

Detection and measurement of radioactive substances in water and food: a narrative review

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Abstract

Contamination of food and water with radioactive substances is a serious health problem. There are several methods to detect and measure radioactive materials, some of which have been developed in recent years. This paper aims to discuss the methods of detecting and measuring radioactive substances in food and water. The principles and the advantages and disadvantages of each method have been discussed. The results showed that some of these methods, such as spectrometry γ -ray high-purity germanium, portable radon gas surveyor SILENA, RAD7, and inductively coupled plasma mass spectrometry, have a higher sensitivity for detection and measurement. The spectrometry γ -ray high-purity germanium method has attracted more attention than other methods because it can measure a wide range of radionuclides with high resolution.

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Introduction

Although radioactive minerals have always existed in the earth's crust, their radioactivity has become known in the last century. Radioactivity is the property of a nucleus emitting energy. The nuclei of radioactive atoms are constantly changing and disintegrating (Anderson *et al.*, 1957). Radioactive radiation is generally classified as α , β , and γ . Regarding α radiation, it is present in natural and synthetic radionuclides with a high atomic weight and very low penetrating power of high-energy particles. The risk of α radiation is very significant for human beings since it irritates sensitive tissues. β radiation, which also exists as a natural and artificial radionuclide, includes the kinetic energy of electrons with an average penetrating power. Lastly, γ radiation is pure electromagnetic radiation with high penetrating power; therefore, it has greater health risks for humans (Harley, 1979).

Different radioactive materials have different radiation levels. For example, potassium-40, cesium-137, radium-226, and radium-228 have γ -ray radiation. Polonium-238, polonium-239, polonium-240, plutonium-210, and americium-241 have α -like radiation. Strontium-90 has β radiation (Guérin *et al.*, 2017; Heldal *et al.*, 2017). Radionuclides in the environment are produced from both natural and artificial sources. Human beings are always under the influence of energies from both natural and artificial sources. The food chain is one of the ways of transferring radioactive substances to the human body. Food contains natural and synthetic radionuclides, resulting in an effective internal dose after being consumed by humans (Hatif *et al.*, 2018). Many radioactive substances will be damaged when they enter the body (Harley, 1979). Strontium-90 is considered a dangerous contaminant in agro-food chains because, having a strong chemical affinity with calcium, it can be rapidly absorbed and deposited in the bones where it is stored (Iammarino *et al.*, 2015). Radon gas causes tetanus, bronchitis, hemoptysis, lung cancer, and trachea and bronchial cancer. Cesium-137 behaves similarly to sodium and potassium, which are distributed throughout the body and accumulate in soft tissues, resulting in serious health issues such as gastric cancer. High levels of uranium accumulate in the kidneys and ultimately lead to cancer. Radium and strontium behave similarly to calcium and bind to bones. Potassium-40 rapidly reaches the entire bloodstream and tissues and causes cancer (Pól *et al.*, 2010; Guérin *et al.*, 2017; Rahimi *et al.*, 2018). The type of radiation, the half-life, and the source of their production should be specified to analyze radioactive substances (Harley, 1979).

Various types of food naturally contain radionuclides that are transferred from the soil to crops and from water to fish (Vosniakos, 2012). The levels of these natural radionuclides in food and drinking water are generally low and considered safe for human consumption. However, the concentrations of these radionuclides vary among different types of food due to environmental conditions, agricultural practices, and other factors that influence their transfer from the environment to crops and animal

products (Charles, 2001). The concentrations of natural radionuclides also differ within food categories such as vegetables, cereals, fruits, meat, and fish. Additionally, radiation doses from food consumption vary depending on the types of food consumed in different countries (Mercuri *et al.*, 2016). Therefore, it is crucial to monitor radioactivity levels in food and educate consumers about potential risks. Typically, radiation doses from consuming food range from a few tens to a few hundred μSv per year (FAO and WHO, 2017). On average, the global population receives a total radiation dose of about 0.3 mSv each year from radionuclides of natural origin in the diet, representing 10% of the average annual radiation dose of 3 mSv from all sources received by an individual (Charles, 2001).

Many methods are sensitive, selective, fast, and precise and can optimally measure radioactive materials. In recent years, several methods, including portable radon gas surveyor SILENA (PRASSI) (Sahin *et al.*, 2013), RAD7 (Rahimi *et al.*, 2018), spectrometry γ -ray high-purity germanium (Nasreddine *et al.*, 2006), inductively coupled plasma mass spectrometry (ICP-MS) (Pól *et al.*, 2010), and the α spectrometer method (Rožmarić *et al.*, 2012). Gas flow proportional counter and scintillation counting, liquid scintillation counting (LSC), a combination of α -particle, γ -ray spectrometry, and a scintillation counter (Jobbágy *et al.*, 2010) were used with their advantages and limitations. All measurements require an initial sampling method or a radioactive separator (Caroli *et al.*, 2013). The purpose of this review article is to discuss different methods of detecting and measuring radioactive substances in water and food.

Preparation of samples before measurement

The main difference between measuring radioactive substances in food and other analytical experiments is that the initial sample amount is higher compared to other methods (1 to 20 kg) (Harley, 1979). However, for the measurement of pure β -radiation, such as toxic radio strontium-90, since sample preparation is very difficult, a relatively small amount of sample is analyzed (Weller, 2017). For instance, strontium-90 is isolated from a complicated organic mixture through combustion and precipitation processes and then further extracted using strontium-specific resins in solid-phase extraction (Weller, 2017). The equipment should be thoroughly cleaned and used between every preparation to prevent cross-contamination (Nasreddine *et al.*, 2006). A test sample is a form of food ready to be eaten or cooked (Harley, 1979).

There are several steps to prepare samples before the measurements: i) remove dust, surface contamination, and non-edible parts of foods such as citrus peel, apple kernels, shellfish, and softwoods, the outer leaves of leafy vegetables, and the upper parts of root vegetables (Harley, 1979; Jibiri *et al.*, 2007; Lou *et al.*, 2013). For example, for different species of fish, the non-edible parts such as skin, lungs, and ovaries are separated, and the edible parts are mixed and shredded for analysis (Yokota *et al.*, 2017). To remove dust and surface contamination from mushrooms and vegetables, they are completely washed with tap water, the non-edible parts are separated from the edible parts, and then they are prepared for the next step (drying) (Lou *et al.*, 2013). For water and liquid samples, their organic compounds are removed by adding nitric acid and acidifying the environment (Kudo and Kobayashi, 1979); ii) sample size reduction and water removal could be made *via* high-temperature drying, ashing, filtration, and centrifugation, which can ultimately increase the concentration of radioactive materials in

