ORTEC®

Food Monitoring Note
Nuclear Spectroscopy Systems

Radioactivity and its Measurements in Foodstuffs

Where Does Radioactivity in Food Come From?

Radioactivity is, and always has been, present in all foodstuffs to some extent. Recent events have made the general public aware of this and most do not know about radioactivity and radiation.

There are four possible sources of radioactivity in foodstuffs: one natural and three artificial.

1. **Natural Radioactivity:** Potassium is an essential constituent of all cellular tissue, one isotope of which, potassium-40 (^{40}K), is naturally radioactive. Most foodstuffs and water also contain small quantities of uranium, thorium, and their daughter products Thus, some irradiation of the body following food intake or drinking is inevitable. Since homeland security became of concern and radiation monitors have been installed to monitor people and freight, the extent to which many objects are naturally radioactive has become very apparent through so-called “innocent alarms” occurring at border crossings and freight terminals.
2. **Industrial Radioactivity:** Effluent and discharges from nuclear power, fuel fabrication and reprocessing plants, fossil fuel power plants, laboratories, and even hospitals contribute small quantities of radioactivity to the environment which can, in principle, contaminate food. Many industries, most notably the oil industry, make use of radioactive sources. These are generally highly controlled and sealed and so are unlikely to contribute to radioactivity in food.
3. **Weapons Testing:** Airborne debris from the atmospheric weapons tests – particularly in the 1960s – caused relatively major contamination of crops and other foodstuffs. Even today some residual activity from these tests can be measured in soil and plants. The cumulative activity from these tests was much greater than that, for example, released from Chernobyl.
4. **Accidents Involving Radioactivity:** Two examples are the Windscale (UK) accident of 1957 when an accidental release caused much local contamination of herbage and milk, and the Chernobyl reactor fire, in 1986, which resulted in widespread, but variable, contamination of foodstuffs in Europe and South West Asia. Unfortunately a third must now be added to this list, the accident at Fukushima Daiichi following the 2011 earthquake in Japan

Note that the sterilization of foodstuffs by irradiation induces no detectable activity in food.

How Does Artificial Radioactivity Get Into Food?

Airborne radioactivity mainly consists of nuclides attached to dust particles which settle directly onto the ground or can be washed out by rain. Some radionuclides are gases and these are mainly washed out by rain. Thus, both vegetation and soil can be contaminated. Deposited material may become quite strongly attached to leaves and possibly transferred throughout the plant by foliar uptake. In the longer term, activity can be transferred to herbage via root uptake from the soil. Plants will naturally take up trace elements – particularly if they are metabolically essential – and their radioactive isotopes, if present and chemically available, will naturally follow. In addition, contaminated soil may adhere to the plant. Different plants and mushrooms concentrate different nuclides because of their varying chemistry.

