I , ^{199}I , ^{200}I , ^{201}I , ^{202}I , ^{203}I , ^{204}I , ^{205}I , ^{206}I , ^{207}I , ^{208}I , ^{209}I , ^{210}I , ^{211}I , ^{212}I , ^{213}I , ^{214}I , ^{215}I , ^{216}I , ^{217}I , ^{218}I , ^{219}I , ^{220}I , ^{221}I , ^{222}I , ^{223}I , ^{224}I , ^{225}I , ^{226}I , ^{227}I , ^{228}I , ^{229}I , ^{230}I , ^{231}I , ^{232}I , ^{233}I , ^{234}I , ^{235}I , ^{236}I , ^{237}I , ^{238}I , ^{239}I , ^{240}I , ^{241}I , ^{242}I , ^{243}I , ^{244}I , ^{245}I , ^{246}I , ^{247}I , ^{248}I , ^{249}I , ^{250}I , ^{251}I , ^{252}I , ^{253}I , ^{254}I , ^{255}I , ^{256}I , ^{257}I , ^{258}I , ^{259}I , ^{260}I , ^{261}I , ^{262}I , ^{263}I , ^{264}I , ^{265}I , ^{266}I , ^{267}I , ^{268}I , ^{269}I , ^{270}I , ^{271}I , ^{272}I , ^{273}I , ^{274}I , ^{275}I , ^{276}I , ^{277}I , ^{278}I , ^{279}I , ^{280}I , ^{281}I , ^{282}I , ^{283}I , ^{284}I , ^{285}I , ^{286}I , ^{287}I , ^{288}I , ^{289}I , ^{290}I , ^{291}I , ^{292}I , ^{293}I , ^{294}I , ^{295}I , ^{296}I , ^{297}I , ^{298}I , ^{299}I , ^{300}I , ^{301}I , ^{302}I , ^{303}I , ^{304}I , ^{305}I , ^{306}I , ^{307}I , ^{308}I , ^{309}I , ^{310}I , ^{311}I , ^{312}I , 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^{913}I , ^{914}I , $$

Fig. 3. Pathways of radioactive materials released to the atmosphere (from Ref. 3).

The most useful measure of radioactive concentration is amount per unit area. Thus, it is important to know the depth of the sampling, the bulk soil density, and the concentration distribution with depth.

Site selection

A critical factor governing the validity of direct sampling is whether or not it is representative of the environment. Any sampling should reflect variability with time and location. Therefore, the number of sites, the site locations, and the sampling frequency are critical. Statistical methods are useful for designing sampling plans for environmental surveys.^{17,18} However, an initial exploratory sampling and an test program should be carried out first in any case to evaluate the variability factor. Then statistical techniques can be applied to calculate the number of samples required for a given set of points in time and space.

Different sampling schemes are described qualitatively by Slonim,¹⁵ in Cochran,¹⁶ which is considered the standard text, the technical details of each are given, with all the accompanying algebra. There are three basic ones:¹⁷ systematic or grid sampling, random sampling (encompassing simple random sampling and stratified sampling), and judgment sampling.

Systematic Sampling. This method involves laying

Fig. 4. Preliminary soil-sampling sites near a nuclear facility (from Ref. 19).

out a grid of uniformly spaced sample locations. The advantages are general simplicity and easy implementation in the field. However, one disadvantage becomes apparent when there is a systematic pattern in the radionuclide distribution. In such a situation, the measured radionuclide concentrations will be too high or too low depending on the nature of the overlap of the grid and the concentration pattern. Another disadvantage is the uncertainty in the variability of the radionuclide concentration. Figure 3 shows a distribution of sampling sites suggested in SRC Regulatory Guide 4.5. The Guide advises that 13 sites (out to 16 km downwind) should provide an adequate picture of background levels around a plant.¹⁹ It is also suggested that no or more samples be taken close to the center of the most heavily populated area near the plant.

Random Sampling. Under this scheme, sample location coordinates are chosen by some chance process where each coordinate has a known selection probability. In simple random sampling the selection probabilities are all equal. A grid is laid out and the sample coordinates are selected from a table of random numbers. When sampling is conducted to satisfy radiation standards, it must be random because then only will it have statistically predictable attributes.