the samples. Different temperatures and time scales are used for drying, depending on the nature of different foods and the desired radionuclide. For example, to measure radium-226 in nuts, a temperature of 80°C for 14 hours or a temperature of 110°C for 24 hours should be used (Ezzulddin *et al.*, 2017; Al-Ghamdi, 2018). Moreover, to measure potassium-40, tritium-232, and radium-226 in vegetables, a temperature of 110°C for 72 hours is used. The samples are then homogenized by being dried, milled, or crushed and sieved. The obtained ash is isolated for a stable equilibrium and conversion of radioactive substances for a certain period; for example, to convert radon to lead and bismuth, one month of isolation is required; to create a balance between radium and bismuth and to prevent the decay of radium into radon gas, 21 to 24 days of isolation are needed; and to create a balance between radon-224 and radon-226, 4-week isolation is required (Nkuba and Mohammed, 2014; Ezzulddin *et al.*, 2017; Al-Ghamdi, 2018). When the concentration of radionuclides such as cesium-137 in the water sample is low and there is a large volume of water (about 100 L), to condense and reduce the volume of water, the water sample is passed through the absorbent resins (cesium-137) and then the resin is transferred (Binesh *et al.*, 2017); iii) when the separation method does not depend on the element's mass, different carriers (for example, barium for radium) are used: the carrier is practical in glass containers to prevent a decrease in nucleotides or the unwanted absorption of the nucleoside (Harley, 1979); iv) in critical situations, rapid preparation methods are applied. In such cases, digesting the sample with microwaves, adding stable radioactive isotopes to the sample, using centrifuges instead of filtration, and using chromatographic columns containing resin are used to increase the action rate; v) when the amount of radioactive material in the sample is very low, it is convenient to load and dispose (scavenging) of the sediment containing radionuclides by adding a carrier group. This method is suitable for separating radioactive material from the whole sample; vi) in chemical separation, different methods such as deposition, ion exchange, distillation, and electrolysis are used (Harley, 1979). In this case, when the level of some actinides such as Pu, Th, and U isotopes is less than the 0.1 maximum permitted levels, and the sample size is large, the separation will be done by ion exchange or chromatography column (Caroli *et al.*, 2013; Brandhoff *et al.*, 2016).

Accurate chromatography separation is essential so that the signals are separated and do not interfere with one another, as they reduce the detection limit (Brandhoff *et al.*, 2016). In certain cases, selective solvents used to extract the radioactive isotopes are employed to increase the analysis rate. For example, direct uranium extraction from water using a lipophylic toluene-based scintillation solution mixed with bis (2-ethylhexyl) phosphoric acid. This specific reagent was also extensively used for strontium-90 determination (Forte *et al.*, 2001).

Methods of diagnosis and measurement

Portable radon gas surveyor SILENA

This method is a portable sensing system and one of the oldest methods with suitable properties for measuring the concentration of radon gas in water (Forozani and Soori, 2011; Binesh *et al.*, 2012). In this method, the concentration of radon and radium in water samples is directly measured through the chamber (Sahin *et al.*, 2013). The tool detector is a scintillation cell coated with silver-activated zinc sulfide [ZNS (Ag)] with a volume of 1830 cm^3 (Binesh *et al.*, 2011), indicating the radon activity in m^3 of water

by counting the α -particles emitted from radon and its daughter nuclei. The PRASSI pump circuit operates at 3 L/min for gas sealing (Binesh *et al.*, 2012). The sensitivity of this system is 4 Bq/m³ in a continuous state for one hour (Binesh *et al.*, 2011).

Principles

This measurement system comprises a bubbler and a dryer column, which are used to measure radon concentration in water samples carried out in a closed-loop circuit. A short plastic hose connects the plastic can to the faucet for sampling. After a few minutes, the water sample is collected in the canal by decreasing the water flow rate (Binesh *et al.*, 2011). Water sampling from external sources (other than the faucet) is performed under reverse pressure from a 25-cm depth of water because the water movement allows radon loss to be high. Radon levels are abundant in deep springs and waters. Therefore, sampling is about -30-25 cm below the free water level. The samples are stored in a cool place and transferred to the laboratory in the shortest amount of time possible. The specimens are left in a dark place for 25 to 60 days to reach equilibrium (Binesh *et al.*, 2011; Forozani and Soori, 2011).

The numbers represented by the device are based on Bq/m³, which is calculated using Equation 1, radon gas density (Bq/L) (Binesh *et al.*, 2011).

$$Q_{Rn} \left(\frac{Bq}{L} \right) = Q_{PRASSI} \times \frac{V_{tot}(m^3)}{V_{(sample)}} \quad [Eq. 1]$$

where: Q_{Rn} is the concentration of radon, Q_{PRASSI} is the value recorded by the device, V_{tot} is the total volume of air connections and V_{sample} is the volume sample, and a correction factor in the delay measurement is within the brackets.

Advantages

This device is light and portable; its most crucial feature is high sensitivity and short response time. This method is one of the most reliable methods for measuring radon-222.

RAD7

The detector of this device is a solid-state semiconductor type and scintillator based on the number of α -particles emitted from radionuclides such as radon-222 and thoron (radon-220) and converts α radiation directly into an electrical signal (Rahimi *et al.*, 2018; Chmielewska *et al.*, 2020).

A special kit of RAD H2O accessories is used to measure radon activity in water samples using a RAD7 detector. This kit has 40 and 250 mL vials, connecting hoses, aerator broadcasts, and a RAD7 detector (Chmielewska *et al.*, 2020). The kit is attached to a bubble kit capable of removing radon from the water sample in the closed loop and a scintillator coated with zinc sulfide that can detect α activity (Snihs, 1983). This method uses a closed-loop aeration design where the volume of air and water is kept constant (Al-jnaby, 2016).

Principles

The inner hole of the RAD7 consists of a hemisphere with a solid-state silicon semiconductor detector located in the center of the hemisphere. Applying a high voltage (2000-2500 V) to the conductor generates an electric field, which causes the positively charged particles to be directed to the surface of the detector (Ravikumar and Somashekar, 2014; Chmielewska *et al.*, 2020;). Then, the electrical signal recorded by the radionuclide decay is

amplified, filtered, and arranged based on its power (Mehra and Bala, 2014). The device bubbles use a closed cycle for 5 to 10 minutes so that more than 95% of radon is separated from water. The pump is then switched off, and it takes 5 minutes to have a steady balance between water, air, and radon daughters attached to the system detector. Air filters are used to prevent dust particles and charged ions from entering. The radon gas concentration is then measured by passing through the air filter tube. During the measurement period, radon-222 gas decays into the chamber, which leads to the production of α -particles. Each α -particle is recorded by the detector due to its energy, which is determined by the number of particles recorded. The humidity should remain below 10% and should be removed from the radon before the measurement begins because detection efficiency decreases with increasing relative humidity. Radon gas enters the glass lamp containing calcium or calcium salts to remove any moisture (Mehra and Bala, 2014; Ravikumar and Somashekar, 2014; Rahimi *et al.*, 2018; Hatif *et al.*, 2018). It should be noted that if the humidity exceeds 10% before the end of the first counting cycle, an error will occur (Abojassim *et al.*, 2017). Water testing in a 250-mL protocol is typically completed within 30 minutes, and the radon concentration in the water is reported as an average (Al-jnaby, 2016).