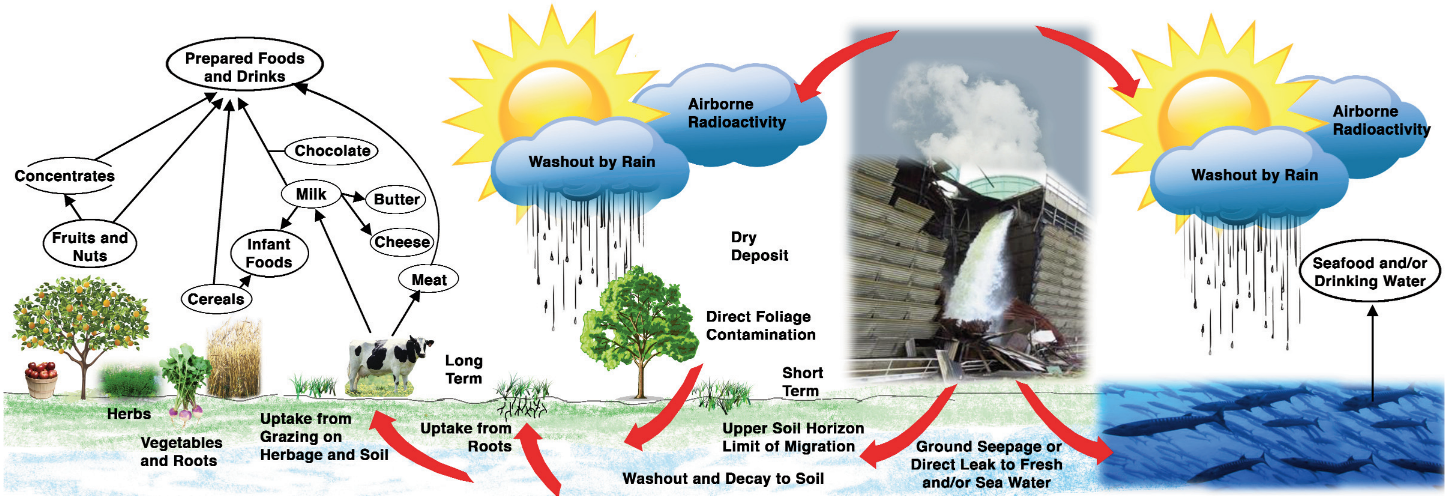
Vegetation, either as fresh food or silage, transfers activity to animals. The subsequent concentration of radioactivity in animal meat depends particularly on the area grazed. Lactating animals produce radioactive milk. There may be some concentration as the material goes along the food chain (biomagnification).

Milk products (e.g., dried milk, whey) will contain higher concentrations of activity simply because of moisture removal. There are numerous foodstuffs sold containing mixtures of milk powders and cereals.

Any foodstuffs which are are processed by removal of water (e.g., fruit juices) will naturally concentrate the activity also.

In the case of a reactor accident, where radioactive material is released from a reactor containment into the surrounding earth, as well as this airborne path, contamination will be present in the soil and groundwater near the plant. In a coastal facility, contamination might leak into the ocean contaminating fish and other organisms generally considered by humans to be seafood.

Figure 1 provides a graphic representation of the primary mechanisms by which radioactivity enters the food chain.



What Nuclides are Found in Food and at What Level?

The most important nuclides to be assessed following the release of radionuclides from a uranium-fuelled reactor to the environment are: ^{134}Cs, ^{137}Cs, ^{131}I, and other gamma emitters, the beta emitters: ^{89}Sr, ^{90}Sr, and tritium, and the alpha emitters: ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, and ^{242}Cm. In general, the radionuclides of major importance in the contamination of food and environmental samples (which are part of the pathway to food) are (Ref. 1):

Air	^{131}I, ^{134}Cs, ^{137}Cs
Water	^{3}H, ^{89}Sr, ^{90}Sr, ^{131}I, ^{134}Cs, ^{137}Cs
Milk	^{89}Sr, ^{90}Sr, ^{131}I, ^{134}Cs, ^{137}Cs
Meat	^{134}Cs, ^{137}Cs
Other Foods	^{89}Sr, ^{90}Sr, ^{134}Cs, ^{137}Cs
Vegetation	^{89}Sr, ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{131}I, ^{134}Cs, ^{137}Cs, ^{141}Ce, ^{144}Ce
Soil	^{90}Sr, ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, ^{241}Am, ^{242}Cm

Biological concentration processes in fresh water and marine systems can result in rapid transfer and enrichment of certain radionuclides. The radionuclides which enter such systems can in certain cases be rapidly accumulated by plankton and algae, which serve as food for organisms such as oysters, clams, shrimp. Radionuclides of particular concern in fresh water and in marine food chains include: ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{131}I, ^{134}Cs, ^{137}Cs, ^{141}Ce, ^{144}Ce and some of the transuranic elements.

