Analysis of Foods for Radioactivity*

by Naomi H. Harley

INTRODUCTION

The analysis of foods for radioactivity should not be considered as a primary defense against human intake. The first indication should always come from information on releases or from measurements of radioactivity in the airborne or waterborne releases. Once the existence of contamination has been established then the foods can be analyzed to evaluate potential hazard to man.

In contrast to most other pollutants the effects of radiation are considered to have a linear response regardless of the level, thus, there is no threshold and no absolutely safe limit. Instead it is necessary to set some lower level below which the radioactivity in foods is no longer of interest as compared with other sources of radiation or other hazards of life. The analytical significance of this is that the lower limits of detection for radioactive substances have been brought down to very low levels and the simple yes or no testing for acceptability that satisfies regulations for many other pollutants in foods cannot be used.

The radionuclides of interest in the case of contaminating events are almost all present now in foods in measurable quantities. Short-lived nuclides are the exception and the transuranic elements are only present at levels that require considerable effort in analysis. Since most of the radionuclides are already present, measurements made for background information should produce a numerical answer, not merely an indication that the amount is less than some pre-set value. The accumulation of background data provides a valuable baseline for evaluating excursions following a contamination event, The natural activity data are equally valuable since the amount of information on food concentrations is presently insufficient for valid comparisons with manmade radioactivity.

This report will describe the requirements and considerations for establishing a system to produce acceptably accurate measurements of radionuclides in foodstuffs. The basic concepts will be described, but to maintain the necessary brevity, the detailed procedures that might be used will be given only by reference.

During the preparation of this report, FDA has proposed certain recommendations for State and local agencies on Accidental Radioactive Contamination of Human Food and Animal Feeds. This material appeared in the Federal Register for December 15, 1978, page 58790, and is interesting background material for this topic.

CLASSIFICATION OF RADIONUCLIDES

The most useful classifications of a radionuclide are those based on the characteristics of the radiation emitted and by the identity of the chemical element. The former is both a guide to the nature of the hazard involved and to the measurement required. The chemical species (e.g., element, oxidation state) regulates the metabolic pathways in the biosphere as well as the nature of any chemical separations required in the measurement procedure.

The emitted radiations are generally grouped as alpha (α), beta (ß), and gamma (γ). Alpha radiation is characteristic of the natural and artificial radionuclides of high atomic weight and consists of energetic particles with very low penetrating power. Its hazard is significant only within the body, where alpha-emitting nuclides can irradiate specific sensitive tissues. Beta radiation appears in both heavy and light natural and manmade radionuclides, and consists of electrons possessing kinetic energy and having modest penetrating power. Gamma radiation is pure electromagnetic radiation and is extremely penetrating. Thus, it can be a hazard externally as well as when it is

*Excerpt from an OTA Working Paper entitled "Analysis of Foods for Radioactivity." A complete copy of the paper can be obtained from the National Technical Information Service. (See app. J.)

54-515 O - 79 - 15

present in the body. For the present purpose, we are only concerned that the penetrating nature of gamma radiation allows its direct measurement in foodstuffs, while alpha and beta emitters generally must be separated from the bulk constituents of the sample before measurement is possible.

There are other processes in radioactive disintegration that produce emissions. Alpha emission is usually accompanied by low-energy gamma rays that may be used for measurement. X-rays can be produced by electron capture and some gamma emitters decay by internal conversion, a process where a fraction of the gamma rays are converted to monoenergetic electrons. These processes do not really modify our measurement concepts but the decay modes of the significant radionuclides must be known for their accurate measurement.

The physical half-life of the radionuclide tends to control its persistence in the environment. For example iodine-l 31, with a half-life of about 8 days, is important for only a few weeks, while cesium-137, with a half-life of about 30 years, may be a problem for centuries.

A third classification that has some value is the source that produced the radionuclides. Knowledge of the source of radioactive contamination gives a good indication of the nuclides that are to be expected in the sample. This is of considerable assistance in planning the analysis since requesting a complete analysis for all radionuclides or even for all types of radioactivity in a single sample would lead to a lengthy and expensive operation. Many radionuclides are not potent health hazards, particularly those that are not metabolized by the body. The general groups of nuclides to be expected include the natural activities, specifically radioactive potassium and members of the uranium and thorium series, and the artificial fission products, transuranic elements, and other activation products that result from nuclear weapon explosions and nuclear reactor operations.

Fission products are a very complex mixture at the time of formation but the short-lived radionuclides die out rapidly and the mixture becomes simpler within a few days, The transuranics (plutonium, americium, etc., formed by activation of the basic fissionable material) are of some interest because of their high toxicity when incorporated into the body but present evidence indicates that their uptake through the gut is relatively small and that dietary intake is not a significant problem. This should be true particularly if the relative hazard of other radioactive contaminants probably present in the same sample is taken into account, The other activation products are frequently elements that make up steel or other metal containers or structural elements. Radioactive manganese, chromium, cobalt, zinc, and iron are particularly common and result from interactions of the materials with neutrons released in the nuclear reaction. It is worth pointing out that contamination of foodstuffs with single nuclides is extremely unlikely, and that more than one member of any group will probably be present in any sample.

DISTRIBUTION OF RADIONUCLIDES

The source of the radionuclides involved will generally control their distribution in the environment and their consequent transfer to the food chain. The sources considered here will include natural radioactivity, releases from operation of nuclear reactors and processing plants, and fallout from nuclear weapons tests.

