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Monitoring of Natural Radioactivity in Drinking Water and Food with Emphasis on Alpha-Emitting Radionuclides

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Abstract

Alpha spectrometry is an indispensable technique in the radiology lab for the analysis of natural radionuclides. While the powerful ICP/MS is used more and more for the analysis of uranium and thorium, other radionuclides, such as ^{226}Ra, are difficult to analyze with this technique due to their very high specific activities. The following chapter is introduced by a description of the problems, which may occur when working in the ultra-trace level. A description of the commonly used extraction and enrichment techniques for alpha nuclides and a short survey of the commonly applied detection techniques are given. The main application of alpha spectrometry in our laboratory is the monitoring of tap and mineral waters. Besides water, some specific food categories, such as fish, seafood, spices or healing earths, are monitored for their content of natural radionuclides.

Keywords: alpha spectrometry, mineral water, seafood, spices, healing earth

1. Introduction

Uranium and thorium are radionuclides with long half-lives. Their abundances in the earth's crust are 12–13 ppm rsp. 2.5 ppm for thorium and uranium. They exist in minerals, such as pitch blend (uranium) or monazites (thorium).

From the earth's crust, these elements migrate to the groundwater layers. There, they can be transferred to surface waters. Therefore, the consummation of water is a main direct source for the ingestion of alpha nuclides. In addition, they are taken up by aquatic organisms (fish, mussels) and enriched in these organisms (e.g. Po-210 (^{210}Po) is enriched in the intestinal tract of mussels and fish). Alpha nuclides are enriched in farmland soils by irrigation and the application of phosphate fertilizers, which contain respectable amounts of uranium and thorium. The consummation of vegetables cultivated on such farmlands and the consummation of meat and meat products from these farms is another possibility for the intake of alpha nuclides.

There exist three main decay series of natural radionuclides starting from the radionuclides uranium-238 (^{238}U), thorium-232 (^{232}Th) and uranium-235 (^{235}U). The ^{238}U decay chain produces several radionuclides, which are of radiological concern. These are ^{238}U, ^{234}U, radium-226 (^{226}Ra), radon-222 (^{222}Rn), lead-210
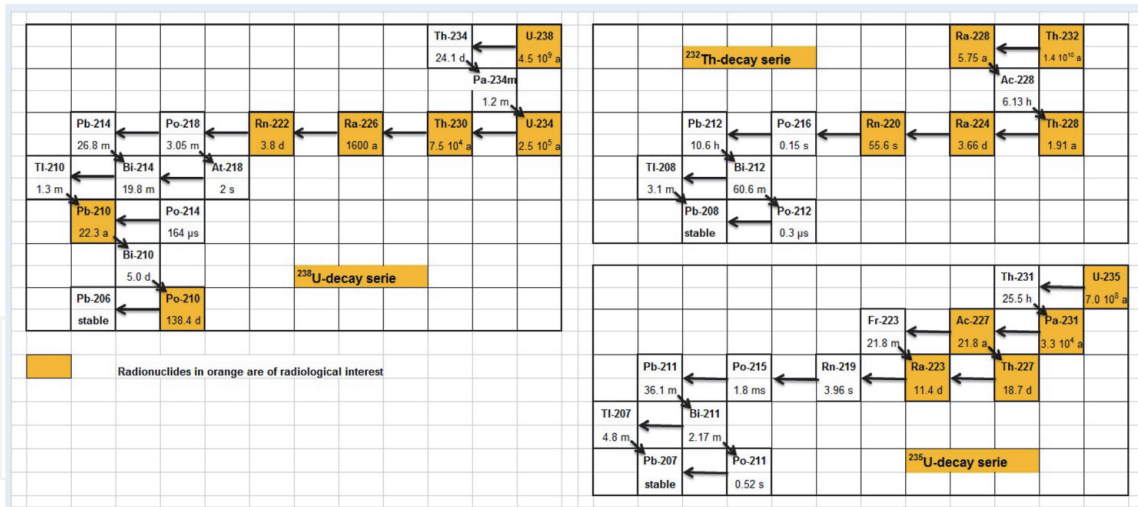
Figure 1.
Natural decay series of uranium and thorium. Alpha decays are symbolized with a horizontal arrow. The small diagonal arrows indicate a beta decay. In the nuclide box, the half-life is given. All data from [1].

(^{210}Pb) and ^{210}Po. The chain ends at the stable lead isotope ^{206}Pb. The ^{232}Th-decay chain comprises ^{232}Th, radium-228 (^{228}Ra), thorium-228 (^{228}Th), radium-224 (^{224}Ra) and radon-220 (^{220}Rn), which are of radiological relevance. The chain stops at the stable lead isotope ^{208}Pb. The Uranium-235 decay chain produces the following relevant radionuclides: ^{235}U, protactinium-231 (^{231}Pa), actinium-227 (^{227}Ac), thorium-227 (^{227}Th) and radium-223 (^{223}Ra). The chain stops at the stable lead isotope lead-207 (^{207}Pb). **Figure 1** shows the decay paths of the three decay series.

Radionuclides with longer half-lives, more than days, are of interest. Nevertheless, all radionuclides in the three decay chains are of radiological concern. The short-lived radionuclides cannot be considered separately. Therefore, the long-lived radionuclides and their short-lived daughter nuclides are assessed together. The conversion factor of a specific radionuclide considers the radiation of the following short-lived daughter nuclides.