In the case of a reactor core meltdown such as in Japan, the following nuclides are considered to be of concern:

Radionuclides with half-lives of 6 hours or greater[2]: ^{90}Y, ^{91}Sr, ^{93}Y, ^{96}Nb*, ^{97}Zr, ^{99}Mo, ^{105}Rh, ^{109}Pd, ^{111}Ag, ^{113}Pd ^{115}Cd, ^{121}Sn, ^{125}Sn, ^{126}Sb, ^{127}Sb, ***^{131}I***, ***^{132}I***, ^{131m}Te, ***^{132}Te***, ***^{133}I***, ***^{135}I***, ^{140}La, ^{142}Pr*, ^{143}Ce, ^{143}Pr, ^{146}Ba, ^{147}Nd, ^{149}Pm, ^{151}Pm, ^{152m}Eu*, ^{153}Sm, ^{156}Sm, ^{157}Eu, ^{239}Np.

Radionuclides of long term importance: ^{9}H, ^{89}Sr, ***^{90}Sr***, ^{91}Y, ^{93m}Nb, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{110m}Ag, ^{113m}Cd, ^{115m}Cd, ^{121m}Sn, ^{123}Sn, ^{124}Sb, ^{125}Sb, ^{129}I, ***^{134}Cs***, ***^{137}Cs***, ^{141}Ce, ***^{144}Ce***, ^{147}Pm, ^{160}Tb, ***^{238}Pu***, ***^{239}Pu***, ***^{240}Pu***, ***^{241}Am***, ***^{241}Pu***, ***^{242}Cm***, ^{242}Pu, ^{243}Am, ***^{244}Cm***.

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[1] The presence of high levels of the radionuclides of cerium, zirconium, ruthernium and transuranic elements in foods and environmental materials indicates the presence of hot particles which may be of special importance in considering exposure by inhalation and/or ingestion.

[2] Bold italic type denotes radionuclides are of major concern. Asterisks denote shielded fission products.

Guideline Levels

Governments are concerned about limiting radiation doses to populations and have introduced regulations to prevent the distribution of food which exceeds levels of contamination regarded by the Government concerned as significantly detrimental.

The relationship between activity of material ingested and consequent dose is extremely complicated, depending on the biochemical behavior of the element concerned in the body, its chemical form, and the nuclear properties of the active isotope. Cesium however, exhibits very straightforward chemical properties, and it is possible to obtain a very approximate relationship between ^{137}Cs and ^{134}Cs activity and dose from ingestion. Thus an intake of 100 Bq can be equated to about 1 µSv (one millionth of a Sievert) of received dose.

To put the quantities in perspective, let us consider a foodstuff containing 1000 Bq/kg of cesium activity. Suppose also the restrictive limit of 1 mSv (one thousandth of a Sievert) per year to individual members of the public was the rule. Then a person could consume 100 kg of that food during the year assuming this was the only significantly radioactive food eaten.

There is also the concept of collective dose which assumes conservatively that damage attributable to a particular level of dose will occur irrespective of whether that does is received by one person or shared by many. Conventional risk estimates suggest that a fatality is likely with 100 Sv of radiation dose. Thus, ten thousand tons of the contaminated foodstuffs would have to be consumed by the population under consideration before a death could be attributed to the radiation.

Legislation Concerning Radioactive Levels in Food

A month after the Chernobyl accident, the only forms of radioactivity that were to be particularly subjected to import limitation were isotopes of iodine and cesium.

By this time, virtually all iodine had decayed. However, the cesium isotopes concerned were (and still are):

Cesium 137	Half-life 30 years
Cesium 134	Half-life 2.3 years

At the time of the Chernobyl accident, the ratio of these two isotopes distributed across Europe was approximately 2:1.

Different countries can propose their own activity levels on imports notwithstanding the advice of the recommendations of a published FAO/WHO CODEX (see next page). Variations on the limits among countries arise partly from differing eating habits, but chiefly from differing perceptions of risk.

Most countries monitor imported food and reject shipments exceeding regulation limits. Some countries will require a certificate of actual activity level to accompany the shipment.