Natural activity may be of concern when it is enhanced by man's intervention, say by mining to bring material to the surface and processing the ore to yield either products or wastes that may concentrate the radionuclides. Good examples are radium in uranium tailings, in phosphate rock waste, or in slags from phosphorus production, Radium may enter the food chain by dissolving in ground water and transferring through plant roots.

Nuclear reactors in normal operation release chiefly the radioactive noble gases that are not of interest in considering foods. Reactors do contain large inventories of fission products, transuranics, and other activation products, however, and accidental releases can contaminate vegetation by deposition or through the water pathway. Gaseous releases would most likely involve the volatile elements such as iodine and tritium or those with volatile precursors, such as strontium-90 and cesium-137, Aqueous releases would follow failure of the onsite ion exchange cleanup system and any water-soluble elements could be involved.

Processing plants could also have either gaseous or aqueous releases, but only fuel reprocessing is likely to be a significant contributor. In

this case, the fission products are aged before processing and iodine and the gaseous precursor radionuclides are not released. Tritium and carbon-14 are the major airborne products, while the waterborne radionuclides are the same as for reactors.

Atmospheric nuclear weapons tests distribute their fission products, transuranics, and other activation products globally, with local deposition being more or less, depending on the size of the weapon and the conditions of firing (high altitude, surface, underground).

In summary, the deposition of airborne material on vegetation or on soil is the route by which foodstuffs become contaminated and the subsequent behavior of the radionuclide is controlled by its chemical nature, including solubility and plant or animal metabolism.

CONTAMINATION OF FOODSTUFFS

Contamination of foods can occur either through atmospheric deposition or by transfer with water. In the first case it is possible for the radioactive material to be in the form of insoluble particulates rather than in a more available form where it will follow the chemistry of the elements involved. A knowledge of the pathway is not absolutely necessary but it does assist in deciding on the proper preliminary treatment of the sample of evaluating exposure. For example, surface contamination may have a different significance than the same material present in a plant through root uptake.

Since pathway information is not always available i t is generally considered proper to measure radioactivity in samples that have been prepared as if for eating, so as to approximate the true expected intake. This will usually result in stripping off or washing off of a considerable fraction of surface contamination. Cooking is generally not part of the preparation, as the mode of cooking and the use o f j u ices, cooking water, and the like cannot be predicted,

Milk is often recommended as an indicator food for studying radioactive contamination. It has many advantages:

1. It is available locally at most desired sampling locations.
2. It is marketed rapidly, so that short-lived radionuclides. such as iodine-l31, can be evaluated.
3. It is a major diet component in the United States, both directly and as an ingredient of prepared foods.
4. There is a lot of background information available on previous contaminating events.

It is worth noting that some of these "advantages' are the factors that contribute t o the role of milk as a source of human exposure.

Milk is a poor indicator of many contaminants. The natural activities, the transuranics. and the activation products have relatively low concentrations in milk. The first two are low because of poor biological transfer and the last because their pathways are almost entirely through the aquatic or marine food chain. Thus, the best approach is to know what is in the environment through other monitoring systems, and to design the food analysis program to fit the circumstances.

Other monitoring data are also necessary in fixing the geographical extent of a contaminating event. As a general rule, nuclear tests are global in radionuclide distribution, with enhanced levels near the test site. Releases from other nuclear operations tend to be more local in their effects and the food-monitoring plan can be modified to suit.

SAMPLING

The mechanics of obtaining representative food samples will not be considered here since the procedures are common to all types of food analysis. There are certain points that must be considered however. The first is whether the measurements are being made to determine human intake or the source of the contamination. In the latter case the early approach used by FDA(1) for radioactivity is appropriate. There, each sample was identified as to its place of origin. For evaluating intake i t is possible to collect total diet samples for a particular population group and to measure the radioactivity in this composite diet. This approach has been described by FDA(2) and by EPA(3) and is normally applied in institutions where mass feeding is carried out. A more elaborate procedure is to simulate a total diet by measuring a number of component food classes selected on the basis of

statistical information regarding consumption. This approach was originally used by the Atomic Energy Commission(4) and was applied originally to three major cities. This approach does allow identification of specific food types that are contaminated but requires much greater effort and cost in analysis.

The use of indicator foods as an intermediate type of monitoring is widespread. As mentioned previously, most of the systems depend on the sampling of milk which is available in most parts of the country either with specific information as to place of origin or the general area of the milkshed. Part of the reason for using milk is the ease in sampling but it also has significance as a primary food for the youngest and most susceptible population group and it also does tend to pick up several of the fission product nuclides of dietary significance such as radiostrontium, radiocesium, and radioiodine. Most other foods give limited geographical or seasonal coverage and are less satisfactory.

The preservation of samples in the field during transport and in the laboratory awaiting analysis is only an esthetic matter in the case of radionuelides. Decomposition processes do not change the radioactivity, and sample contamination by radioactivity is unlikely. Thus, freezing, formaldehyde addition, or any other method that will maintain the sample is adequate. Any additive should be checked to assure it does not contain significant amounts of the radionuclide sought.

For the purpose of this report we will assume that an adequate quantity of a representative sample is available for analysis and that another portion is available for storage, either permanently or until the analytical results are accepted as satisfactory.