Advantages

The most significant feature of this device is that its electronic detector is portable (Mehra and Bala, 2014; Hatif *et al.*, 2018). This device can evaluate inaccurate measurements, and the results are analyzed within 30 minutes. The device sensitivity is similar to or even better than the liquid scintillation methods, and no harmful chemicals are needed (Tabar and Yakut, 2014). Samples are analyzed immediately at the sampling site (Duggal *et al.*, 2018). Another advantage of the solid-state detector in detecting radon generation is its ability to determine the energy associated with α -particles entering the device electronically, such as radionuclides polonium-218. Po-specific iodine can be identified by the energy of 6.00 MeV α radiation or polonium-214 by the energy of 7.69 MeV (Mehra and Bala, 2014). This system can detect the sample pulse/signal from annoying noises (Tabar and Yakut, 2014).

Spectrometry γ -ray high-purity germanium

Germanium semiconductor detectors were introduced in 1962 and are currently the desired detectors for high-resolution γ -ray spectrometric studies (Khandaker, 2011). These detectors can be used to detect photon beams and specific actinide rays. As an example, photons of 185.7 kV 235U can be readily observed with low energy γ spectroscopy (D). High-purity germanium is embedded in a cylindrical lead shield with an inner coating of copper and cadmium. The thickness of the cylindrical shield is much greater than that of the internal coating. The cylindrical shield around the system reduces background radiation and radiation (Görür *et al.*, 2012). High-purity germanium detectors located in conventional lead shields have different types with different relative efficiencies, including i) a 100% efficiency coaxial detector (CANBERRA, Canberra Packard, Schwadorf, Austria) with electrolytic copper crystals with carbon window; ii) a CAN detector with 150% crystalline efficiency made of electrolyte copper (good dimensions: 32.5 mm, depth 73 mm) and thin copper walls; iii) 170% coaxial detector (EURISYS, Canberra Packard, Schwadorf, Austria) with crystalline and al-made window; iv) 200% efficiency detector (CANBERRA, Canberra Packard, Schwadorf, Austria) with electrolytic crystals made of copper (ideal dimensions of 25 mm diameter and 60 mm depth) and fine walls made of thin copper; v) thal-

limum doped sodium iodide crystal [NaI(Tl)]: the detector is used for radioactive measurement in Marin. The NaI(Tl) detector comprises a crystalline structure and typically demonstrates an energy resolution of 7.0 for the absorption of 662 keV γ -rays (L'Annunziata, 2012) with poor energy resolution. Only if the energy and probability of the photons emitted by the radionuclides are high enough and the background is low enough will they be sufficiently resolved (Jibiri *et al.*, 2007).

Principles

This method has two detectors and an electronic device. The detector is high-purity germanium with a significant spectroscopy component (Görür *et al.*, 2012), which receives the radiation and converts it into an electrical peak as a core component. Since this peak is weak, changes are required to make it ideal for stabilization, amplification, and isolation for proper analysis and inference; therefore, it is improved in the amplifier. The second part of the system is an electronic component, which includes a power supply, an along-to-digital converter amplifier, a multichannel analyzer, a computer, and a printer. Some devices also have a pre-amplifier, usually with a detector (L'Annunziata, 2012). In the γ spectrometer (Figure 1) (Yoo *et al.*, 2015), the specimens are placed in the detector chamber, and the device is adjusted. The count, the number, and the file's name are recorded. Then, the counting begins for 12-24 hours, depending on the type and concentration of the radioactive elements. The peaks of the factors measured on the monitor determine their type and amount. For example, cesium-137 shows a count at the peak of 661 Kev. The γ -rays emitted in the energy range between 50 keV and 3000 keV are used to measure the energy spectrum. Raw data are obtained from Pc and are analyzed, and final responses are extracted (Binesh *et al.*, 2011). Different γ -ray energies are used for measuring the activity of radionuclides for radioactive materials, for example, 351.92 keV for lead-214, 609.31

Kev for bismuth-214, and 911.07 keV for thorium-232. Energy calibration and efficiency are performed using mixed γ radionuclide reference standards (Görür *et al.*, 2012; Meli *et al.*, 2016).

Advantages

The advantage of this method is that, while it is cost-effective, it can also be used to detect a wide range of radionuclides (L'Annunziata, 2012). High-purity germanium has high purity, low ionization energy required to generate electron pairs, high conductivity, fast response time (swift electron motion), high resolution, and good resolution between two rays (their energy levels are very close to one another); also, it is simple with high efficiency (L'Annunziata, 2012; Khandaker, 2011). In this method, measuring the activity of radionuclides that produce high-energy γ -rays is relatively simple.

Disadvantages

A large sample size is needed because of the high detection limit in the γ spectrometry method.

α spectrometry method

The α spectrometer has a semiconductor silicon detector (Skwarzec *et al.*, 2003) widely used for α -emitter nucleic acid analysis (L'Annunziata, 2012). Many actinides are α -emitters (Fukushima's forgotten radionuclides). The usual analysis of actinide-emitter α has complex and time-consuming chemical separations (extraction chromatography, sedimentation, electrical deposition, *etc.*) with relatively low counting efficiency. Since higher detection limits and relatively small sample sizes are required in critical situations, ion chromatography on a chemical resin can reduce the time needed for sample preparation (Forte *et al.*, 2001; Jobbágy *et al.*, 2010; Meli *et al.*, 2016). The enumeration efficiency of these methods is strongly influenced by the total sol-

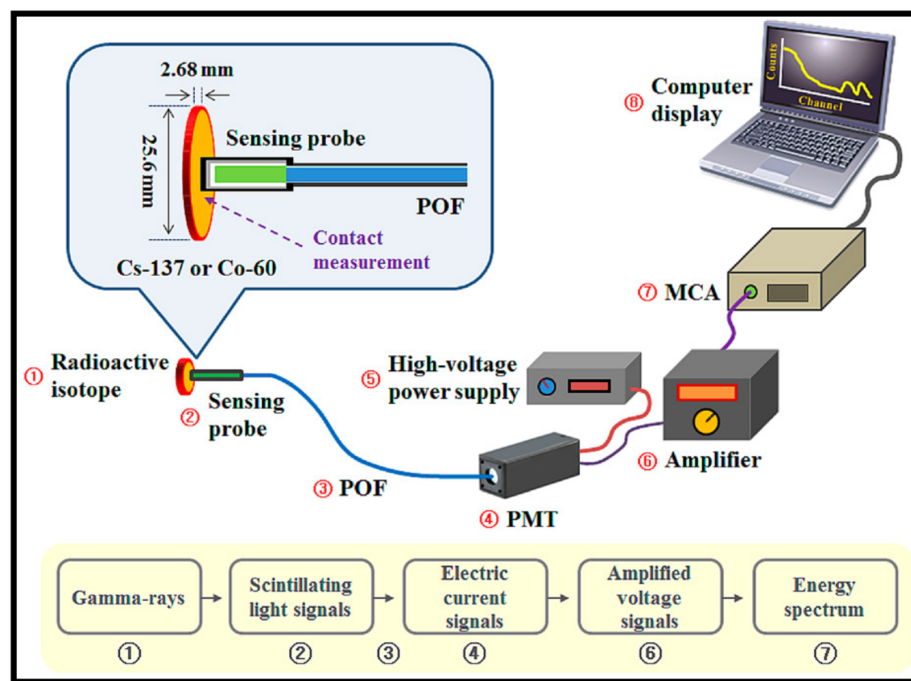


Figure 1. Schematic of spectrometry γ -ray high purity germanium. Reproduced from: Yoo *et al.*, 2015. Cs-137, cesium-137; Co-60, cobalt-60; POF, plastic optical fiber; PMT, photomultiplier tube; MCA, multichannel analyzer.

uble solids and the chemical composition of the water or sample (Jobbágy *et al.*, 2010).