Radon nuclides are radionuclides of the decay chains, which are gaseous. Therefore, they can be translocated by outgassing from the soil. That means, the radon nuclide and its follower nuclides are transported away from the soil or other environmental matrices that contain radium isotopes. Consequently, radon nuclides can be dislocated from its point of origin. ^{222}Rn can leave the soil due to its relatively long half-life of 3.8 days. One estimates that about 50% of the radon can leave the soil from a depth of 1 m [2]. In the air, the decay of the radon produces after several decay steps the longer-lived ^{210}Pb, ^{210}Bi and ^{210}Po, which are transported on dust particles through the atmosphere and may reach again farmland and surface waters. For the two other decay chains, this effect can be neglected. ^{219}Rn and ^{220}Rn are disintegrated before they have left the soil. In addition, the decay of these two radon species produces only short-lived radionuclides.

2. EU and Swiss legislation for radionuclides

Since 2018, in Swiss legislation most limit values for radionuclides for food were deleted after the adaption of the ordinances to the legislation of the European Union. The *Federal Ordinance on Contaminants and Constituents in Food* from 1994 was expired [3], and a new *Ordinance on Maximum Limits for Contaminants* was put into force in May 1, 2017 [4]. In this new ordinance, limit values for radionuclides in

food at so-called emergency situations are included. For "planed exposition situations" (this means normal situations without any fallout event), no limit values at all are defined. Fortunately, limit values for radiocesium, an important beta-emitter in fallout, are regulated in the *Ordinance on the Importation and the Placing on the Market of Food, which is contaminated following the Accident of the Nuclear Power Station of Chernobyl (Chernobyl Ordinance)* [5] and the *Ordinance on Food originating in or consigned from Japan (Fukushima Ordinance)* [6]. Unfortunately, the limit values for radiocesium are different in each ordinance.

In Switzerland, a better legal basis exists only for drinking water. The *Ordinance on Drinking Water and Water from Public Baths and Shower Facilities (TBDV)* includes a limit value for uranium and guide values for tritium and radon and the parameter *indicative dose* [7]. This ordinance is based on the *European Council Directive 2013/51/Euratom* [8]. Fortunately, this council directive prescribes some

Radionuclide		TBDV	Derived activity limits according to Euratom	WHO guidance level
natural radionuclides				
Lead-210 (^{210}Pb)	Bq/L		0.2	0.1
Polonium-210 (^{210}Po)	Bq/L		0.1	0.1
Radium-226 (^{226}Ra)	Bq/L		0.5	1
Radium-228 (^{228}Ra)	Bq/L		0.2	0.1
Radon (^{222}Rn)	Bq/L	100	100	—
Thorium-228 (^{228}Th)	Bq/L			1
Thorium-230 (^{230}Th)	Bq/L			1
Thorium-232 (^{232}Th)	Bq/L			1
Uranium-238 (^{238}U)	Bq/L		3.0	10
Uranium-234 (^{234}U)	Bq/L		2.8	1
Uranium*	µg/L	30	—	
Artificial radionuclides				
Americium-241 (^{241}Am)	Bq/L	0.1	0.7	1
Carbon-14 (^{14}C)	Bq/L		240	100
Cesium-134 (^{134}Cs)	Bq/L		7.2	10
Cesium-137 (^{137}Cs)	Bq/L		11	10
Cobalt-60 (^{60}Co)	Bq/L		40	100
Iodine-131 (^{131}I)	Bq/L	6.2	6.2	10
Plutonium $^{239 + 240}$Pu	Bq/L		0.6	1
Strontium-90 (^{90}Sr)	Bq/L	4.9	4.9	10
Tritium (^{3}H)	Bq/L	100	100	10,000
Indicative dose (ID)	mSv	0.1	0.1	

TBDV, Swiss ordinance on drinking water and water for public baths and shower facilities [7]; Euratom, council directive 2013/51/Euratom [8]; WHO [9].
uranium is calculated from the activity of ^{238}U.

Table 1.
Guidance levels for drinking water in EU and Switzerland compared to the WHO guidance levels.

more guidance limits, "derived activity limits", for artificial and natural radionuclides. Therefore, these activity limits are also applicable in Switzerland. **Table 1** summarizes the actual valid guidance and limit values for drinking water existing in Europe and Switzerland.

3. Collection and conservation of water samples

In this short chapter, important tips for a good analytical practice are given. Sample collection and storage can be the source of basic errors, which can no more be eliminated, even using a sophisticated analytical technique. At first, we must remember the concentration level we are working at.

3.1 Specific activity

When analyzing radio traces, one must keep in mind the very low chemical concentrations one is dealing with. Due to short half-lives (between hours and days), the specific activity of many species is very high. This means that very low, mostly non-weighable chemical concentrations correspond to measurable activities in the mBq range. Practical work with such traces requires the addition of inactive carriers. The following table illustrates this effect [10]. ^{232}Th and ^{238}U are exceptions. Both nuclides decay with half-lives of billion years. Therefore, common activities correspond to weighable chemical amounts (**Table 2**).

These very low concentrations must be kept in mind, when samples are collected and prepared for radio trace analyses. The problems, which can occur, are losses of the analytes or contaminations during collection, transport, conservation and preparation of samples.