SAMPLE PREPARATION

The measurement of radioactivity is a physical process and it is most efficient when the radioactivity from a relatively large sample can be placed close to the detector. This means that direct measurements of bulk samples are only useful at relatively high levels of contamination and that most measurements are preceded by preparation and possibly chemical separation to reduce the bulk of the material and to improve the efficiency of the measurement.

As mentioned previously, sample preparation may include removal of inedible portions of the foods or those portions not generally eaten. For example, citrus rinds, apple cores, outer leaves of leafy vegetables, and aboveground portions of root vegetables would normally be discarded. The general goal is to prepare the foods as if for cooking or consumption.

Foods generally have a high water content and a primary method of bulk reduction is drying at room temperature, at elevated temperatures, or freeze drying, Most of the radionuclides of interest are not volatile under these conditions and losses must be considered only for elements such as tritium and iodine. The dried material can be reduced further by ashing at elevated temperatures, by cold ashing with activated oxygen, or by wet ashing with oxidizing acids. The sample dry-ashing process is most likely to lead to loss of volatile elements but with care even cesium, polonium, and lead can be retained, The other processes should not lead to losses of elements of interest with the exception of iodine and tritium, mentioned above, and of carbon.

Another approach that is essentially one of reducing bulk is to extract either the original sample or the dried or ashed material with acids or other solvents and thus remove the desired elements from the bulk of the sample. This requires considerable testing beforehand to be certain that the process operates in the desired manner.

All of these procedures reduce the bulk of the sample and in the case of extraction may also separate the desired constituents from some of the remaining inert material. They do not, however, separate the radionuclides of interest completely from the other radionuclides present in the sample. Such separations will be covered in the next section, but they may not be necessary if the measurement technique can provide both qualitative and quantitative information. In most cases this limits the possibilities to gamma spectrometry on the prepared sample.

If the samples are going to be subjected to chemical analysis, the sample preparation must include the dissolution of the dried or ashed material. This has already been done, of course, in preparing the wet ashed or extracted solutions. The radionuclides of interest in ashed foods should be soluble in strong acids if ashing temperatures have not been excessive, and this treatment is usually accepted. If there is concern that

insoluble particulate may be present, it is necessary to use more drastic methods such as fusion to bring the sample into solution. This should not be necessary, however, if human hazard is the problem under consideration.

At the time of preparation, if not before, the basis of measurement must be established. Depending on the use of the data, wet weight, dry weight, ash weight, volume, or even numerical count (e. g., eggs) has to be determined. Frequently, this may have to be done in the field, but fortunately, relatively crude measures are adequate, Since unforeseen questions often arise, it is recommended that as many of these quantities be measured as is possible.

The requirements for measurement of radionuclides in foods are such that sample preparation is best handled by the group responsible for the rest of the analysis. The chief difference from other types of analytical work is the initial sample. The usual range is from 1 to 20 kg, and the reduction of this amount of material is a specialized problem.

RADIOCHEMICAL SEPARATIONS

Radiochemical separations are required to isolate the desired radionuclide both from the remaining bulk constituents and from other radionuclides which would interfere in the measurement. In addition, it is necessary to convert the final product to a form suitable for presentation to the counter. This may involve elect redeposition, precipitation, or other processes. There are a number of manuals giving the details of specific radiochemical procedures and these details will not be repeated here. There are a few generalities however that may be of interest.

The actual mass of radionuclide that is measured is almost always vanishingly small. This means that many of the normal chemical reactions used in analytical chemistry will not take place; for instance precipitates will not form. For this reason it is common to add a few milligrams of carrier material which is preferably the inert form of the same element. Where this does not exist it is frequently possible to use similar elements as carrier, such as the substitution of barium for radium. The inert form then follows normal chemistry, carrying the radionuclide with it. It is also worth noting that even when the separation technique does not depend on the mass of element present, a carrier may still be useful in preventing unwanted coprecipitation or absorption on glassware.

Because of the high degree of purification required in radiochemistry, it has been customary to make a number of repeated separations either identical or different to insure purity. To carry these out in a reasonable time it is better to lose a small amount of the nuclide sought while removing a large fraction of the undesired material rather than retaining all of the nuclide and much of the undesired material. Fortunately, it is possible to measure these losses in analysis and to make a correction at the end. One approach is to measure the carrier at the end of the analysis either by gravimetric or instrumental analysis. If there was none of the carrier substance present in the initial sample, the fraction recovered will be equal to the fraction of desired radionuclide recovered. Where carriers are not available or where they are present in variable amounts in the original sample it is frequently possible to use radioactive tracers, that is radioactive isotopes of the same element or a similar element. The fraction of the amount added that is left at the end of the analysis can be used to determine the recovery of the radionuclide sought.

In the chemical separations there is considerable use of classical analytical chemistry based on precipitation and in most cases it will at least appear as *a* final collection step to put the desired radionuclide in a condition suitable for counting. In addition the general techniques of ion exchange, liquid extraction, distillation, and electrolysis are used.

Since many of the separations in radiochemical analysis are carried out in small volumes the centrifuge is widely applied, It has become common practice to redissolve and reprecipitate rather than to wash precipitates carefully. This may be repeated several times very rapidly and will generally give good decontamination (separation from other radionuclides). As noted above, where the element sought and the contaminant have similar properties it is preferable to use two different precipitations, a precipitation followed by an extraction, or any two widely different steps to achieve good decontamination.