One type of random sampling in which the selection probabilities are weighted is called stratified sampling. Several separate strata are formed by grouping units having similar levels of radioactivity. Then each stratum is randomly sampled. For example, one might use a field meter to survey for gross gamma-ray activity in a potential soil-sampling area. The area could then be broken down into smaller areas which have different count rates. Then each area could be randomly sampled. Past experience²⁰ has shown that the variability of radionuclide concentrations increases with the mean concentration. Therefore, the number of samples required in a stratum increases with its mean concentration and its size.

Judgment Sampling. Here, the sample locations are chosen on the basis of someone's judgment. Such samples are almost always biased. The sampling in most ecological studies is based to a greater or lesser extent on the investigator's judgment.

Johnston¹⁸ has derived an "index of adequacy" to evaluate the statistical validity of a given sampling program. This index depends on knowing: (1) how much the concentration of each particular radionuclide

varies in the environment, (2) the confidence level desired by the scientist, and (3) the level of statistical risk to the population.

Site selection is critically important to correct interpretation of the data. The site must remain undisturbed for the time interval of interest. It should be at least 125 m (400 ft) away from the edge of any construction. When soil is sampled as part of a preoperational survey, areas should be selected so that they can be resampled later. A reasonable sampling frequency is every three years.

The site should be representative in terms of meteorological and climatological factors. Generally it should be at the center of a large flat open area. It should be located near air sampling sites and food crop sampling sites. Lawn areas or areas with short stands of vegetation are desirable because ground cover tends to hold the deposits in place. High stands of vegetation or tree canopies may act as filtering agents and limit deposition. In such cases it may be necessary to sample the grass and soil separately.

The site should be nearly level with moderate to good permeability. There should be little runoff and no overruns during heavy rains. Crests of ridges, level farm land, and lawns are frequently ideal. In some instances a worst case situation should be selected so that excessive accumulations occurring in low spots, at the foot of slopes, or in flooding areas can be measured. Soils having high earthworm or rodent activity will be unevenly mixed down to significant depths.

Vegetation Sampling. Vegetation samples should be taken at the same time and place as soil samples so that total deposition can be estimated. Perennial vegetation provides the best data on deposition of airborne radionuclides. Hand clippers can be used for sampling vegetation. A minimum of 1 kg should be collected for analytical sensitivity. Food crops or forage can be sampled separately to measure the concentration of radionuclides in the food chain. Forage should be sampled when it is being grazed. Vegetation and soil sampling locations should be near air sampling locations for comparison purposes. Samples should be taken of the same vegetation species at each location.

In areas covered with tall grass, the grass and the soil should be sampled separately. The procedure is to clip the grass in a 1-m square down to 2 cm from the ground. The soil is then sampled by taking five cores. Two such plots should be sampled to achieve the proper area representation. The grass can be analyzed separately or analyzed with the soil. Since the total surface-deposited contaminants are of interest, the vegetation must also be included in the analysis.

General Principles of Soil Sampling. No single soil-sampling method is adequate for all soil types and all locations. Methods which work well in sandy loam may not work well in dry, loose soils. Two procedures are described in Guide 4.5: a coring method of general utility used by the Health and Safety Laboratory (HASL)²³ and a depth profile technique useful for dry sandy soil. Coring means the side of the sample and should be limited to areas having a finely divided and workable soil structure. The depth profile method does not work well in rocky or stony soils. If the rock problem is severe, it is usually advisable to change sampling sites. If trowing is not feasible, larger diameter cores can minimize the effects. All

rocks should be included. After the sample is dried and weighed, the rocks can be removed by sifting.