Problems must arise from food concentration. For example, a quantity of fresh milk may be well inside the regulatory limit. However, when dried and converted to milk powder having only 10% of the liquid, it may then exceed the limit even though the total quantity of activity has not increased.

Half-LIfe, Becquerels, Sieverts, Rems, and Curies – what do they all mean?

- **Half-Life** The activity, or rate, of nuclear disintegration of a radioactive source is proportional to the amount that is present. As it decays, there is less left to decay and so the activity decreases. The half-life is the time over which the activity decreases to 50% of its original value. Half-lives vary from seconds to thousands of years. Long half-life nuclides are more of a problem than short half-life emitters.
- **Becquerels** The Becquerel is an activity of one disintegration per second. It is usual to express specific sample activities in Becquerels per kilogram (Bq/kg) or Becquerels per liter (Bq/l).
- **Sieverts** When radiation strikes cellular tissue, the tissue may be damaged. Radiation may strike a person externally, as in a medical chest x ray, or internally if the radioactive material has been inhaled or ingested, as in the case of food.

  The amount of damage caused or "dose" depends on the radiation type (alpha, beta, or gamma rays) and the energy of the radiation. The dose is measured in Sieverts, Sv, where one Sievert is the dose from one joule, J (0.24 calories), of energy deposited by ionizing radiation in one kilogram of tissue, making allowance for radiation type and the way in which the energy is deposited in huma tissue.
- **Rems** The Rem (Roentgen equivalent man) is an older dose unit.
  100 Rem = 1 Sv.
- **Curies** The Curie (Ci) is an older, inconveniently large activity unit.
  1 Ci = 3.7 x 10^{10} Bq.
- **Nano-curie** (nCi) is sometimes used, where 1 nCi = 37 Bq.

A CODEX issued by the United Nations FAO/WHO (Ref. 2) provides the following guidelines.

Radionuclides

Commodity Code	Product Name	Representative Radionuclides	Dose per unit intake factor in Sv/Bq	Level in Bq/kg	Type	Reference	Note/Remarks
	Infant foods*	^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am		1	GL		
	Infant foods*	^{90}Sr, ^{106}Ru, ^{129}I, ^{131}I, ^{235}U		100	GL		
	Infant foods*	^{35}S**, ^{60}Co, ^{89}Sr, ^{103}Ru, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{192}Ir		1000	GL		
	Infant foods*	^{3}H***, ^{14}C, ^{99}Tc		1000	GL		
	Foods other than infant foods	^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am		10	GL		
	Foods other than infant foods	^{90}Sr, ^{106}Ru, ^{129}I, ^{131}I, ^{235}U		100	GL		
	Foods other than infant foods	^{35}S**, ^{60}Co, ^{89}Sr, ^{103}Ru, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{192}Ir		1000	GL		
	Foods other than infant foods	^{3}H***, ^{14}C, ^{99}Tc		10000	GL		

* When intended for use as such.
** This represents the value for organically bound sulphur.
*** This represents the value for organically bound tritium.

Scope: The Guideline Levels apply to radionuclides contained in foods destined for human consumption and traded internationally, which have been contaminated following a nuclear or radiological emergency. These guideline levels apply to food after reconstitution or as prepared for consumption, i.e., not to dried or concentrated foods, and are based on an intervention exemption level of 1 mSv in a year.

Application: As far as generic radiological protection of food consumers is concerned, when radionuclide levels in food do not exceed the corresponding Guideline Levels, the food should be considered as safe for human consumption. When the Guideline Levels are exceeded, national governments shall decide whether and under what circumstances the food should be distributed within their territory or jurisdiction. National governments may wish to adopt different values for internal use within their own territories where the assumptions concerning food distribution that have been made to derive the Guideline Levels may not apply, e.g., in the case of wide-spread radioactive contamination. For foods that are consumed in small quantities, such as spices, that represent a small percentage of total diet and hence a small addition to the total dose, the Guideline Levels may be increased by a factor of 10.