One process that is frequently of value is called scavenging. This term is usually applied to a pre-

cipitation carried out to remove contaminating radionuclides after the bulk matrix of the sample has been removed. The process involves the addition of a group carrier, precipitation, and discarding of the precipitate. A typical example would be an addition of iron carrier followed by hydroxide precipitation to remove rare earths and other heavy metals in a determination of an alkali or alkaline earth. Frequently the scavenging procedure is repeated to improve decontamination, but it is only rarely that the scavenge precipitate is redissolved and reprecipitated to recover any of the desired constituent that may have been absorbed.

After suitable radiochemical separations have been made, it is necessary to collect and mount the sample for counting. This is most often done by precipitation or by elect redeposition. In the section on measurement, the requirements for sample preparation are discussed: for example, the sample area and mass should be reproducible and the sample should be mounted on a sample holder identical with that used for the counter standard. Since it is frequently necessary to determine the weight of precipitate for recovery determination, this factor must also be considered.

The selection of a mounting technique is usually a compromise between convenience and the counting requirements. Besides consideration of the type and energy of emission, practical matters such as the counter size and sample mounts available must be weighed,

In certain cases, the total amount of sample available may be limited and analysis for several radionuclides may be required. Procedures should be on hand for the sequential analysis of single samples, even though separate samples are used for routine work.

MEASUREMENT

The method of measurement to be selected depends on the type of radiation, the form of the sample, and to some extent on the amount of radioactivity. It is necessary that the complete analytical procedure be designed so that the sample is brought to a suitable form for the equipment and conditions that exist.

It is possible to measure the total gamma, total beta, or even the total alpha activity on a sample of food, Unfortunately, such data are valueless in estimating human exposure. The accuracy of the determination is very poor, natural potassium usually interferes, and the chemical and radiation characteristics needed to evaluate possible hazard are not known. It is possible, however, to set a particular total activity level as a screening level for a specific food. If the measured value is below the screening level, no analyses for individual radionuclides are performed. In such a case, the measurements should be considered as internal data only and the numerical results should not be published. Any report should merely list the samples as having activities below the stated screening level.

Qualitative identification of radionuclides on original samples of foods is limited to gamma emitters at relatively high levels. The sensitivity can be increased if some bulk reduction, as described under “Sample Preparation, ” is carried out. The identification of alpha and beta emitters depends on radiochemical separation for element identification and the measurement of energy or half-life on the separated material for radionuclide identification. The latter step may be omitted if other considerations limit the possibility to a single radionuclide.

The equipment available for quantitative measurement of radioactivity is sufficiently sensitive for all foreseeable cases. Instruments for detection of the three major emissions are described here, and their applications are shown in table 20 (chapter VIII) in terms of the detection limits for various radionuclides.

It is worth pointing out that a number of the instruments described are not commercially available. They have been in the past, but the low demand has removed them from the market, The necessary electronic components are available but the mechanical assemblies for the detectors are not, and the larger laboratories tend to build their own systems.

Alpha Emitters. The measurement of alpha activity is best carried out on a very thin sample to avoid self-absorption of the alpha particles. This is even more true for spectrometry since degradation of the original alpha energy will give a spectrum with poor resolution. The measurement of the total alpha activity can be carried out either in thin-window counters or by scintillation counting with zinc sulfide phosphor. Both techniques have high efficiency but the scintillation method can give a considerably lower background with a consequently lower limit of detection. Unfortunately, neither system is readily available as a

complete unit and must be assembled from commercially available components. A somewhat less sensitive technique is to use the liquid scintillation spectrometer described in the next section.

Alpha spectroscopy also has two possibilities, the Frisch grid ionization chamber or the silicon diode solid-state detector. The Frisch grid unit can handle large area samples but is slightly poorer in resolving closely separated energies. On the other hand, the silicon diodes available are all quite small and can only count samples of about 1-cm diameter with high efficiency. This is not a serious limitation, since the Frisch grid systems are not presently available in this country. This means that the form of the sample presented to the counter is limited. Most metals are electrodeposited onto small smooth discs of stainless steel, nickel, or platinum, with evaporation of pure solutions being used in some cases.

A specialized technique is available for measuring radium-226, by means of its gaseous daughter product radon-222. If the sample can be put into solution and stored for about 3 weeks the radon-222 will be close to equilibrium (equal activity) with the radium-226. The gaseous radon can then be transferred to a scintillation counter or an ionization chamber for measurement of its alpha activity. This is rather specialized, and the equipment is not listed in this report since radium-226 can be determined by other procedures. For example, separation and counting as the sulfate or chromate using barium as carrier is quite adequate.

Beta Emitters. Beta counting is a little more flexible in the mass of material that can be present at the time of counting. This is true for higher energy beta emitters but carbon-14 and tritium present a problem. Since each individual beta emitter gives off particles with a range of energies from zero to a characteristic maximum, beta spectrometry is not possible for food samples. Some qualitative information may be obtained from absorption measurements, which allow an estimate of the characteristic maximum energy of single emitters or simple mixtures (5).