The probable uniformity of deposition should govern the size and number of samples. Core depths of 17 to 30 cm may be necessary, depending on local rainfall and tillage practices. To prevent mixing, it may be necessary to freeze the cores before segmenting. With the proper techniques and tools, it is feasible to sample the top 1.25 cm. However, in sites with heavy vegetation and high precipitation, a surface depth of 5 cm is recommended. A thick root mat may prevent removal of even a 5-cm plug, in which case a deeper plug should be cut. A total surface area of 460 to 930 cm² (1/2 to 1 ft²) provides a reasonably good estimate of the actual radioactive deposit.²⁴

On a test plot of 9 m² (100 ft²), 5 to 10 separate cores of several inches in diameter may be composited to provide the recommended sample of 200 g dry. The procedure used at HASL²³ for ⁹⁰Sr assays uses 10 cores taken in a 4.6-m (15 ft) straight line. At LLL,^{21,22} we have been taking five cores at the corners and center of a 7 m × 7 m (10 ft × 10 ft) square, and have composited them at the Laboratory.

Anything that will take a core or a plug and that has equal cross-sectional area, through its entire depth may be used for taking the soil sample. For surface sampling a 5 cm (deep) × 8 cm (diameter) cookie cutter is adequate. For coring we have found a 25-cm depth to be sufficient in soils with reasonable cation exchange capacity.

HASL Coring Method. The sampling tools used at HASL²³ are an 8.9-cm-diameter (1.4-in) topsoil cutter that takes a 5-cm-deep (2.0-in) sample, and an 8.9-cm-diameter (1.25-in) barrel orchard auger that cuts an 8.9-cm-deep (3.5-in) sample.

The topsoil cutter is used to remove the soil to a depth of 5 cm, and the auger is used to take an inner sample to the desired depth. The soil from ten 10-cm cores is composited to make a single sample weighing from 18 to 16 kg; the total area sampled with this method is 620 cm² (9.67 ft²). In desired, the soil samples (5 cm in depth) may be kept separate.

HASL Depth Profile Method. This method²⁶ of taking soil samples at various depths is advisable in sandy loam only. When used in other soils, it may result in contamination of subsol layers. The procedure is as follows:

- (1) Lay a tarpaulin (about 2 m²) near the clipped area.
- (2) Dig a trench about 60 cm wide, 90 cm long, and 60 cm deep next to the clipped area, placing the dirt on the tarpaulin.
- (3) Make the trench soil perpendicular to the surface by smoothing the sides with a trowel or shovel.
- (4) Take the first sample by pushing a three-sided square pan (15 cm × 15 cm × 5 cm deep) with cutting edges on the open side into the wall of the trench 5 cm below surface.
- (5) Score the first cut with a sharp knife, remove the sample and seal it in a plastic bag.
- (6) Enlarge the flat shelf by cutting away a 15-cm by 15-cm area of topsoil on either side of the sample cut to a depth of 5 cm.

- (7) Brush away soil particles from the shelf.
- (8) Repeat the sampling procedure, and continue until the desired depth is reached.
- (9) When all the samples have been taken, fill the trench back with dirt from the tarpaulin.

This technique samples 225 cm² of surface area at one location. Other locations must be sampled to make the data representative of the area.

LLL Coring Method. At the Lawrence Livermore Laboratory (LLL), we have used a circular corer (shown in Fig. 5) developed by Lindeken at LLNL.²² Its

Fig. 5. Soil coring tool used at LLL for sample collection.

cylinder is 6.35 cm in diameter and 25 cm in length, and it is equipped with a specially hardened cutting edge to minimize damage in rocky soils. The replaceable cutting edge is shrink-fitted into the barrel of the sampler. Mechanical support is provided by a steel collar welded to the top of the sampler, and the collar is protected by an adiprene cover plate. Cross-contamination between samples is eliminated by using a new plastic liner inside the corer each time the sampler is driven into the soil. The liner rests on the shoulder of the cutting edge insert and is prevented from rising by a lucite spacer.

The procedure used with this corer is as follows:

- (1) An undisturbed site which meets the necessary criteria is selected. A 3 m x 3 m (10 ft x 10 ft) square is laid out. Coordinate distances are measured to fixed landmarks.
- (2) The liner is inserted into the corer. The spacer is laid on top of the liner, and the adiprene cover is placed on top of the assembly.