Radionuclides: The Guideline Levels do not include all radionuclides. Radionuclides included are those important for uptake into the food chain; are usually cotained in nuclear installations or used as a radiation source in large enough quantities to be significant potential contributors to levels in foods, and could be accidentally released into the environment from typical installations or might be employed in malevolent actions. Radionuclides of natural origin are generally excluded from consideration in this document.

In the table, the radionuclides are grouped according to the guideline levels rounded logarithmically by orders of magnitude. Guideline levels are defined for two separate categories “infant foods” and “other foods”. This is because, for a number of radionuclides the sensitivity of infants could pose a problem. The guideline levels have been checked against age-dependent ingestion dose coefficients defined as committed effective dose per unit intake for each radionuclide, which are taken from the “International Basic Safety Standards” (IAEA, 1996).

Multiple radionuclides in foods: The guideline levels have been developed with the understanding that there is no need to add contributions from radionuclides in different groups. Each group should be treated independently. However, the activity concentrations of each radionuclide within the same group should be added together.

Be Prepared

The majority of significant radioactivity likely to be encountered if foodstuffs emit penetrating gamma radiation. The energies of the gamma rays are always characteristic of the radionuclide concerned. Thus, if a device is available which measures not only the quantity of activity but the gamma-ray energy of each component in a sample of foodstuffs, then a measure of each radionuclide in the sample can be made. Such as device is a gamma-ray spectrometer.

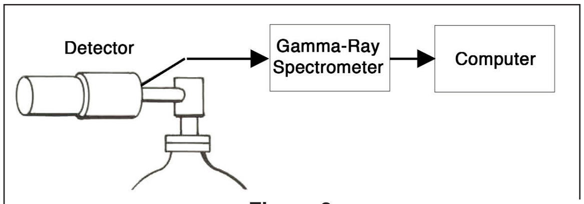

Figure 2.

It is a commonly held belief that a hand-held monitor of the Geiger counter type can be used. This is not so. At these low levels, more sophisticated instruments are required. A lead shield is also required to screen out (as much as possible) other radiations external to the sample itself. This implies that while a mobile laboratory can be constructed in a truck, it is not possible to build a lightweight portable or backpack type instrument to detect the low levels of activity prescribed in the regulatory limits.

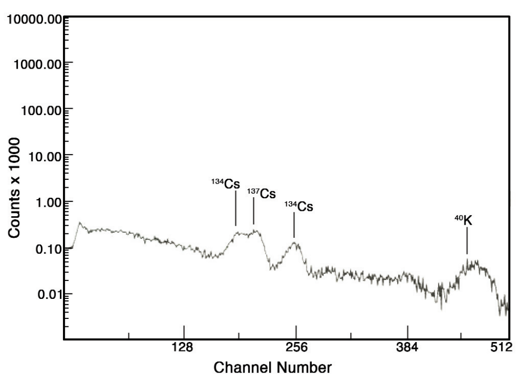

Figure 3.

Figure 2 is a block diagram showing the components of a gamma-ray spectrometry system.

The system generates a pulse-height spectrum that represents the true energy distribution of radiations striking the detector and can be used to determine the gamma-emitting components of the foodstuffs. The spectrum is displayed on a screen as a histogram, and computer software analyzes the distribution, automatically displaying the estimated quantity of each isotope within the foodstuff.

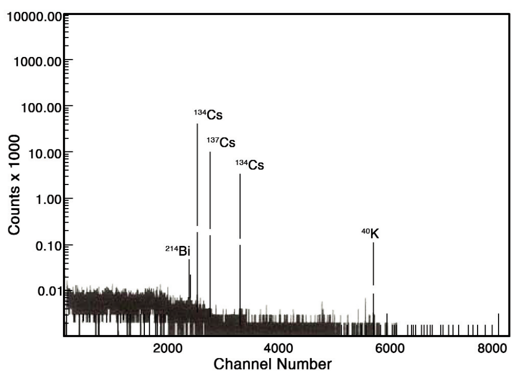

Figure 4.