3.2 Sample collection and storage

From natural waters, it is a major problem to obtain representative samples, e.g. out of a river. Trace element concentrations depend on depth, salinity and turbidity of a river or lake. Groundwater samples collected by using a pump should be taken some minutes after beginning of the pumping to avoid the collection of the standing water in the pipe. At this point, we cannot explicate profoundly the challenges for adequate sampling. We recommend consulting the relevant literature, e.g. [11–13].

Radionuclide	Half-life	Specific activity MBq/kg	Activity limits in tap water (Bq/L)*	Corresponding concentration μg/L (ppb)
^{222}Rn	3.8 d	5.7 10^{12}	100	2 10^{-11}
^{210}Po	138 d	1.66 10^{11}	0.10	6 10^{-10}
^{210}Pb	22.3 a	2.8 10^{9}	0.20	4 10^{-8}
^{228}Ra	5.75 a	1.0 10^{10}	0.20	1 10^{-7}
^{226}Ra	1600 a	3.4 10^{7}	0.50	3 10^{-6}
^{238}U	4.4 10^{9} a	12.5	3	8.1
^{232}Th	1.4 10^{10} a	4.06	1	24

Activity limits according to ordinance on drinking water and water for public baths and shower facilities (TBDV) [7].

Table 2.
Specific activities of some dose-relevant natural radionuclides.

Another important point is the choice of adequate sample containers. For metal trace analysis, no glass containers should be used. The active surface of glass bottles acts like an ion exchanger. SiOH groups are potent ligands for metal ions and may adsorb ions from the water sample. One possibility to omit these adsorptive effects is the conservation of the sample with mineral acid, such as nitric acid, hydrochloric acid or sulphuric acid. SiOH groups are then protonated, and the adsorption of cations is hindered. One can also add carrier ions to obtain the same effect. This is a commonly used technique in radiochemical analyses. Addition of stable isotopes of the analytes in higher concentrations prevents such adsorptive effects. Another possibility is to precipitate radio traces by adding carriers (co-precipitation). Another possibility is to catch and stabilize the analyte with a chelating agent, such as EDTA, to prevent absorptive losses. Samples may also be dried at 105°C for conservation or by freeze-drying, for example, of milk.

To overcome the possibility of such losses of analytes, we recommend using plastic bottles of polyethylene, PVC or Teflon, whenever possible. Pyrex (borosilicate glass) or soda glass containers should be avoided [13, 15, 16]. The containers should be soaked in diluted acid and rinsed thoroughly with distilled water before use. Before the sampling, the bottles are rinsed several times with the sample water [12].

It is also advisable to stabilize samples with the addition of conc. Nitric or hydrochloric acid, if the applied procedure does allow this (pH should be below 1). A stabilization is necessary when samples cannot be analyzed immediately (losses or transformation by bacteria, co-precipitation with suspended matter, etc. must be avoided) [11, 12, 16].

A special case is the sampling of the gaseous radon. Here, losses may occur when samples are collected under turbulences or bottles are not completely filled, leaving some headspace. Radon will outgas partially. Here, it is advisable to collect the water without turbulences in glass bottles under water, if possible. The bottles must be filled to the top without letting back any air bubbles. For sampling of water from taps or valves, a simple method is to let stream the water through a plastic tube connected to a funnel and to fill the bottle from the bottom up to the top without turbulences. Then, the bottle is closed with a glass stopper displacing the water in the bottleneck. Normally, the radon activities in water are in the Bq/L range, so loss effects by adsorption effects on glass walls may not be noticed. The bottles should be transported to the lab at low temperature, but a freezing of the sample should be avoided. The samples must be analyzed within a week due to the fast disintegration of the radon [11].

3.3 Quality of standards and chemicals

One should take special care to choose acids and other chemicals of high quality. They should be of sufficiently high purity. Concentrated mineral acid solutions (e.g. 65% nitric acid, 95% sulphuric acid) and other chemicals should be of the same quality used for ICP, AAS or XRF analysis. These are, for example, mineral acids of suprapur quality from Sigma-Aldrich, Merck, Roth, etc. For these products, specification data sheets with the declaration of minimum trace amounts of most metals are available. For every method, a careful check of the whole blank (chemicals used, demineralized water, glassware for sample preparation, etc.) is important and is a matter of course.

4. Sample preparation techniques

Alpha rays have a short reach due to its high mass and dual positive mass (two protons and two neutrons). Therefore, the matrix absorbs most alpha radiation. Alpha rays can leave the sample only from very thin surfaces. To detect alpha rays,

one must eliminate the matrix without losing the alpha nuclides. The following survey is not a complete review of the commonly used techniques. It is more focused on the applied procedures at the state laboratory of Basel City. This focus allows us to report from our long-time experience in alpha spectrometry.

4.1 Oxidation of the matrix

There are several common techniques for the preparation of samples. For non-volatile and thermal stable analytes, solid samples can be calcinated in an oven (dry ashing). For volatile analytes, e.g. [210]Po, where losses are possible at temperatures over 200°C or radiocesium (400°C), the matrix can be oxidized with mineral acid/peroxide in a microwave oven at moderate temperatures around 200°C.

In water samples, the analytes can be concentrated by evaporation or distillation of the water. Other procedures include direct evaporation of the sample on surfaces or by vacuum sublimation (e.g. Frisch-grid ionization chambers or gas proportional counters). The German DIN prescribes the evaporation of the water phase in the presence of barium as a carrier for the analysis of [226]Ra. After a second precipitation with sulphate as a cleanup to remove thorium and polonium, the analyte can be measured [17].