- (3) The corer is driven into the soil with a 10-lb sledge hammer until the adiprene cover plate is in contact with the soil.
- (4) Rods are inserted into holes in the collar, and the corer is rotated as it is pulled out of the ground.
- (5) The corer is placed on the ground, and the liner is capped at the top. The sampler is inverted to allow the liner to fall out, and the bottom of the liner is capped.
- (6) The liners are stored upside down so that high-activity soil will not be shaken down into low-activity soil.

This corer does not perform well in powder-dry loose-grain soils. They fall out the bottom as the sampler is pulled out of the ground. In these soils, a new step 4 is used. A hole is dug next to the corer. A shovel is inserted under the corer, and the assembly is lifted out on the shovel.

Normally, the 25-cm sample core is sectioned (beginning at the top) into segments of 2, 3, 5, 5, 5, and 5 cm in length. The depth segments from the five cores are then composited to cover a surface area of 160 cm².

Figure 6 compares the ¹³⁷Cs concentrations from five cores taken in a 9-m² (100-ft²) plot located near the Rancho Seco Nuclear Generating Station at site TLD-10.^{23,24} The slopes are 0.10, 0.13, 0.16, 1.1, and 1.19 cm⁻¹. Although the slope differences are significant, the concentrations agree quite closely. They are 50, 55, 57, 50, and 44 nCi/m². The composite concentration is 53 nCi/m², while the average is 50 ± 10 nCi/m².

LLL Depth Profile Method. Another sampler in use at LLL is shown in Fig. 7. It was developed at the University of Utah by Dr. Robert Lindsten.²⁵ It is essentially a 10-cm cube made out of welded steel with two open sides composite one another and a sharpened bottom edge. Two-cm increments are scribed on the sides. It is used in the following manner.

- (1) The sampler is pushed into the soil to the 10-cm depth. A small oak block is placed on the top and a light sledge hammer is used.
- (2) The soil is cleared away around the sampler down to the 8-cm depth and out to about 10 cm.
- (3) Each successive 2-cm increment is scooped up with a flat trowel or spatula and stored in a container.
- (4) When the 8-cm depth is reached, the sampler is driven down to 15 cm and the scooping procedure is repeated. Finally, the corer is driven down to 20 cm, where the final sample is taken.

This procedure results in 13 depth increments.

Several comparisons have been made with the two soil samplers at the same site.^{23,26} The results are shown in Table II. The results for the circular cores are composites of five cores (160 cm²), while the results for the square sampler core from one sample (100 cm²). The profiles, shown in Fig. 8, agree quite well. At site TLD-G, the concentration in the top sample (2 cm deep) is depressed for both corers. This

Fig. 6. Comparison of ^{137}Cs profiles from five cores and the composite in a 100 ft² plot Site 110-19 near Rancho Seco Power Plant. The core profile slopes are 0.10, 0.13, 0.36, 1.14, and 1.19 m^{-1} . The concentration, (in parentheses) for the composite is 44 : 1 nCi/m² while the average of the five cores is 50 : 10 nCi/m².

occurred quite frequently in the samplings. It is caused by mixing of the grass roots in the soil, which lowers the sample soil weight and the ^{137}Cs concentration on a per gram basis.

Sample Analysis

Soils can be stored in polyethylene bags or bottles. Vegetation is subject to spoilage, and it

should be frozen or dried immediately after sampling. Soil preparation procedures are as follows:

- (1) Break up the soil clumps and pull off the vegetation and root mat for the surface sample. Mix in the vegetation homogeneously, and weigh the sample.
- (2) Air-dry the sample for three days or dry it in a forced-air oven for 24 hours at 50°C.

FIG. 7. Diagram of soil sampler with square cross-section.

Table 1. Comparison of ^{137}Cs soil concentrations using square sampler and circular corer.