Two detector types are available. The first is a sodium iodide scintillation detector system (NaI) that is efficient, robust, and relatively inexpensive. However, its discriminatory powers are limited.

Figure 3 shows a peak from the natural ^{40}K that is well separated from the cesium isotope. However, the peaks from ^{137}Cs and ^{134}Cs overlap, making it difficult to determine these isotopes separately.

A more sophisticated detector is a solid-state, high-purity germanium detector system (HPGe) which exhibits remarkable discriminatory power and can distinguish hundreds of different gamma-ray energies in one spectrum.

Figure 4 shows the spectrum from a food sample demonstrating not only the well separated cesium peaks, but a tiny amount of radium.

There are some additional points to consider.

To measure a large food sample as efficiently as possible, the material is contained in a re-entrant vessel (called a Marinelli beaker) which surrounds the detector as much as possible (Fig. 5).

Standard isotopes are used to obtain calibration data. These standards should be traceable to a recognized calibration facility, such as NPL or NBS, to demonstrate to the legislating authority the accuracy of the measurements. Several commercial source suppliers also maintain calibration facilities and their sources are traceable to the national facility.

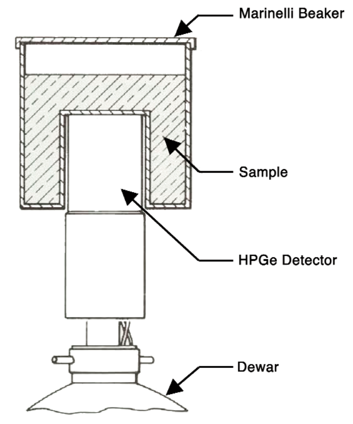

Figure 5.

NaI System

The ORTEC FoodGuard-1 Sodium Iodide Food Monitor (Fig. 6) consists of a 3-in. x 3-in. NaI detector mounted inside a standard lead shield. The digiBASE digital integrated spectrometer connects directly to a photomultipler tube base.

With a suitable PC and customized software for food monitoring application, ease of use and simplicity of operation are assured, allowing rapid investigation or screening of suspect samples, one at a time or in batches.

With a 1-kg sample, a detection limit of 370 Bq of ^{134}Cs or ^{137}Cs can be reached in measurement time of about a minute. Analysis of the data after the acquisition takes a few seconds.

Unlike “gross counting” systems, the computer provides a visual display of the spectrum, which gives reassurance that the only artificial nuclides present are the two cesium isotopes, although the moderately poor resolution of the NaI detector limits the degree of certainty that this is the case.

For example, while the primary nuclides of interest, as a result of the Chernobyl accident, were ^{134}Cs and ^{137}Cs, some instances of ^{110m}Ag contamination were reported.

The main peak of ^{110m}Ag occurs too close in energy to the ^{137}Cs peak for the two to be easily distinguished by an NaI detector.

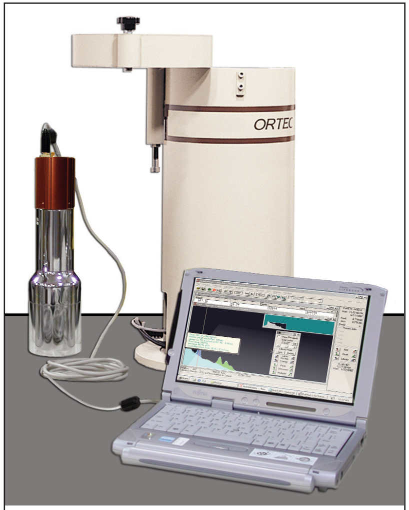

Figure 6.

HPGe System

The ORTEC FoodGuard-2 HPGe Food Monitor (Fig. 7) consists of a 30% (or greater) relative efficiency high-purity germanium detector inside a lead shield with a DSPEC Pro digital integrated gamma spectrometer operating an advanced spectrum analysis software tailored to match the food monitoring application.