4.2 Liquid-liquid extraction

Liquid samples can be water samples or aqueous extraction solutions of solid samples. They may be extracted with nuclide-specific, extractive cocktails. Jack McDowell et al. have developed a set of extractants/fluors for different alpha nuclides [18]. Some special applications with the photon electron rejection alpha liquid spectrometry (PERALS) system are described in [19–21]. For the extraction of uranium with the extractor/scintillator URAEX, several investigations to optimize analytical procedures were published [22–24]. Véronneau et al. [25] investigated the extraction of polonium with different extractors/scintillators. Our preferred system is PPBO[1]/TOPO[2], commercially available as POLEX™. The PERALS system combined with a set of specific extractant/scintillator is a powerful analytical tool for the analysis of alpha nuclides (**Table 3**) [27].

Reagent	Extractable radionuclides	Literature data
ALPHAEX	U, Th, Pa, Hf, Zr, Pu (IV)	23, 28
POLEX	Specific for [210]Po, [237]Np	25, 29, 30
RADONS	Specific for [222]Rn	31
RADAEX	[226]Ra (and daughters)	20, 21, 29, 32
STRONEX	Specific for [90]Sr	33
THOREX	[228]Th, [230]Th, [232]Th and Zr, Hf, U, Eu, In and other nuclides	34, 35
URAEX	Specific for [234]U and [238]U	21, 23, 24

Table 3.
Extractant/fluor solutions for PERALS-α-spectrometry [26].

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[1] 2-(4′-biphenylyl)-6-phenylbenzoxazole
[2] Trioctylphosphine oxide

Many other extraction systems were published. For example, Leeuwen et al. published an overview on the selective extraction of radium with means of chelators [36, 37].

4.3 Adsorptive surfaces

The use of some specific active surfaces, which are suitable for the adsorption of radionuclides from aqueous solutions and are commercially available, was published.

Such selective phases can be made of pure metals, such as copper, nickel or silver. ^{210}Po will auto adsorb on such a surface in an acidic, reductive milieu using ascorbic acid or hydroxylamine [38–41]. For recovery control of the adsorption process, polonium-208 (^{208}Po) or polonium-209 (^{209}Po) can be added as tracers.

Manganese dioxide-coated surfaces adsorb radium nuclides selectively. Based on the work of Moore and Reid [42], Glöbel and Berlich [43] and Surbeck [44], robust procedures for the analysis of radium in water were developed by Eikenberg et al. and Surbeck [45, 46]. Ra disks are commercially available from nucfilm [47].

For uranium, there also exists a selective surface based on diphonix resin supported on polycarbonate [48]. Diphonix-coated disks are commercially available from nucfilm [47]. They work well below activities of 1 Bq/L because of the restricted load capacity. For example, Surbeck suggests analyzing for uranium only after the elimination of radium by adsorption with a MnO_2 disk. Otherwise overloading effects with radiumnuclides may be noticed [49].

Alpha nuclides in a liquid milieu can be adsorbed electrolytically onto stainless steel disks (electroplating, electrodeposition). The *Mitchell method* describes the deposition of actinides (uranium, americium, polonium) in a HCl milieu [50]. The *Talvitie method* is used for the deposition of actinides (uranium, thorium) in the presence of a HSO_4^-/SO_4^{2-} milieu [51]. Many applications were published for uranium, thorium, radium, plutonium and other alpha nuclides [52, 53]. In our laboratory we have experiences in analyzing uranium and thorium in acid extract solutions of spices [54]. Our method is based on working sheets from the Paul Scherrer Institute (PSI), which describes the production of a homemade electrodeposition unit and the application for the enrichment of uranium or thorium on stainless steel planchettes [51, 55–58]. Frindik et al. give a method for the determination of many alpha nuclides, including such as plutonium and americium [59].

4.3.1 Micro-precipitation

For radium, micro-precipitation processes based on manganese dioxide are well used. Many alpha nuclides, such as americium or plutonium, are analyzable. To equalize losses of analytes in the precipitation step, the use of radioactive tracers (internal standards) is important [60]. Thorium, uranium, plutonium, americium and curium species are analyzable [61, 62].

4.3.2 Filtration

Radionuclides are collected on impregnated filters more or less selectively. Best examples are radium-specific filters from 3 M Empore (Radium Rad disks). ^{228}Ra can be analyzed indirectly via its short-lived daughter ^{228}Ac, which is built on the filter surface by the disintegration of the enriched ^{228}Ra. This is a very interesting approach for the analysis of the dose-relevant ^{228}Ra [63, 64].

5. Measurement equipment

For a general survey, we recommend the *Handbook of Radioactivity Analyses* [65]. Another somewhat older standard book is *Radiation Detection and Measurement* from Knoll [66]. In this chapter, we describe the most used analytical techniques for alpha nuclides as we know.

5.1 Alpha particle spectrometry

Passivated implanted planar silicon detectors (PIPS) or silicon barrier detectors are widely used in analyzing alpha nuclides. It is necessary to obtain thin, homogenous alpha sources. Thick sources show broad peaks due to some degradation cause by self-adsorption. High-resolution alpha spectrometry needs thin sources. For this purpose, stainless steel plates or adsorptive surfaces are ideal. Very thin micro precipitates are also suitable. Most alpha nuclides are energy resolved, with one exception. The alpha energies of plutonium-239 (^{239}Pu) and plutonium-240 (^{240}Pu) differ only 10 keV. They may not be resolved in the alpha spectrum and therefore often given as the activity sum of both nuclides.