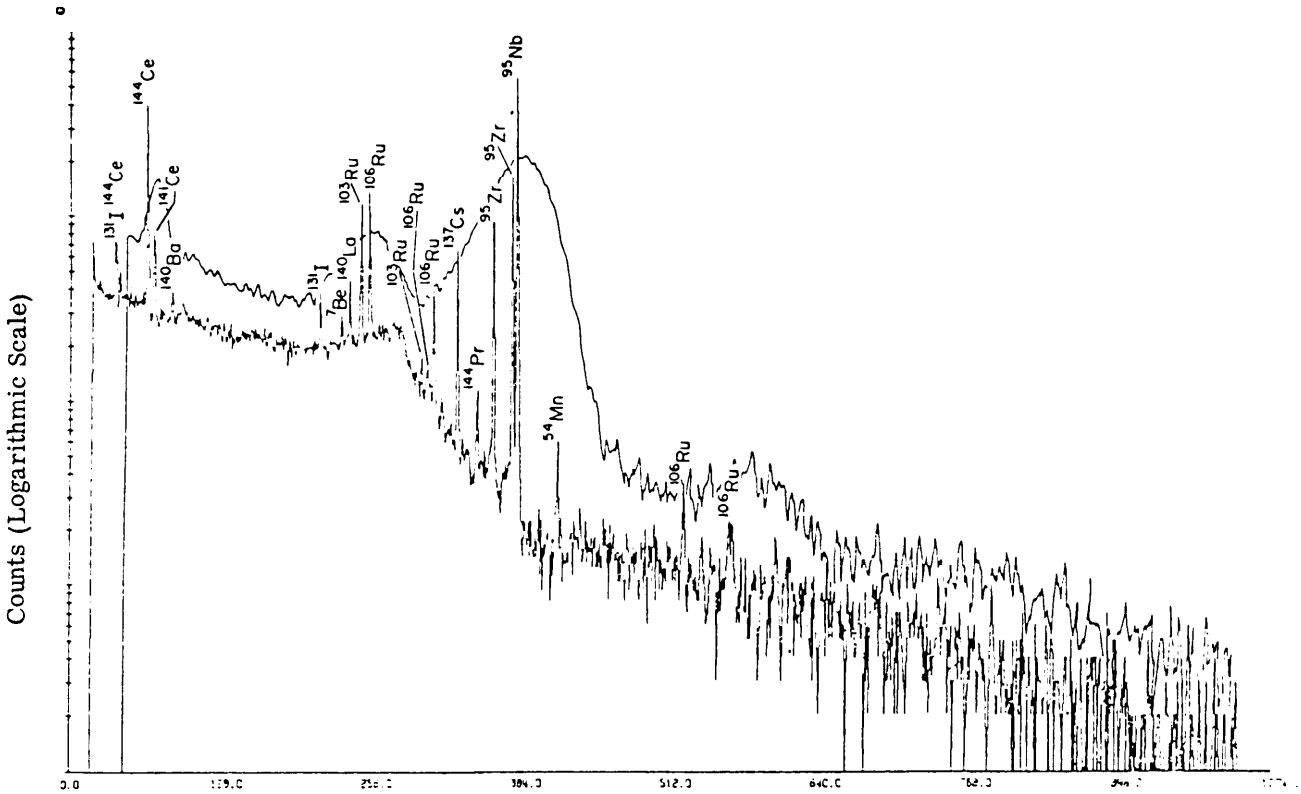
The available counting equipment for quantitative measurement includes geiger counters, proportional counters, and scintillation counters. The geiger counter is relatively inexpensive and requires only simple electronics but is not popular and is generally not available as a counting system. The thin-window proportional counter is used widely and has both reasonably high efficiency and low background. For low-level samples the background can be further reduced by anticoincidence techniques. These add to the complexity and cost of the system but are sometimes necessary.

Scintillation counting can be performed in two ways. Solid scintillators can be used for counting chemical precipitates collected on filter papers and liquid scintillators can be used whenever the sample can be made miscible with the scintillating solution itself. This can even be done with solids by suspending them in a scintillating gel. The advantage of liquid scintillators is their high efficiency, even for the low-energy emitters carbon-14 and tritium. Scintillation systems for counting precipitates are not commercially available at present. There are, however, many liquid scintillation systems on the market, most of them with automatic sample changers. They enjoy a good demand in medical and biological studies with tritium and radiocarbon. These use high levels of activity and short counting time. The better systems also have a provision for rather crude spectrometry. They can distinguish qualitatively and quantitatively among carbon-14, tritium, alpha emitters, and higher energy beta emitters.

Gamma Emitters. Gamma rays are so penetrating that the detector must have a considerable mass to absorb enough to produce a response. For spectrometry, complete energy absorption is required, so solid detectors are most useful. Sodium iodide is a popular detector, since crystals can be fabricated in large sizes and the crystal is transparent to the scintillations produced by radiation. Sodium iodide is used for most of the gamma counting today, and is still useful in spectrometry. It has high efficiency but poor energy resolution and is now applied to samples where some separation has taken place. Milk is a good natural example, as the metabolism of the cow removes most gamma emitters other than isotopes of cesium and iodine.

More complex spectra can be resolved with the solid-state germanium diode detector. A comparison of a sodium iodide with a germanium diode detector spectrum is shown in figure I-1. It is possible to resolve the sodium iodide spectrum of a sample containing several radionuclides but this requires a sizable computer and very careful work to allow for drift in instrument characteristics and for the possibility of other radionuclides than those sought being present. The diode detector gives much better resolution and present spectrometer systems frequently include sufficient computer capacity to resolve the resulting spectra and produce quantitative data. At the same time, interferences from radionuclide im-

Figure I-1 .—Comparison of Gamma Spectra Taken With a Sodium Iodide Detector (Upper Curve) and a Germanium Diode Detector (Lower Curve). Note that the main single peak in the sodium iodide spectrum is resolved into 9 peaks for 6 radionuclides with the germanium diode spectrum “



purities are greatly reduced compared to spectra from sodium iodide detectors. The efficiency of the diode is low and, for many analyses, a spectrometer can only be used for one measurement a day. Another disadvantage is that the detector must be kept at liquid nitrogen temperature to maintain its detection capability. Newer diodes have been developed that do not require storage at liquid nitrogen temperatures; however, they must be cooled during measurement. Since the system is in use most of the time, this may not be a significant advantage.