Principles

This method requires long preparation and source counting (Meli *et al.*, 2016). The post-prepared sample (ash) is directed to the planchet source/stainless steel plate/discs of 50mm diameter. The counting of α -particles is done in 80,000 seconds. A radioactive substance such as radium-226 should be measured immediately after preparation to minimize the growth effects of the radionuclide (Desideri *et al.*, 2007). The measurements are made at a distance of 5 mm from the detector (Idoeta *et al.*, 2018) (Figure 2) (Liu *et al.*, 2017).

Advantages

The α spectrometry is a rapid screening method with the best energy resolution of α -particles and the lowest background. When the α source is prepared correctly, no significant self-absorption is detected. Volatile compounds can be measured if the sample and preparation conditions are selected with care (*e.g.*, polonium-210) (Jobbágy *et al.*, 2010). This method is also simple, sensitive, inexpensive, and does not require expensive equipment (Desideri *et al.*, 2007; Meli *et al.*, 2016). In this method, some α -particles produced in the uranium-238, uranium-235, and thorium-232 series can be measured directly (Bonotto *et al.*, 2009). The α spectroscopy has the highest sensitivity for accurately evaluating uranium isotope ratios, which is essential for detecting low and attenuated amounts in environmental samples (Forte *et al.*, 2001).

Disadvantages

Since the surface of the detector is small, there is less sensitivity, and only samples with higher relative concentrations of α -emitters can be analyzed. However, high-resolution silicon detectors and deconvolution software can solve this problem (L'Annunziata, 2012). Water evaporation time, soluble solids in water, and the number of samples that can be prepared simultaneously for counting are considered limitations (Desideri *et al.*, 2007).

Gas flow proportional counter and scintillation counting

Gas flow detectors, designed to detect all kinds of radioactive

materials, are nuclear detectors. The detectors have a metal chamber filled with gas that contains an anode wire in which the gas flows continuously, and when the radioactive substances pass through the gas, the process of ionization and excitation of gas molecules occurs along its path. Gas-ionization rays are transmitted into the room. The electrons are absorbed into the anode wire and collected to produce an electric pulse. High-density ions can be recombined with electrons at low anode voltages. Almost all electrons are accelerated to the anode at high voltages, and the detector is known as an ionization chamber and counter. The ionization chamber is the simplest type of gas-filled detector. It contains a chamber with two electrodes with an electrical potential that generates an electric current by absorbing opposing ions. Ion flow is measured by electrical amplification in dose units. In addition, these detectors can measure low-energy α - and β -particles (Al-Ghamdi, 2018). The flow proportional counting method used to measure α and β in drinking water samples requires more chemical treatment in comparison with evaporative methods. Some properties of the detector are affected by sample preparation. The amount of sample in the detector plays a vital role in obtaining reliable results to create a uniform thickness and form a homogeneous layer of residual material in the planchet. Time is also an essential factor; for example, the half-life of radium-224 is 3.66 days, and due to the delay between sampling and analysis of α -rays, it cannot be detected *via* conventional methods. Therefore, it is advisable to evaluate α activity within 48 hours. It is also preferable to count the samples on a scale that is proportional to the low background gas flow. In general, the counting time may vary from a few hours to several days. Finally, due to the energy loss and self-absorption of α - and β -particles in the sample matrix, their counting efficiency is much less than 100% (Jobbágy *et al.*, 2010).

Principles

A certain amount of water sample (0.1 to 2 L of the initial sample volume in soluble solids) evaporates slowly and forms a dry residue or sediment. α - and β -particle absorption in the final sediment depends on the density of the source surface, which must be carefully controlled according to different standards and protocols. This value ranges from 0.5 to 25 mg/cm² to obtain satisfactory counting statistics. The precipitate must be heated on an asbestos pad with a gas burner for 1 minute to eliminate nitrates. The surface should be fragile and uniform, especially in saline waters. The

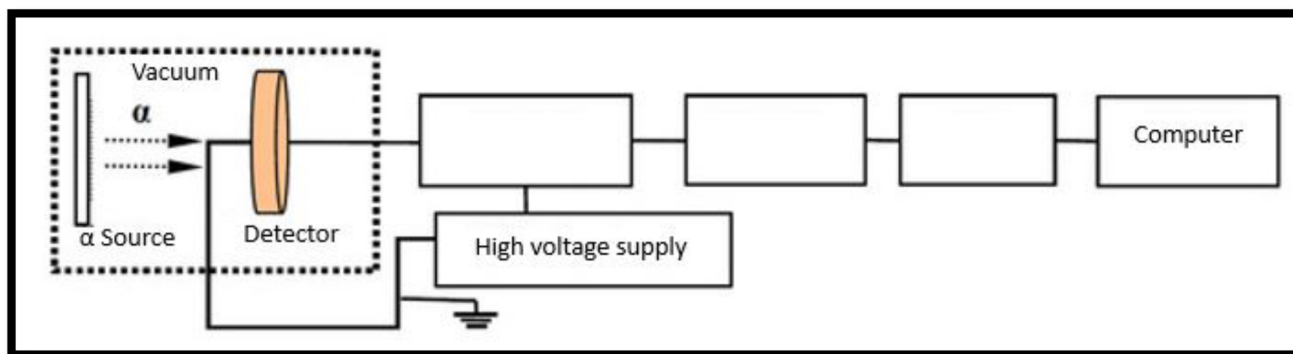


Figure 2. Schematic of α spectrometry. Reproduced from Liu *et al.*, 2017.

final α and β activity of the filtered and dried sediment is counted by a ZnS (Ag) scintillation detector (Jobbágy *et al.*, 2010).

Advantages

This method is beneficial for high-power screening. The α and β activities can be determined simultaneously, reducing analysis time. Several detectors can be installed in a single chamber so that more samples can be measured simultaneously; moreover, the transmission of α and β signals is low (Jobbágy *et al.*, 2010).

Disadvantages

Volatile radionuclides are not detected in a sample that has been evaporated and re-exposed to heat (polonium-210 above 100°C). Therefore, low α and β activity concentrations may not be measured (Jobbágy *et al.*, 2010).

Liquid scintillation counting

The radionuclide in LSC is mixed with a fluorine cocktail (Liu *et al.*, 2017) (Figure 3). The decay energy is transferred to the cocktail, and radionuclide activity is measured by photon counting using a photomultiplier tube (PMT). Aromatic solvents are used due to the high density of electrons associated with these solvents. When they react with β -particles, a large amount of fluorescence is produced. PMTs are not sensitive to the fluorescence wavelength of aromatic solvents; thus, the scintillator absorbs solvent energy and quickly detects light (L'Annunziata *et al.*, 2020). The gross α and β activities can be measured using LSC (Desideri *et al.*, 2007). The α energy can be partially separated by this method. The obtained resolution is not as accurate as the resolution obtained by α spectroscopy; therefore, the identification of α -emitting radionuclides becomes complicated. In some countries (Italy), the gross activity of α and β in water samples is determined by LSC according to the ISO 11704 standard (Jobbágy *et al.*, 2010). Different types of radiation, including α -, β -, and γ -particles, can be measured by LSC. LSC is the most commonly used method among β detectors (Jobbágy *et al.*, 2010).