Detector systems are available from Ortec [67] and Mirion (former Canberra) [68]. They are available as multichamber systems to handle the long counting times (e.g. 24 hours) in the daily routine. In our laboratory, we use PIPS detectors to detect alpha nuclides of polonium, radium, uranium and thorium (**Figure 2**).

5.2 Liquid alpha scintillation techniques

Liquid scintillation techniques (LSC) with an alpha/beta discrimination possibility are often used [69]. An example is the analyses of radon. Very sensitive analyses of radon in water samples are possible, when the large beta background of the radon daughters ^{214}Bi and ^{214}Pb is discriminated. The consecutive alpha decays of ^{222}Rn, ^{218}Po and ^{214}Po are counted (cross alpha counting) [70]. Three main procedures for the analysis of radon with LSC are possible. The "direct" method consists in mixing of the water with a water immiscible cocktail. The mixture is then counted with LSC. In the indirect method, radon is transferred from the water into the cocktail phase and then analyzed. The third method uses a purging of the radon into a cocktail phase. The simplest and most precise method is the direct method, because only one step in the sample preparation is necessary. Low detection limits are achievable with a small effort in sample preparation (detection limit of about 0.2 Bq/L).

A special alpha scintillation technique is commercially available from ORDELA [26]. The PERALS called alpha spectrometric technique is based on the following procedure. The analyte is extracted with a selective extracting agent, which contains a specific fluor. With the help of the fluor, α-, β- and γ-rays are converted into photons. The β- and γ-photons of the extract solution are discriminated by the use of a pulse shape discriminator. The discrimination is based on the longer relaxation times of alpha decays. The discriminator eliminates the fast-relaxing β- and γ-induced photons. As resolution of the emissions is much poorer than PIPS detectors, nuclide-specific extractions systems must be used. Methods are available for the analysis of polonium, radon, radium, uranium, thorium and other alpha nuclides [18], see 4.2 (**Figure 2**).

5.3 Gas proportional counting

Gas proportional counting is a suitable analyzing technique when equipped with the possibility to separate alpha from beta rays. Loaded planchettes or disks and thin
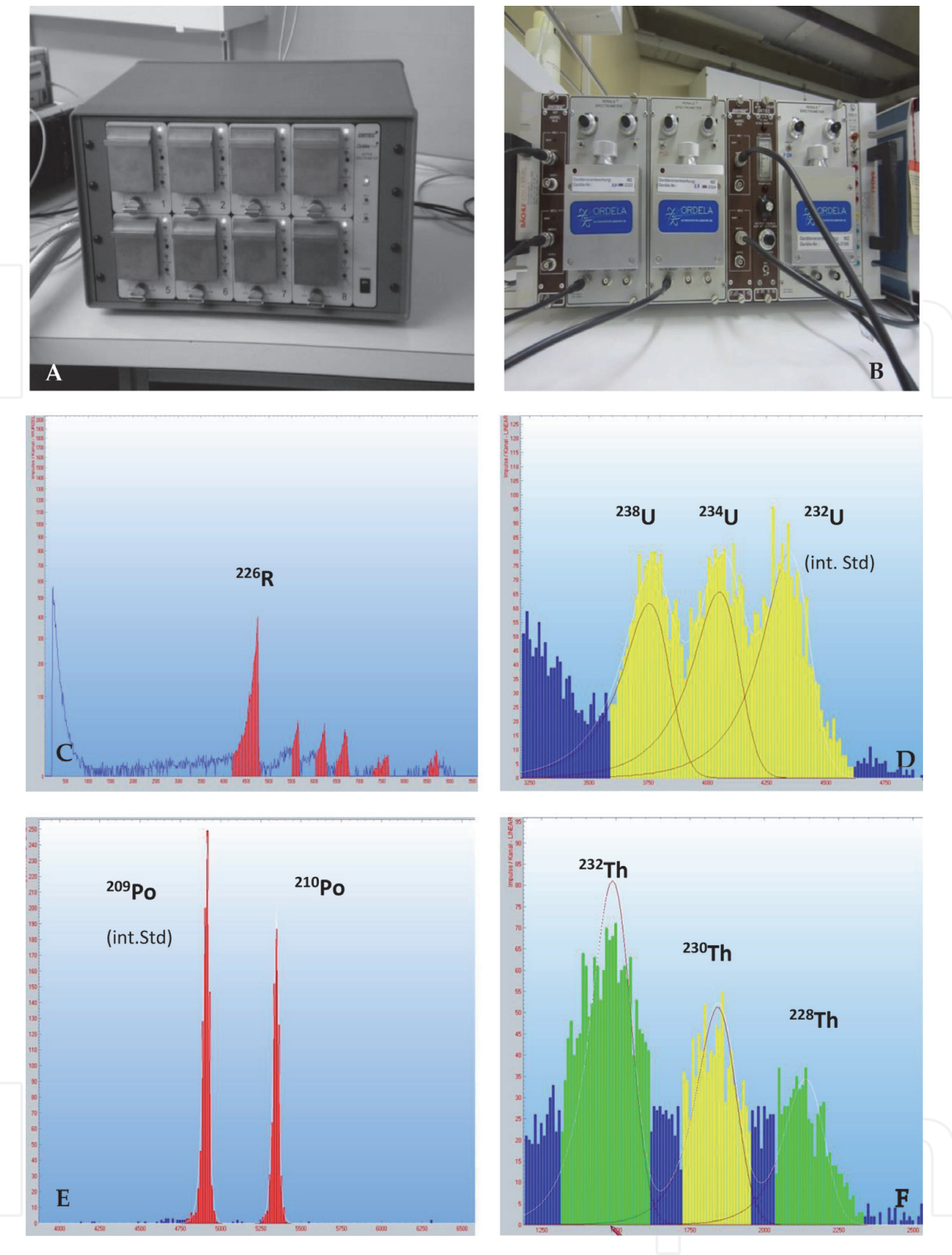
Figure 2.
Common equipment for alpha spectrometry. A, PIPS alpha spectrometer with eight counting cells; B, PERALS alpha liquid scintillation counters; C, alpha spectrum of radium in mineral water; D, PERALS spectrum of uranium in san Pellegrino mineral water; E, PIPS alpha spectrum of polonium in a mineral water; F, PERALS spectrum of thorium in a mineral water.