Diode spectrometers may also be used to measure the low-energy gamma-rays that accompany alpha emission. This allows direct measurement in some environmental samples, but the levels in foods have not been high enough for this technique.

General Requirements. The choice of a counting procedure depends on the precision required. In turn, the relative precision of a quantitative counting measurement is inversely proportional to the square root of the number of counts obtained. Thus, any improvement in precision must be obtained by increasing the number of counts. This can be done by using larger samples, by counting for longer times, or by using counters with higher efficiency. A secondary improvement is possible for low-activity samples by decreasing the background. Each of these improvements has some drawback, and selection of the optimum balance requires a degree of experience to weigh

cost, manpower, and quality. For example, handling larger samples increases the effort in sample preparation and radiochemistry while longer counting times require more counters, and increased counter efficiency or lower background is expensive.

Counting-room operation requires the maintenance of detailed records on standardization, background, and sample measurement. This information can frequently allow the recovery of bad data or the correction of calculational errors. In addition, maintenance of control charts(5) will signal when instrument problems arise.

Experience with modern nuclear instrumentation has been good, and downtime of the order of 5 percent is common. This requires that service be available immediately or the next day following a breakdown. The increasing complexity of measuring systems tends to preclude in-house servicing in most cases, but some diagnostic capability and competence in minor repairs is very valuable.

CONTROLS

An analytical laboratory carrying out measurements of radioactivity in foodstuffs will require a program to document the validity of the measurements, This is necessary in legal cases and is also highly desirable when data are being presented for use in decisionmaking. A suitable program of quality control should be carried on in addition to the necessary calibrations and standardizations.

Calibration is the determination of the relationship between a desired quantity and the response of a particular instrument. In the case of radioactive materials this may mean that the instrument must be calibrated with each radionuclide to be measured unless there is evidence that the instrument response is independent of the energy or other characteristics of the radiation. Alternatively a complete response v. energy calibration may be substituted. These calibrations, to have a legal standing, should probably be traceable to the U.S. National Bureau of Standards or comparable authority. This turns out to be a requirement that is far from trivial in the effort required.

Fortunately, a complete calibration is not required at frequent intervals and, depending on experience, may not be needed oftener than every few months. In the meantime of course it is necessary to have assurance of the proper operation of measuring equipment but this can be done with simple standards or even with samples that are reproducible over a period of time. For example, a simple counter standard may be run every morning before starting operations just to be sure that the counter is working properly. Similar standards should also be available for checking spectrometer energy response.

An ideal quality control program should include the checking of the complete procedure from sample preparation through measurement. This requires that standard samples be available for testing the full procedure. Additional controls would include running of blind duplicate samples to test reproducibility of analyses, and blank samples to check on the possibility of laboratory or reagent contamination. These three types of samples should make up at least 10 percent of the laboratory output if the quality of measurement is to be followed closely. It is also necessary that the data be published with the ordinary laboratory results and that any deviations from the expected results should be used to make suitable corrections in laboratory operations.

There is always some danger that a laboratory may drift out of control in spite of an adequate internal quality control program, This is possible since an internal program may emphasize consistency while the absolute values drift. Therefore, some part of the quality control effort must be devoted to intercomparisons with established groups, such as IAEA or EPA. This should be done on an annual basis, at least.

It must be noted that standard samples are frequently not available, and recourse must be had to "spiking," which is the addition of a known amount of a radionuclide to a sample that is relatively free of that nuclide. Recovery of the spike is not necessarily a good test of a chemical procedure, since a natural sample may be more difficult to analyze.

A final component of a good quality control system is a careful, responsible review of all the data produced. This means checking the arithmetic, knowing the characteristics of the equipment and, hopefully, having a sixth sense that recognizes that certain results do not look right. This is especially true when relying on automatic counters or on computer processing of the data.

STAFF AND FACILITIES

A minimum facility could be designed around a staff of six, including a B.S. or M.S. senior chemist with experience, a B.S. junior chemist and three technicians for chemistry, sample preparation, and counting-maintenance, A- secretary-administrative assistant could handle reports, local purchasing, and similar duties. With this small staff, considerable versatility and flexibility would be required.

The measurement of radioactivity in foods requires fairly extensive facilities to handle the varied analyses that might be called for, In addition to a modest office space, separate rooms would be necessary for sample preparation, wet chemistry, measuring equipment, and for maintenance support and storage. The first two laboratory rooms would require hoods, chemical benches, and laboratory safety devices such as showers and eye fountains. Since the samples to be measured are normal foods there are no special requirements for radiation safety, If the samples were radioactive enough to be a personnel hazard in the laboratory they certainly would not require this type of precise measurement.