Principles

An aliquot of water (50-200 mL) is acidified to a pH=1.5-2.5 and evaporated onto the hot plate to reach a volume of about 10 mL. They are sometimes stirred to eliminate radon and its short-lived daughters to prevent salt deposition (Yokota *et al.*, 2017). The water sample is mixed with a scintillation cocktail in a 20 mL polyethylene vial. Choosing the correct type of vial is crucial. Given the potassium-40 content, glass vials have a higher degree of backing compared to polyethylene vials because organic solvents may penetrate the walls of polyethylene vials. Using low-emission polyethylene coated with teflon or low-potassium glass vials with copper warheads is recommended to achieve a low background and to prevent scintillation cocktails from spreading on the walls of the counting vial (Jobbágy *et al.*, 2010).

The presence of some metal ions in water affects the counting; therefore, it is of great importance to properly adjust the type of vial, cocktail, and α/β pulse discrimination to determine the counting efficiency for LSC α/β measurements. Some factors, such as the physicochemical properties of the radionuclides, soluble anions, and energy released from particulate matter (Orita *et al.*, 2017), should be used during calibration measurements (Jobbágy *et al.*, 2010). The use of LSC and specialty resins were developed for this purpose. As the water passes through specific chromatographic resins, the resin is extracted from the column, then dried and mixed with a suitable scintillation cocktail, and is then counted by LSC (Jobbágy *et al.*, 2010; Hatif *et al.*, 2018).

Advantages

This method has high detection efficiency (100%) and a low background count tool for determining gross α/β activities, which is suitable for specific isotope analysis when rapid information is needed (Jobbágy *et al.*, 2010). LSC is a valid test for measuring non-volatile natural α -emitting radionuclides in drinking water (counting radium-226 determination in complex samples using liquid scintillation) (Yokota *et al.*, 2017). Compared to other methods, the easy sample preparation, optimal detection power, and high speed of this technique have made its overall performance satisfactory (Caroli *et al.*, 2013). The high counting efficiency in a

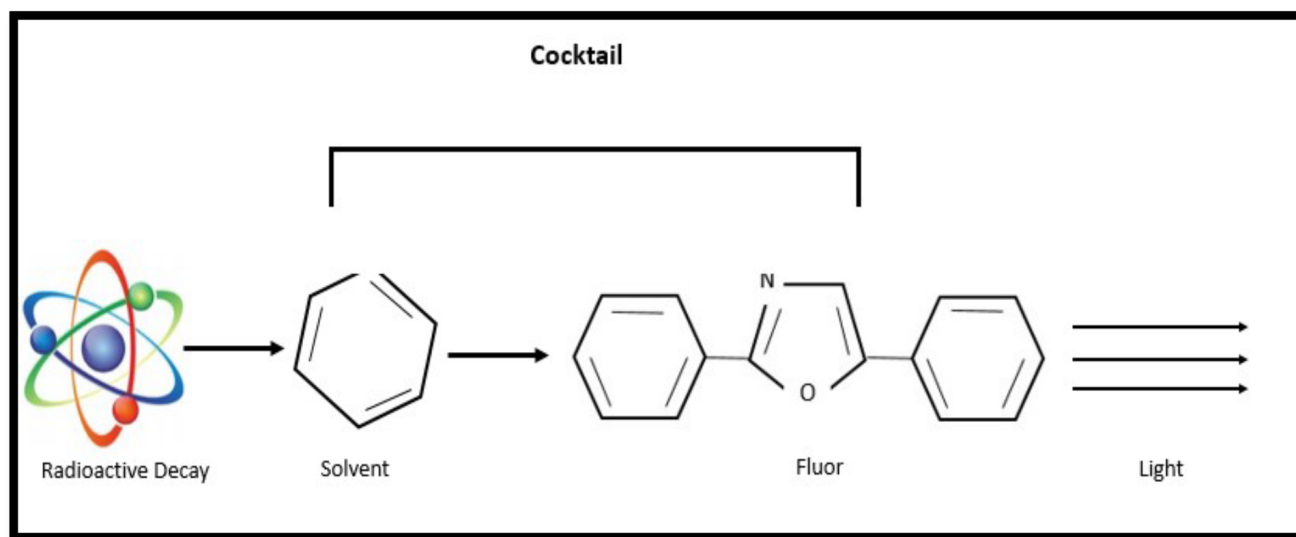


Figure 3. Liquid scintillation counting for the determination of β emitter.

scintillating cocktail is due to the fact that the sample is thoroughly dissolved and homogenized (Jobbágy *et al.*, 2010).

Disadvantages

In this method, the resolution is poor, and complete separation of β and α energy may not be performed, which is considered an obvious disadvantage (Caroli *et al.*, 2013). This technique is also expensive (Desideri *et al.*, 2007), and because of the installation of a detector on one device, only one sample is counted at a time (Jobbágy *et al.*, 2010). Most methods, like LSC, cannot be used for food analysis. The few available methods for food analysis have several drawbacks, including time-consuming processes, the use of dangerous chemicals, low repeatability and sensitivity, and interference from other radioactive substances. Additionally, there is a lack of validation procedures, which is a significant limitation for laboratories responsible for official food controls, as they need fully validated radiochemical procedures to meet ISO 17025 requirements. For this reason, several studies have focused on the validation and optimization of these methods in food. In the study of Iammarino *et al.* (2016), a multi-matrix ultrasensitive radiochemical method was optimized and validated for the determination of radiostrontium in solid food (wheat and derived products, seafood, meat, and dairy products) by liquid scintillation counting (Iammarino *et al.*, 2016). The findings indicated that the minimum detectable activity was 8.0 mBq per kg, with a mean repeatability of 14.5% and a recovery rate of 90.5%. Marchesani *et al.* (2022) developed and validated a rapid and highly sensitive technique to detect strontium-90 in solid food items like meat, dairy products, seafood, vegetables, and animal feed. This method involves the use of a specialized resin for extraction and ultra-low-level liquid scintillation counting for detection. The result showed that the minimum detectable activity was 11 mBq kg⁻¹, with an average repeatability of 10.7% and a recovery rate of 100.1%. In another study conducted by dell'Oro *et al.* (2014), an analytical technique using liquid scintillation counting to accurately measure low levels of radiostrontium in milk was developed. The results demonstrated strong selectivity, with a detection limit of 0.006 Bq L⁻¹, a repeatability value of 13% expressed as coefficient of variation%, and a mean recovery value of 102.5% (dell'Oro *et al.*, 2014).

Spectrometry combination of α -particle and γ -ray

This method uses α - and γ -ray spectroscopy to determine α and β gross radioactivity in water (Bonotto *et al.*, 2009; Khandaker, 2011).