precipitates can be counted. Methods exist for polonium and other alpha emitters. It is also possible to extract and analyze alpha and beta nuclides, e.g. ^{210}Pb and ^{210}Po.

5.4 Track detectors

Track detectors are used since many years for the measurement of ^{222}Rn in air. The tracks on an exposed foil produced by alpha particles are counted with a microscope after chemical etching of the foil.

5.5 Gamma-ray spectrometry

Gamma-ray spectrometry is only suitable for the analyses of alpha nuclides in samples, where the analytes are present in higher activities (soil, sediments, etc.). Uranium, for example, is only detectable via its daughter nuclides (e.g. ^{234}Th for ^{238}U). Therefore, it is important to render data plausible according to the individual decay time within the decay chains. Radionuclides, which are in secular equilibrium, should show equal activities. Interferences may pretend excessive activities.

A second challenge is the weak photon emissions of alpha nuclides. In addition, these emissions are in the lower keV range, and the correlation to a specific radionuclide is often doubtful because of interferences. ^{232}Th is such a radionuclide with a main emission line at 63.8 keV. It can easily be mixed with ^{234}Th (63.3 keV). Such lines of weak intensity need counting times of days to get reasonable signals [71]. In drinking water analysis, there are two important radionuclides, ^{228}Ra and ^{210}Pb, which are beta emitters and cause dominant dose contributions. ^{228}Ra can be analyzed indirectly via its short-lived beta daughter ^{228}Ac. ^{210}Pb shows only a very weak line at 46.5 keV. The analysis is only suitable with a germanium detector equipped with a beryllium or carbon window that allows the transmission of low-energy lines. For both radionuclides, counting times are days or weeks.

^{226}Ra can be analyzed directly with its emission line at 186.21 keV (3.56% emission probability) but which interferes with a more intensive gamma line of ^{235}U (185.72 keV, 57.2%). Here, it is advisable to quantify ^{226}Ra via its daughter nuclides (^{214}Bi and ^{214}Pb), after the radioactive equilibrium is established. ^{228}Th and ^{224}Ra are alpha nuclides of the ^{232}Th decay chain. Their activities are calculated via the daughter nuclides (^{212}Pb, ^{212}Bi). ^{238}U may be analyzed via its daughter ^{234}Th. As mentioned before, these radionuclides are only quantifiable in samples, which show relatively high activities (e.g. soil samples) [71]. In **Table 4**, radionuclide decay energies and common detection methods are listed. Passivated implanted planar silicon detectors (PIPS), liquid scintillation alpha/beta-counting and PERALS are commonly used analytical methods.

6. Applications

In this chapter, some few applications of alpha spectrometry for the examination of drinking water and food samples are described. In our laboratory, the main application of alpha spectrometry is for the routine control of water samples, e.g. tap water or mineral waters. Fish and seafood, spices and healing earths are food categories, which may have incorporated relevant quantities of alpha nuclides.

6.1 Monitoring of mineral waters from the Swiss market

Natural mineral water should be water of good microbiological quality. It is collected in groundwater layers or rock formations. This water reaches the surface by one or several naturally or artificially built exit points and can be collected there. Mineral waters are classified according to their mineral contents (from very low mineral content to high mineral content) or specified for a dominant constituent (e.g. rich in magnesium, iron, fluoride). Another classification respects the content of carbonic acid (non-carbonated water, sparkling water, etc.). In all cases, minimum or maximum concentrations are defined in the Swiss Ordinance on Beverages [72] and other ordinances. Radionuclide activities in mineral waters may vary in a wide range and are caused by complex solubility and transport processes in the aquifer. The hydrogeological conditions, the uranium and thorium content of the