Fort Calhoun Site	Slope (cm^{-1})		Concentration (mCi/m^2)	
	Square	Circular	Square	Circular
TLD-G	0.09 ± 0.004	0.18 ± 0.007	139 ± 5	133 ± 11
GM-6	0.11 ± 0.01	0.15 ± 0.02	185 ± 9	175 ± 13

- (5) After drying, weigh the sample again to determine the weight-loss from drying.
- (6) Remove the rocks and weigh them separately.
- (7) Crush the soil sample, and blend it for about 30 minutes.
- (8) Ball-mill or pulverize the sample, and pass it through a coarse sieve with a pore size no larger than 100 mesh.
- (9) Store the sample in a polyethylene bottle.
- (10) Collect a subsample by spreading out the sample, quartering it, and taking scoops from each quarter until the desired sample weight (not less than 200 g) is collected.

For gamma-spectral analysis at LLL, the soil samples are first shaken and mixed thoroughly. Then 200 to 300-g aliquots are sealed in 200-ml thin-walled (8-10 mil) aluminum cans. The cans are counted for all gamma-emitting radionuclides in shielded $\text{Ge}(\text{Li})$ spectrometers equipped with Compton-suppression systems.²⁴ Results are in nanocuries per gram of dry soil. The field-bulk density, the area, and the depth should be recorded. The field activity is then expressed in nanocuries per unit area.

FIG. 8. Comparison of the ^{137}Cs profiles of samples taken with a square-cross-section sampler and a circular-cross-section corer.

LLL Terrestrial Gamma-Ray Surveys at Preoperational Nuclear Power Plants

Baseline terrestrial gamma-ray surveys have been carried out at five proposed sites: Fort St. Vrain, Colorado; Fort Calhoun, Nebraska; Cooper Nuclear Station, Nebraska; Rancho Seco, California; and Diablo Canyon, California.^{25,26} A pressurized ion chamber was used to measure the total exposure from cosmic rays and terrestrial radionuclides. A portable $\text{Ge}(\text{Li})$ spectrometer was used to make *in-situ* measurements of the soil concentrations of radionuclides and of the individual external gamma-exposure rates from radionuclides. Fifteen to 19 locations within a 10-mile radius were surveyed at each power plant site, including most of the utilities' thermoluminescent dosimeter (TLD) stations.

The locations were laid out as close to the reactor's TLD sites as possible, taking into consideration the prospects of future disruptions for plowing, construction, or other major soil disturbances. The primary concern was to measure the concentration and variation of fallout fission products before startup.

The *in-situ* $\text{Ge}(\text{Li})$ detector results depended on soil density and depth distributions of radionuclides.

Fig. 9. Gamma-ray spectrum of a 200-g surface soil sample taken at site LLL-K at the Cooper Nuclear Station. The sample was analyzed with 300-mm x 300-mm NaI(Tl) detectors.

To measure these parameters, soil cores were taken at the corners and center of a 10-ft square centered at the detector. The cores were then sectioned as described above, and the section samples were dried, weighed, blended, composited, and counted on laboratory Ge(Li) detectors. Figure 9 shows a gamma-ray spectrum of a typical surface soil section taken at site LLL-K at Cooper Nuclear Station. The spectrum includes gamma rays from ^{214}Pb and ^{214}Bi daughters, from ^{40}K and from ^{137}Cs as the only fission product routinely found at each site, other observed man-made radionuclides were ^{92}Zr , ^{144}Ce , and ^{129}Sb . At all the sites, the natural radionuclides were uniformly distributed with depth.

Figure 10 shows soil profiles of ^{137}Cs for three permanent pasture sites (LLL-K, 52, and 33) at Cooper Nuclear Station. The solid lines are least-squares fits to an exponential function. The slopes are 0.33 cm^{-1} , 0.34 cm^{-1} , and 0.13 cm^{-1} , respectively. The concentrations are $148 \pm 9\text{ nCi/m}^2$, $150 \pm 4\text{ nCi/m}^2$, and $146 \pm 9\text{ nCi/m}^2$, respectively. Site LLL-K has been used as a pasture since 1952. For comparison, we show in Fig. 11 results from sites (Fort Calhoun TLD-C and Diablo Canyon TLD-13) where exponential distributions were not found. In the top distribution plot, the ^{137}Cs concentration appears constant down to 12.5 cm and assumes a sharp decrease below that depth. In the bottom plot the distribution is exponential down to 7.5 cm where it flattens out. These discontinuities may be due to some kind of disturbance or to different slopes in the five cores which were composited.