Principles

An alternative method to determine gross α and β radioactivity in water is using α -particle and γ -ray spectroscopy. This method includes 2-stage evaporation of water samples. A relatively inexpensive sodium iodide activated with a trace amount of thallium scintillation detector is used for counting γ -ray and a surface barrier detector for α -particles. The water sample evaporates in two stages. The two detectors being used for γ -ray counting are relatively inexpensive thallium scintillation and a barrier-level detector for α -particles (Jobbágy *et al.*, 2010).

Advantages

This method requires no specific reagents or laboratory equipment. Sample preparation is simple, and the measuring equipment is cheap, with the minimum detectable activity of gross α (0.001 Bq/L) and gross β (0.03 Bq/L). Direct detection of radionuclides is possible after system calibration (Jobbágy *et al.*, 2010).

Disadvantages

This method requires both types of detectors in the laboratory. An analyst is needed to analyze the data and calibrate the system. This method is more time-consuming, and complex compared to other standard techniques. Quantitative data on the radionuclides of α radiation in the sample do not appear to be accurate; therefore, more sophisticated and reliable methods should be used for specific nucleoid analysis, for example, isotope separation step and α spectrometry or LSC (Sujo *et al.*, 2004; Bonotto *et al.*, 2009; Jobbágy *et al.*, 2010).

Scintillation counter

Scintillation means producing small flashes of light. Some crystals, such as sodium iodide, convert ionization and radiation-stimulated products into scintillation optical pulses. The light pro-

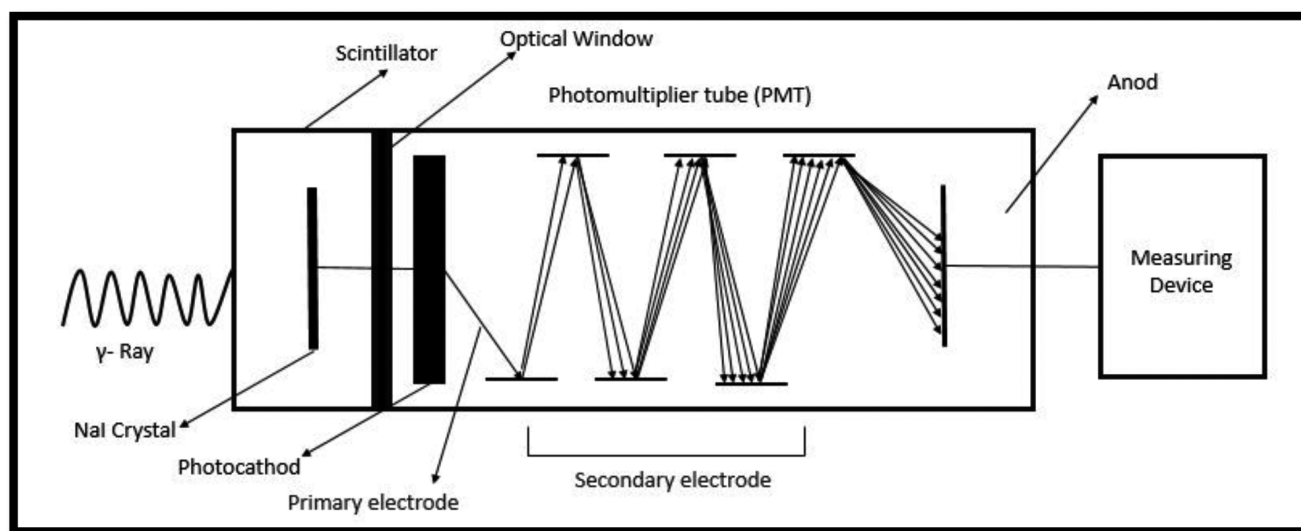


Figure 4. The main components and operation of a scintillator detector.

duced is proportional to the energy the radiation/photon particles store. The small light pulse is transformed into an electric pulse by an electrical component called a PMT. The size of the amplified electric pulse is proportional to the energy stored by the photon/radiation particle. Thallium-activated sodium iodide and cesium iodide crystals and a wide range of plastics are used as the dominant sodium iodide detector for high-resolution γ detection, which is affordable. Many types of plastic scintillators are commercially available and are suitable for detecting charged particles or neutrons in rapid time. The essential components and agents of the scintillation detector operation are shown in Figure 4. The counter with solid scintillation detector materials such as ZnS (Ag) is used for water analysis. ZnS (Ag) sensors and plastic detectors are used for sample preparation, allowing simultaneous collection of α and β (Jobbágy *et al.*, 2010).

Advantages

This method is easy to maintain and operate. Operational and maintenance costs are relatively low (Jobbágy *et al.*, 2010).

Disadvantages

Detectors should be kept in the dark and the lead compartment. The counting efficiency is lower than that of counters proportional to gas flow, but it can be significantly increased by a vacuum. Detection of volatile nucleoids is also not possible, and self-absorption may be significant. Besides, several transitions occur between α and β signals (Jobbágy *et al.*, 2010).

Inductively coupled plasma mass spectrometry method

Several non-radiometric methods based on mass spectrometry (MS) have been introduced for the quantitative determination of radionuclides. Different types of MS instruments largely depend on the variety of ion sources and the detection unit. The most successful methods are as follows: inductively coupled plasma, direct current plasma, microwave-induced plasma, spark source, thermal ionization, laser microprobe, and secondary ion systems. Specifically, ICP-MS has the potential to detect radionuclides in a wide range of environmental, clinical, and nutritional materials (Caroli *et al.*, 2013). ICP-MS is the latest susceptible and non-radiometric method (Desideri *et al.*, 2007).

Principles

This method measures the mass-to-charge ratio. Sample molecules (as vapor) become ionized and charged molecules in the ionizing portion. The ions pass through the magnetic field of the analyzers and are separated by mass-to-charge ratio. These ions are sent to the recorder to detect the signal and obtain a proportion of voltage (Pól *et al.*, 2010).

Advantages

Currently, ICP-MS is one of the leading techniques for detecting trace elements with high accuracy. This method could screen several elements with a wide dynamic range (ng/L to mg/L). In addition, with its rapid analysis, it can measure all actinide elements in concentrations of less than 1pgmL^{-1} in liquid samples within 1 minute (Santos *et al.*, 2010). This method has a plasma temperature range of 6000-10,000 K, which is relatively free of contamination and interference in the sample matrix. The detection limit in this method is equal to or better than atomic absorption spectroscopy (Caroli *et al.*, 2013). The cost is low, and the technique is suitable for analyzing small samples. Moreover, accurate methods such as

chromatography eliminate potential interference and are widely used to analyze long-lived radioisotopes (L'Annunziata, 2012). This method can determine the contamination of foodstuffs with plutonium, americium, and neptunium at the same time within 3 hours of sampling (Brandhoff *et al.*, 2016). ICP-MS was also extensively used to determine chemical yields in radiochemical analysis (Iammarino *et al.*, 2016). ICP-MS is the fastest method to measure the concentrations of isotopes because no pre-processing is required and the analysis is performed directly and quickly, in a few minutes. Moreover, the number of sample preparation steps can be significantly reduced due to the separation of elements by mass/charge ratio (Forte *et al.*, 2001; Santos *et al.*, 2010).