If the laboratory is to be in continuous operation it is most likely that the principal chemical and measurement systems will have to be available at least in duplicate. This and similar considerations lead to the conclusion that a certain minimum size and sample throughput are not necessary if the radioactivity laboratory is part of a larger operation which can furnish support. One continuing problem is the need for electronic maintenance of radiation measuring equipment. This is rather specialized and must be considered either when staffing the laboratory or in locating it where such services are readily available.

It is not always possible to purchase the ideal counting equipment for a particular purpose. The total demand for many systems is small and commercial instrument makers are not interested. Large laboratories can make some of their own equipment but this is not possible for the group described here. The construction of alpha and beta radiation detectors requires services of a first-class machinist plus sufficient electronic know-how to transfer the detector signal to the available commercial equipment. Where such capability already exists in the overall organization, it may be fruitful to copy advanced noncommercial instruments, but staffing specifically for this function is probably not economical. Thus, most small laboratories must operate with the less-than-optimum commercial equipment,

If significant uses are to be made of gamma spectrometry there will be a need for at least modest computing facilities. These can be part of the purchased spectrometer system but this is an expensive method if suitable computer time is available otherwise. The former approach is used here, and it might be noted that the spectrometer computer has capacity for other work.

Additional space is required not only for stocking necessary reagents and materials but also for storage of incoming samples and for storage of residual material from samples that have been run. As a general rule it is desirable to take a larger sample than would be required and to set aside a portion of this for possible contingencies or if the data are later brought into question.

The output of a group this size should run between 1,000 and 4,000 samples[1] a year, depending on the difficulty and the activity level, One limiting factor is that, at background levels, each counter can only turn out one or two samples per day. Following a contaminating event, it should be possible to process smaller samples and to count for shorter times, both of which would allow higher output. It might be possible to add 3 more laboratory staff plus 50 percent more space and equipment dollars and essentially double the output. Further increases would probably run into other bottlenecks due to the need for added support.

Table I-1 details the costs for space modification and furnishings ($140,000), for equipment ($200,000), and for supplies ($20,000). These costs are based on a building in place, lights and partitions in place, and utility stubs in each room, It is also assumed that building services are provided.

[1] A sample is defined as a complete gamma spectral analysis or a single radionuclide analysis that requires preparation plus radiochemistry.

Table I-1. —Costs for Space, Furnishings, Equipment, and Supplies

	Cost
Space modification and furnishings	
Chemical ***laboratory—20 x 30 ft.***	
Base cabinets	$ 8,500
Wall cabinets	2,500
Storage cabinets	1,800
Sinks (2)	1,200
Hoods (3)	7,000
Services and Installation	40,000
Preparation area— 10x 30 ft.	
Base Cabinets	3,800
Wall cabinets	1,800
Storage cabinets	1,500
Sink	600
Hood	2,500
Benches	600
Services and Installation	25,000
Counting **area— 10x 20 ft.** *(air-conditioned)*	
Benches	1,200
Rack	1,000
Base cabinets	1,700
Wall cabinets	600
Desk, etc.	500
Services and installation	12,000
Storage ***area— 10x 20*** ft.	
Acid storage	800
Solvent storage	600
Cabinets	3,000
Services and Installation	1,500
Office area —library 10x 20 ft.	
Desks, etc (4)	2,000
Cabinets, bookcases	3,000
Services and Installation	1,000
Equipment	
Chemistry area	
Hot plates (2)	600
Stirring plates (6)	900
pH meter	350
Demineralizer	500
Centrifuges (3)	1,800
Platinum ware(12 items)	12,000
Preparation area	
Large furnace	3,500
Small furnaces (2)	1,700
Large oven	1,400
Small oven	600
Vacuum oven	1,000
Still and tank	2,100
Blender	500
Analytical balance	2,800
Scales (2)	1,400
Canning unit	500
Bulk freeze drier[a]	14,000
Hot plates (2)	600
Counting area	
4-position alpha spectrometer and detectors multichannel analyzer and output	21,000
Germanium diode gamma spectrometer with detector, shield, electronics, PDP-11 computer and output	100,000[b]
4 general-purpose proportional counters	10,000
Liquid scintillation spectrometer automatic sample changer with alpha-counting capability	18,000
Maintenance-oscilloscope, signal generator, digital voltmeter	4,500
Storage area	
Refrigerators (2)	800
Freezers (3)	1,500
General	
Safety equipment	600

Supplies

A rough estimate would be $20,000 for the initial inventory of reagents, glassware, and other supplies. The annual cost would be about $3,500 per person in the lab, or $17,500.

Total costs

Setting up the laboratory would cost on the order of $350,000. Operation should run about $100,000 for salaries, $17,500 for supplies, and about $50,000 per year for equipment maintenance and replacement.

[a]Desirable, but not absolutely essential.
[b]Additional detectors—up to 4—could be added for about $30,000 each.

APPENDIX I REFERENCES

1. E. P. Laug, et al. 'radioactivity in Foods,'" *Journal AOAC* 44, 52.3-30 (1961), also "Strontium-90 Analyses of Human and Animal Foods, *Radiological Health Data* 1, 36-40 (May 1960).
2. Teen-Ager Diet Survey, Food and Drug Administration, *Radiological Health Data* 4, 18-22 (1963)
3. R. E. Simpson, E. J. Baratta, and C. F. Jelinek, "Radionuclides in Foods: Monitoring Program. " *Radiation Data and Reports* 15, 647-56 (1974). (This is a later report that summarizes much of the EPA program.)
4. J. H. Harley and Joseph Rivera, *Summary* **of** *Available Data on the Strontium-90 Content of Foods and Total* Diets in the *United States*, U. S. AEC Report HASL-90 (August 1960).
5. J. H. Harley (Editor), *EML Procedures Manual*, U.S. DOE Report HASL-300 (1972, revised annually).