Radionuclide	Half-life	Alpha energies (MeV), % branching	Common methods comments
^{210}Pb	22.3 y	beta	LSC Gas prop. Counter: indirect via ^{210}Bi Gamma spectrometry: weak line at 46.5 keV
^{210}Po	138.4 d	5.407,5 (100)	LSC: PERALS PIPS: Ag and Cu disk
^{231}Pa	3.28 10^4 y	4.762,5 (1,8) 4.795,4 (1,2) 4.819,8 (8,4) 4.939,1 (1,4) 5.023,0 (2,9)	PIPS: electrodeposition Gamma spectrometry
^{224}Ra	3.66 d	5.547,9 (5,3) 5.788,9 (94,7)	LSC: PERALS PIPS: electrodeposition PIPS: MnO$_2$ disk Gamma spectrometry
^{226}Ra	1600 y	4.684,0 (5,95) 4.870,5 (94,0)	LSC: PERALS PIPS: electrodeposition PIPS: MnO$_2$ disk Gamma spectrometry: interfering line at 86 keV
^{228}Ra	5.75 y	beta	LSC Gamma spectrometry: via ^{228}Ac
^{220}Rn	55.6 s	6.404,7 (100)	PIPS: PERALS
^{222}Rn	3.83 d	5.590,3 (100)	LSC: several specific methods
^{228}Th	1.91 y	5.520 (73,4) 5.436 (26,0)	LSC: PERALS B: electrodeposition
^{230}Th	7.54 10^4 y	4.620,5 (23,4) 4.687,0 (76,3)	LSC: PERALS PIPS: electrodeposition
^{232}Th	1.41 10^{10} y	4017,8 (21,0) 4081,6 (78,9)	LSC: PERALS PIPS: electrodeposition
^{234}U	2.45 10^5 y	4.804,5 (28,4) 4.857,6 (71,4)	LSC: PERALS and others PIPS Gamma spectrometry (indirect)
^{235}U	7.04 10^8 y	4.441,7 (18,8) 4.474,0 (7,25)	LSC: PERALS and others Gamma spectrometry
^{238}U	4.47 10^9 y	4.220,2 (22,3) 4.269,7 (77,5)	LSC: PERALS and others PIPS Gamma spectrometry (indirect)
^{239}Pu	2.41 10^4 y	5.192,8 (11,9) 5.231,5 (17,1) 5.244,5 (70,8)	LSC: PERALS PIPS
^{240}Pu	6561 y	5.210,5 (27,2) 5.255,8 (72,7)	PIPS
^{241}Am	432.4 y	5.479,3 (1,7) 5.534,9 (13,2) 5.578,3 (84,5)	LSC: PERALS PIPS Gamma spectrometry
^{244}Cm	18.1 y	5.858,9 (23,3) 5.901,7 (76,7)	LSC: PERALS PIPS

Taken from: [1].

Table 4.
Radionuclide decay energies and common detection methods.

rock formations, solubility behaviour and other chemical characteristics of the radionuclides are crucial for their presence in the water phase.

Besides tap water, mineral waters are the most consumed beverage in Switzerland. Nearly one billion liters are consumed yearly. Today, about 57% of the mineral waters are from Swiss production. 43% are originated from European countries, such as Italy, France, Germany and others [73]. The consummation of such high quantities of water requires rigorous monitoring. However, the law prescribes the declaration of the mineral contents only. Besides a microbiological survey, the focus is on contaminants from agriculture, industry and other productions. In past years, contaminations were detected sporadically. For example, traces of benzene in a French mineral water, *E. coli* bacteria in a mineral water infiltrated by contaminated seawater or high amounts of radium or uranium in French and Portuguese products gave reason for small scandals and violations of the law. Increased uranium activities in groundwater, originating from the intensive use of phosphate fertilizers, gave reason for the establishing of a monitoring programme for drinking water plants in Germany.

Natural and artificial radio contaminants were first monitored systematically in the USA and Germany [74].[3] In Switzerland, first investigations of mineral waters are from 1990. The Federal Office of Public Health mandated the Swiss Paul Scherrer Institute to analyze mineral waters available from the Swiss market [75]. In 2006 and 2007, the state laboratory of Basel City analyzed mineral waters in collaboration with the Office for food safety and veterinary affairs of the state of Basel-Country with the focus on uranium, tritium, radon and heavy metals [76, 77].

In 2018, our laboratory analyzed 46 mineral waters from the Swiss market. It was a complete investigation of natural and artificial radionuclides according to the TBDV [7]. Since 2018, the food law has changed. The former Ordinance of Food Contaminants and Constituents (FIV) was invalidated [3]. Therefore, no more limits exist for radionuclides in mineral waters. Consequently, it was necessary to validate the results according to the guide and limit values of the Swiss drinking water ordinance (**Table 1**).

Mineral water samples were collected at stores of the city of Basel. 21 products were Swiss mineral waters, followed by 12 mineral waters from Italy, 3 from Serbia and 2 each from Germany and France. Other countries were Fiji islands, Spain, Portugal, Norway, Kosovo and Croatia. The samples were analyzed by using the following sample preparation/measurement scheme (**Table 5**).

The aim of such investigations is always to estimate internal doses by consumption of the analyzed food category. Therefore, all dose-relevant radionuclides must be analyzed. These are natural radionuclides and anthropogenic radionuclides, such as ^3H, ^{40}K or ^{241}Am. According to TBDV, the total dose is the sum of the individual doses of all natural radionuclides, with the exception of potassium-40 (^{40}K), tritium (^3H) and ^{222}Rn with its short-lived daughters. So, uranium, radium, polonium, thorium and lead nuclides are the main nuclides which contribute to the dose. The doses due to the ingestion of individual radionuclides were calculated as follows:

$$D_i = c_i * e_{ing,i} * U, \tag{1}$$

with

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[3] Federal ministry for the environmental conservation, construction and nuclear security (2017). Guidelines on compliance with the requirements of the Drinking Water Ordinance in the testing and evaluation of radioactive substances in drinking water. Drinking water is tested for its concentrations of radioactive substances nationwide.