Disadvantages

The main drawbacks of this technique are the initial cost of the equipment and the daily maintenance costs. The mass spectra produced by ICP-MS are relatively simple, but that does not mean the technique is free from spectral interference caused by isobaric ions. Isobaric interactions occur when two isotopes of the same mass are present. Most elements generally have more than one isotope, and there is a good chance of choosing an isotope unaffected by isobaric interference (Caroli *et al.*, 2013). Additionally, organic compounds, acidic conditions, salt, and soluble solids can affect ICP-MS in addition to spectral and non-spectral interactions (Santos *et al.*, 2010).

Conclusions

This review article evaluates different methods of measuring radioactive substances in water and food. The principles, advantages and disadvantages of each method have been assessed. Briefly, it can be said that RAD7 and PRASSI methods have high sensitivity and the ability to measure radionuclides (portable), and this measurement is direct and fast. ICP-MS also has high accuracy. Gas flow proportion can simultaneously measure α and β activity in water with short-half-life radioactive materials. LSC is used to determine α/β gross activity and its high detection efficiency. Combining α -particle and γ -ray spectrometry and scintillation counter are inexpensive and simple methods for the detection of α/β gross. The spectrometry has the highest resolution for α -particles and the lowest background. Spectrometry γ -ray high-purity germanium method is better for measuring because of its ability to measure a wide range of radionuclides with high resolution, efficiency, and speed.

References

- Abojassim AA, Kadhim SH, Mraity HAA, Munim RR, 2017. Radon levels in different types of bottled drinking water and carbonated drinks in Iraqi markets. *Water Supply* 17:206-11.
- Al-Ghamdi AH, 2018. Determination of natural radioactivity concentration in consumed nuts and seeds and their implications in human body. *Univers J Public Health* 6:198-202.
- Al-jnaby MKM, 2016. Radon concentration in drinking water samples at Hilla city, Iraq. *WSN* 52:130-42.
- Anderson EC, Schuch RL, Fisher WR, Langham W, 1957. Radioactivity of people and foods. *Science* 125:1273-8.
- Binesh A, Arabshahi H, Pourhabib Z, 2011. Radioactivity and dose assessment of heavy radioactive pollution, radon and radium from water sources of 3 northern regions in Iran. *Int J Phys Sci*

- 6:7969-77.
- Binesh A, Mohammadi S, Mowlavi AA, Parvaresh P, 2017. Evaluation of the radiation dose from radon ingestion and inhalation in drinking water. *Afr J Water Conserv Sustain* 5:201-05.
- Binesh A, Mowlavi AA, Mohammadi S, 2012. Estimation of the effective dose from radon ingestion and inhalation in drinking water sources of Mashhad, Iran. *Iran J Radiat Res* 10:37-41.
- Bonotto DM, Bueno TO, Tessari BW, Silva A, 2009. The natural radioactivity in water by gross alpha and beta measurements. *Radiat Meas* 44:92-101.
- Brandhoff PN, Van Bourgondiën MJ, Onstenk CGM, Vos Van Avezathe A, Peters RJB, 2016. Operation and performance of a national monitoring network for radioactivity in food. *Food Control* 64:87-97.
- Caroli S, Forte M, Nuccetelli C, Rusconi R, Risica S, 2013. A short review on radioactivity in drinking water as assessed by radiometric and inductively coupled plasma-mass spectrometry techniques. *Microchem J* 107:95-100.
- Charles M, 2001. UNSCEAR Report 2000: sources and effects of ionizing radiation.
- Chmielewska I, Chałupnik S, Wysocka M, Smoliński A, 2020. Radium measurements in bottled natural mineral-, spring- and medicinal waters from Poland. *Water Resour Ind* 24:100133.
- dell'Oro D, Iammarino M, Bortone N, Mangiacotti M, Chiaravalle AE, 2014. Determination of radiostrontium in milk samples by ultra-low-level liquid scintillation counting: a validated approach. *Food Addit Contam Part A Chem Anal Control Expo Risk Assess* 31:2014-21.
- Desideri D, Roselli C, Feduzi L, Meli MA, 2007. Radiological characterization of drinking waters in Central Italy. *Microchem J* 87:13-9.
- Duggal V, Sharma S, Kumar Srivastava A, Mehra R, 2018. Measurement of radon concentration in drinking water in Bhiwani district of Haryana. *J Geol Soc India* 91:700-3.
- Ezzulddin SK, Ahmed AH, Samad AI, Othman SQ, 2017. Radioactivity measurement of nuts and seeds available in Erbil city markets. *AIP Conf Proc* 1888:020022.
- FAO, WHO, 2017. Evaluation of certain contaminants in food: eighty-third report of the joint FAO/WHO expert committee on food additives. Available from: <https://iris.who.int/bitstream/handle/10665/254893/9789241210027-eng.pdf?sequence=1>.
- Forozani G, Soori G, 2011. Study on radon and radium concentrations in drinking water in west region of Iran. *arXiv*: 1106.3646.
- Forte M, Rusconi R, Margini C, Abbate G, Maltese S, Badalamenti P, Bellinzona S, 2001. Determination of uranium isotopes in food and environmental samples by different techniques: a comparison. *Radiat Prot Dosimetry* 97:325-8.
- Görür FK, Keser R, Akçay N, Dizman S, 2012. Radioactivity and heavy metal concentrations of some commercial fish species consumed in the Black Sea Region of Turkey. *Chemosphere* 87:356-61.
- Guérin N, Riopel R, Rao R, Kramer-Tremblay S, Dai X, 2017. An improved method for the rapid determination of ⁹⁰Sr in cow's milk. *J Environ Radioact* 175:115-9.
- Harley HN, 1979. Analysis of Foods for Radioactivity. Available from: <https://www.princeton.edu/~ota/disk3/1979/7907/790720.PDF>.
- Hatifi KH, Kadhim Muttaleb M, Hofdi Abass A, 2018. Measurement of radioactive radon gas concentrations of water in the schools for Abu-Gharaq. *JUBPAS* 26:174-80.
- Heldal HE, Volynkin AS, Skjerdal HK, Komperød M, Naghchbandi P, Hannisdal R, 2017. Radioactive substances in Norwegian farmed Atlantic salmon (*Salmo salar*). Available from: https://imr.brage.unit.no/imr-xmlui/bitstream/handle/11250/2480424/Rapport_24-2017_Lakseprosjekt_endelig.pdf?sequence=2&isAllowed=y.
- Iammarino M, dell'Oro D, Bortone N, Chiaravalle AE, 2015. Beta emitter radionuclides (⁹⁰Sr) contamination in animal feed: validation and application of a radiochemical method by ultra low level liquid scintillation counting. *Ital J Food Saf* 4:4531.
- Iammarino M, dell'Oro D, Bortone N, Mangiacotti M, Chiaravalle AE, 2016. Optimisation and validation of a multi-matrix ultrasensible radiochemical method for the determination of radiostrontium in solid foodstuffs by liquid scintillation counting. *Food Anal Method* 9:95-104.
- Idoeta R, Rozas S, Olondo C, Párraga A, Herranz M, 2018. ²²⁶Ra determination in complex samples using liquid scintillation counting. *J Radioanal Nucl Ch* 318:1773-84.
- Jibiri NN, Farai IP, Alausa SK, 2007. Activity concentrations of ²²⁶Ra, ²²⁸Th, and ⁴⁰K in different food crops from a high background radiation area in Bitsichi, Jos Plateau, Nigeria. *Radiat Environ Biophys* 46:53-9.
- Jobbágy V, Wätjen U, Meresova J, 2010. Current status of gross alpha/beta activity analysis in water samples: a short overview of methods. *J Radioanal Nucl Ch* 286:393-9.
- Khandaker MU, 2011. High purity germanium detector in gamma-ray spectrometry: High-Purity Germanium detector. *Int J Fundam Phys Sci* 1:42-6.
- Kudo K, Kobayashi K, 1979. Determination of trace elements and radioactive materials by substoichiometric isotope dilution analysis. *J Radioanal Chem* 53:163-72.
- L'Annunziata MF, 2012. Handbook of radioactivity analysis. Academic Press, Cambridge, MA, USA.
- L'Annunziata MF, Tarancón A, Bagán H, García JF, 2020. Liquid scintillation analysis: principles and practice. In: L'Annunziata MF, ed. Handbook of radioactivity analysis. Academic Press, Cambridge, MA, USA.
- Liu LY, Wang L, Jin P, Liu JL, Zhang XP, Chen L, Zhang JF, Ouyang XP, Liu A, Huang RH, 2017. The fabrication and characterization of Ni/4H-SiC schottky diode radiation detectors with a sensitive area of up to 4 cm². *Sensors (Basel)* 17:2334.
- Lou Y, Wan L, Ma Y, Li H, Meng Q, Kong Y, Zhu W, Wu D, Cui L, 2013. Survey on radioactive contamination in Beijing following the Japanese Fukushima nuclear accident. *J Radiol Prot* 33:661-8.
- Marchesani G, Trotta G, De Felice P, Bortone N, Damiano R, Nicolini M, Accettulli R, Chiaravalle AE, Iammarino M, 2022. Fast and sensitive radiochemical method for Sr-90 determination in food and feed by chromatographic extraction and liquid scintillation counting. *Food Anal Method* 15:1521-34.
- Mehra R, Bala P, 2014. Assessment of radiation hazards due to the concentration of natural radionuclides in the environment. *Environ Earth Sci* 71:901-9.
- Meli MA, Desideri D, Roselli C, Feduzi L, Benedetti C, 2016. Radioactivity in honey of the central Italy. *Food Chem* 202:349-55.
- Mercuri M, Pascual TNB, Mahmarian JJ, Shaw LJ, Rehani MM, Paez D, Einstein AJ, INCAPS Investigators Group, 2016. Comparison of radiation doses and best-practice use for myocardial perfusion imaging in US and non-US laboratories: findings from the IAEA (International Atomic Energy Agency) Nuclear Cardiology Protocols Study. *JAMA Intern Med* 176:266-9.
- Nasreddine L, Hwalla N, El Samad O, Leblanc JC, Hamzé M,