GENERAL REFERENCES

Radiochemical Methods

National Academy of Sciences, Nuclear Science Series (NAS-NS 3001 ff), Office of Technical Services, Washington (1960 and later), A Series of Monographs on Individual Elements and Techniques.

WHO-FAO-IAEA, Methods of Radiochemical Analysis, World Health Organization, Geneva (1966).

Johns (Editor), National Environmental Research Center, Las Vegas, Handbook of Radiochemical Analytical Methods, EPA-680/4-75-001, February 1975.

Harley (Editor), Environmental Measurements Laboratory, DOE, EML Procedures Manual, HASL-300, 1972 (Updated Annually).

International Atomic Energy Agency, Reference Methods for Marine Radioactivity Studies (Sr, Cs, Ce, Co, Zn), Technical Reports Series No. 118 (1970).

Ibid. (Transuranics, Ru, Zr, Ag, I)

Technical Reports Series No. 169 (1975)

Measurement of Radioactivity

National Council on Radiation Protection and Measurements, A Manual of Radioactivity Measurements Procedures, Report 58 (1977).

National Council on Radiation Protection and Measurements, Environmental Radiation Measurements, Report 50 (1976).

International Commission on Radiation Units and Measurements, Measurement of Low-Level Radioactivity, Report 22 (1972).

International Atomic Energy Agency, Measurement of Short-Range Radiations, Technical Reports Series No. 150 (1973).

International Atomic Energy Agency, Metrology and Radionuclides (Symposium), IAEA, Vienna (1960).

Crouthamel (Editor), Revised by Adams and Dams, Applied Gamma-Ray Spectrometry, Pergamon, New York (1970).

Data Compilations

Lederer and Shirley, Table of Isotopes—7th Edition, John Wiley, New York (1978).

Health, Scintillation Spectrometry, Gamma-Ray Spectrum Catolog (3rd Edition), USAEC Report ANCR-1000 (1974).

Chanda and Deal, Catolog of Semiconductor Alpha-Particle Spectra, USAEC Report IN-1261, March 1970,

Niday and Gunnink, Computerized Quantitative Analysis of Gamma-Ray Spectrometry, USAEC Report UCRL-51061, 4 Volumes (Includes Decay Scheme Data Library).

Quality Assurance

Kanipe, Handbook for Analytical Quality Control in Radioanalytical Laboratories, EPA Report 600/7-77-088 (April 1977),

Ziegler and Hunt, Quality Control for Environmental Measurements using Gamma-Ray Spectrometry, EPA Report 600/7-77-144 (December 1977).

See also:

Metrology Needs in the Measurement of Environmental Radioactivity Environment International 1, 7ff (1978),

GLOSSARY

Activation Products—The radionuclides formed in either fissionable material or in surrounding material during a nuclear reaction, usually by capture of neutrons,

Carrier—A stable element added in radiochemical analysis to provide sufficient mass that a radionuclide can follow various separation steps.

Detector—Any device that transforms the radiation emitted by a radionuclide into a signal that can be handled electronically. The most usual signal is an electrical pulse which can be recorded.

Disintegration-The spontaneous process by which a radionuclide gives off energy in the form of nuclear radiation.

Fissionable Material—Any of the heavy radionuclides which can undergo fission, or splitting into two or more lighter atoms. The fission process is accompanied by a large release of energy.

Fission Products—The radionuclides formed when fissionable material splits into two or more atoms.

Geiger Counter—A detector based on gas ionization which converts any ionization within the detector into a large electrical pulse.

Germanium Diode Detector—A gamma-ray detector composed of very pure germanium, usually activated with lithium.

Indicator Food—A food that is measured to give an estimate of the total dietary intake of man.

Ionization Chamber—A radiation detector based on gas ionization which converts any ionization within the chamber into an equivalent electrical pulse.

Pathway—The route by which a radionuclide is transferred from the source through the environment to man.

Picocurie—See units of radioactivity. Equal to 2.2 disintegrations per minute.

Proportional Counter—A detector based on gas ionization which converts any ionization within the detector into an amplified electrical pulse proportional to the amount of ionization.

Radionuclide—Any atomic species which is unstable and gives off nuclear radiation to attain stability.

Scintillation Detector—A detector which converts nuclear radiation to a pulse of light. This, in turn, can be converted to an electrical pulse with a phototube.

Sodium Iodide Detector—A scintillation gamma-ray detector made up of a single crystal of sodium iodide activated with silver.

Spectrometry—The measurement of the energy of radiation emitted during decay of a radionuclide or mixture of radionuclides. Quantitative as well as qualitative information may usually be derived.

Tracer—A radionuclide added in radiochemical analysis to follow the distribution of the desired constituent in various separation steps.

Transuranic Element—An artificial element having a higher atomic number than uranium, formed by activation, usually with neutrons.

Units of Radioactivity—There are various ways of expressing the disintegration rate of a radionuclide. In this report the disintegrations per minute (dpm) unit is used. In other reports, the curie and its submultiple (millicurie, microcurie, picocurie) are used and the new International Standard nomenclature is the bequerel, equal to one disintegration per second.