Table III compares the ^{137}Cs concentrations at selected locations around each power plant site. The locations were selected because the soil was undisturbed. Note that the measured concentrations and the concentrations calculated using the fitted exponential agree very well. The slopes are very similar, but the concentrations are significantly different from plant to plant. The Rancho Seco and Diablo Canyon sites show averages of 59 nCi/m^2 and 51 nCi/m^2 , respectively;

Fig. 10. Depth distributions of ^{137}Cs at three permanent pasture sites around Cooper Nuclear Station. For sites LLL-K, 52, and 33, the slopes are $0.33 \pm 0.01\text{ cm}^{-1}$, $0.34 \pm 0.01\text{ cm}^{-1}$, and $0.13 \pm 0.01\text{ cm}^{-1}$, respectively; the concentrations are $148 \pm 9\text{ nCi/m}^2$, $150 \pm 4\text{ nCi/m}^2$, and $146 \pm 9\text{ nCi/m}^2$. The solid lines are least-squares fits to an exponential function.

Fig. 11. Best distribution of TLDs at two sites where exponential distributions were not used.

Table III. TLD concentrations at preoperational power plants.

Power station	Site	Type ^a	Slope (cm ⁻¹)	Concentration (nCi/m ²)	
				Fitted	Measured
Dablo Canyon, California	2	Unc	0.15	45	45
	8	Unc	0.12	29	29
	14	Unc	0.16	16	37
Rancho Seco, California	10	PL	0.25	44	43
	11	PP	0.26	46	52
	18	PL	0.20	51	51
Fort St. Vrain, Colorado	AB	PP	0.22	91	101
	PCP	PL	0.14	94	95
	MBF	PL	0.11	91	90
Cooper Nuclear, Nebraska	31	PP	0.13	145	146
	KEN	PP	0.33	145	148
	52	PP	0.28	146	150
Fort Calhoun, Nebraska	A	Unc	0.11	151	152
	GIF	PL	0.20	120	125
	G	PP	0.18	128	133

^aUnc = uncultivated, PL = planted lawn, and PP = permanent pasture.

Table IV. TLD soil concentrations.

Power plant	No. sites	Mean concentration (nCi/m ²)	Range (nCi/m ²)
Fort Calhoun	16	140.67	12.4-150
Cooper Nuclear	17	137.22	61-170
Rancho Seco	18	146.61	126-158
Fort St. Vrain	17	90.94	43-101
Dablo Canyon	14	40.28	29-52

Table V. Average terrestrial radionuclide concentrations.

Power station	Site	Soil (nCi/g dry)	Grass (nCi/g dry)
Fort Calhoun, Nebraska	AB	1.0	1.0
Fort Calhoun, Nebraska	PCP	1.0	1.0
Fort Calhoun, Nebraska	MBF	1.0	1.0
Cooper Nuclear, Nebraska	31	1.0	1.0
Cooper Nuclear, Nebraska	KEN	1.0	1.0
Cooper Nuclear, Nebraska	52	1.0	1.0
Fort St. Vrain, Colorado	AB	1.0	1.0
Fort St. Vrain, Colorado	PCP	1.0	1.0
Fort St. Vrain, Colorado	MBF	1.0	1.0
Dablo Canyon, California	2	1.0	1.0
Dablo Canyon, California	8	1.0	1.0
Dablo Canyon, California	14	1.0	1.0

the Colorado sites average 90 nCi/g for the Fort Calhoun and Cooper sites average 139 nCi/g and 127 nCi/g, respectively. These results are summarized in Table V. Table V summarizes the natural radionuclide results for purposes of comparison.

These data are compared to the TLD measurements and to the TLD (soil) measurements in our published reports,^{1,2} and will be supplemented by similar (soil) TLD and soil sampling measurements at the same reactor sites after the plants have been operational for several years.

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