- Sibiril Y, Parent-Massin D, 2006. Dietary exposure to lead, cadmium, mercury and radionuclides of an adult urban population in Lebanon: a total diet study approach. *Food Addit Contam* 23:579-90.
- Nkuba LL, Mohammed NK, 2014. Determination of radioactivity in maize and mung beans grown in the neighborhood of Minjingu phosphate mine, Tanzania. *Tanz J Sci* 40:49-57.
- Orita M, Nakashima K, Taira Y, Fukuda T, Fukushima Y, Kudo T, Endo Y, Yamashita S, Takamura N, 2017. Radiocesium concentrations in wild mushrooms after the accident at the Fukushima Daiichi Nuclear power station: follow-up study in Kawauchi village. *Sci Rep* 7:6744.
- Pól J, Strohalm M, Havlíček V, Volný M, 2010. Molecular mass spectrometry imaging in biomedical and life science research. *Histochem Cell Biol* 134:423-43.
- Rahimi M, Abadi AAM, Jabbari Koopaei L, 2018. The Measurement of radon gas dissolved in groundwater and determination of annual effective absorbed dose of radon gas in Zarand city in 2016. *JRMUS* 16:1126-37.
- Ravikumar P, Somashekar RK, 2014. Determination of the radiation dose due to radon ingestion and inhalation. *Int J Environ Sci Technol* 11:493-508.
- Rožmarić M, Rogić M, Benedik L, Štok M, 2012. Natural radionuclides in bottled drinking waters produced in Croatia and their contribution to radiation dose. *Sci Total Environ* 437:53-60.
- Sahin L, Çetinkaya H, Murat Saç M, İçhedef M, 2013. Determination of radon and radium concentrations in drinking water samples around the city of Kutahya. *Radiat Prot Dosimetry* 155:474-82.
- Santos JS, Teixeira LSG, Dos Santos WNL, Lemos VA, Godoy JM, Ferreira SLC, 2010. Uranium determination using atomic spectrometric techniques: an overview. *Anal Chim Acta* 674:143-56.
- Skwarzec B, Strumińska DI, Boryło A, 2003. Radionuclides of ²¹⁰Po, ²³⁴U and ²³⁸U in drinking bottled mineral water in Poland. *J Radioanal Nucl Ch* 256:361-4.
- Snihs JO, 1983. Measurement Techniques for radon in mines, dwellings and the environment. Available from: https://inis.iaea.org/collection/NCLCollectionStore/_Public/15/043/15043323.pdf.
- Sujo LC, Cabrera MEM, Villalba L, Renteria Villalobos M, Torres Moye E, Garcia Leon M, García-Tenorio R, Mireles García F, Herrera Peraza EF, Sánchez Aroche D, 2004. Uranium-238 and thorium-232 series concentrations in soil, radon-222 indoor and drinking water concentrations and dose assessment in the city of Aldama, Chihuahua, Mexico. *J Environ Radioact* 77: 205-19.
- Tabar E, Yakut H, 2014. Determination of ²²⁶Ra concentration in bottled mineral water and assessment of effective doses, a survey in Turkey. *Int J Radiat Res* 12:193-201.
- Vosniakos FK, 2012. Radioactivity transfer in environment and food. Springer Berlin, Heidelberg, Germany.
- Weller A, 2017. Determination of strontium-90 in food concentrates from Japan. Available from: <https://www.irs.uni-hannover.de/fileadmin/irs/Arbeiten/Master/mastwell.pdf>.
- Yokota M, Watanabe T, Nomura H, Akimoto Y, Onchi H, 2017. Trend of radioactivity in fisheries products. *Rep Mar Ecol Res Inst* 22:27-36.
- Yoo WJ, Shin SH, Lee DE, Jang KW, Cho S, Lee B, 2015. Development of a small-sized, flexible, and insertable fiber-optic radiation sensor for gamma-ray spectroscopy. *Sensors* 15:21265-79.