This system is capable of detecting radioactive gamma emissions in the energy range of 40 keV to 3 MeV, which covers the emissions of all the nuclides of interest. For an extended energy range down to 5 keV, the ORTEC Gamma-X (GMX) detector can be used.

Figure 7.

With a 1-kg sample, a detection limit of 370 Bq of ^{134}Cs or ^{137}Cs can be reached in a measurement time of several minutes. Analysis times vary depending on the nuclides found, but rarely are more than a few seconds. During analysis, a second sample may be measured on the detector.

Generally speaking, when it comes to HPGe detectors, “bigger is better.” A larger detector of higher than 30% relative efficiency will count samples faster to the same limit of detection. (Put simply, larger detectors are more sensitive.) It has been estimated (Ref. 4) that a single HPGe detector of 90% relative efficiency may count 4 times as many samples to the same detection limit as a single 30% relative efficiency detector. Implemenation of a FoodGuard-2 system based on a 90% relative efficiency HPGe detector is certainly less expensive than 3 or more 30% relative efficiency systems.

The HPGe system has approximately 50 times better resolution than the NaI system. Resolution is the ability to distinguish one nuclide from another of similar energy. The FoodGuard-2 is capable of identifying and quantifying any or all of several hundred nuclides that might be present in the sample (Ref. 3).

For locations with unreliable or without supplies of detector coolant (liquid nitrogen), an electrical cooling option is an attractive alternative. Modern electrical cooling systems are smaller and lower in power than previous units. They can be operated from mains power (100 W) or battery power (minimum 3 hours).

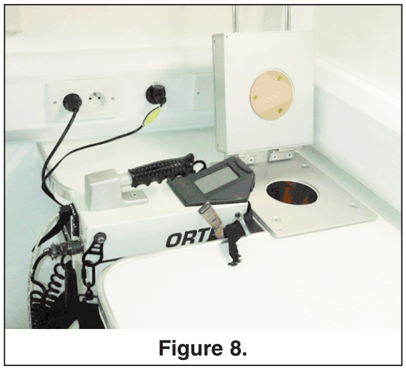

Figure 8.

An ORTEC trans-SPEC or Detective integrated portable spectroscopy system/nuclide identifier used with a suitable lead shield (Fig. 8) can provide an excellent alternative to the more conventional system just described. The "100" models feature a 40% relative efficiency HPGe detector which is ideal for the food monitoring application. In addition, they can also be used in other related screening applications (Ref. 5 and 6).

Which Detector Type Should the Food Industry Use?

The straightforward sodium iodide system (FoodGuard-1) will satisfy many requirements, such as detecting any radiation without regard to nuclide, in which a business is concerned with the distribution and export of foodstuffs under current legislation. It may also be suitable for a manufacturer needing to monitor the individual components of a prepared food. However, where the company concerned has good quality analytical facilities and is concerned about reassuring the public of any possible radioactive component, the more capable FoodGuard-2 system is appropriate.

In either case, the detector should be moderately large in size. The sodium iodide detector should have cylindrical dimensions of 3 in. x 3 in. The germanium detector should have a "relative efficiency" of at least 30–35%. Large detectors are needed to accommodate large samples, and it is usually necessary to measure up to a kilogram (or a liter) of the material to obtain the sensitivity required by some regulations. Indeed, not only is it necessary to measure large samples, but where levels of activity are <10 Bq/kg, a measurement of several hours or even longer is necessary to achieve adequate statistical precision and to ensure that the activity is, in fact, the nuclide in question.

Therefore, when purchasing a system, care should be taken to ensure that not only can it make a suitable measurement, but that it can cope with the required throughput of samples.

What of the Future?

Public awareness and concern about the impact of radioactive contamination has reached a high level which will doubtless lead to new regulations concerning the import, export, and manufacture of foodstuffs in many countries. The food industry can be prepared by the acquisition of the necessary equipment and skills to perform the measurements required.

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