Radionuclide	Sample preparation	Sample volume	Analytical technique	Quantification limit
^{234}U, ^{238}U	Extraction with 5 mL of URAEX	500 mL	PERALS-α-spectrometry	4 mBq/L
^{224}Ra, ^{226}Ra	Adsorption on MnO$_2$ disk	200 mL	PIPS-α-spectrometry	2 mBq/L
^{228}Ra	No sample preparation	1000 mL	γ-spectrometry (via ^{228}Ac)	50 mBq/L
^{210}Po	Adsorption on Ag disk	100 mL	PIPS-α-spectrometry	5 mBq/L
^{210}Pb	Adsorption on Ni disk	200 mL	b-gas proportional counter	50 mBq/L
^{228}Th, ^{230}Th, ^{232}Th	Extraction with 5 mL of THOREX	500 mL	PERALS-α-spectrometry	2 mBq/L
^{222}Rn	1:1 mix with Maxilight cocktail	10 mL	α-counting with LSC	0.4 Bq/L
^{3}H	Mix with 12 mL of Ultima Gold LLT cocktail	8 mL	β-counting with LSC	2 Bq/L
^{90}Sr	Extraction with 8 ml of STRONEX	1000 mL	β-counting with LSC	0.05 Bq/L
^{60}Co, ^{131}I, ^{134}Cs ^{137}Cs, ^{241}Am	No sample preparation	1000 mL	γ-spectrometry	0.05–0.1 Bq/L

Table 5.
Sample preparation and analyzing techniques used for the investigation of mineral waters at the state laboratory of Basel City.

c_i: Activity concentration of the radionuclide i

$e_{ing,i}$: Ingestion factor of radionuclide i [78]

U: Consummation rate (720 L for adult persons and year)

The sum of these individual doses is defined as *indicative dose*. The dose coefficients are listed in **Table 6**.

Results of the monitoring of mineral water in Switzerland in 2018 are presented in **Table 7**.

Uranium was detectable in 45 of 46 mineral waters. The mean activities were 0.02 ± 0.02 rsp. 0.02 ± 0.03 for ^{234}U and ^{238}U. The corresponding chemical concentration was 1.9 ± 2.0 μg U/L, fulfilling the limit value of 30 μg/L. The ratio of ^{238}U/^{234}U of 1.04 shows an undisturbed equilibrium between the two uranium nuclides. The Italian mineral waters *San Pellegrino* and *Varanina* contained 183 and 269 mBq/L of both uranium nuclides, corresponding to 7.0 rsp. 10.6 μg/L.

^{228}Ra is the dominant radionuclide of the radium species. Eleven samples showed activities of ^{228}Ra with a mean of 0.07 ± 0.05 Bq/L. ^{224}Ra and ^{226}Ra were only found in traces (mean 0.04 Bq/L for ^{226}Ra) with one exception. *Pedras Salgadas*, a mineral water from Portugal, showed an activity of 1.4 Bq/L of ^{226}Ra. Here, the guide value of 0.5 Bq/l was violated. Before 2017, this mineral water was banned from the Swiss market due to the then existing limit value of 1 Bq/L for liquid food. ^{224}Ra was only detected in two samples (< 0.002–0.10 Bq/L).

^{210}Pb and ^{210}Po are built at the end of the decay chain of ^{238}U. ^{210}Pb has a relatively long half-life of 22.3 years and is the mother nuclide of ^{210}Bi and ^{210}Po. It is a beta nuclide and commits to the indicative dose. ^{210}Po is an alpha nuclide with a high alpha energy of 5.3 MeV and a half-life of 138 days. Therefore, it is also dose-relevant. Fourteen mineral water samples showed a measurable activity of ^{210}Pb.

Radionuclide	Infants (1–2 years)	Children > 10 years	Adult persons
Americium-241 (^{241}Am)	0.37	0.22	0.20
Lead-210 (^{210}Pb)	3.6	1.9	0.69
Cesium-134 (^{134}Cs)	0.016	0.014	0.019
Cesium-137 (^{137}Cs)	0.012	0.010	0.013
Iodine-131 (^{131}I)	0.180	0.052	0.022
Cobalt-60 (^{60}Co)	0.027	0.011	0.003
Polonium-210 (^{210}Po)	8.8	2.6	1.2
Radium-224 (^{224}Ra)	0.66	0.26	0.065
Radium-226 (^{226}Ra)	0.96	0.80	0.28
Radium-228 (^{228}Ra)	1.70	1.70	1.70
Radon (^{222}Rn)	0.02	0.02	0.01
Strontium-90 (^{90}Sr)	0.073	0.06	0.028
Thorium-228 (^{228}Th)	0.37	0.14	0.072
Thorium-230 (^{230}Th)	0.41	0.24	0.21
Thorium-232 (^{232}Th)	0.45	0.29	0.23
Tritium (^{3}H)	4.8E − 11	2.3E − 11	1.8E − 11
Strontium-90 (^{90}Sr)	0.073	0.06	0.028
Uranium-238 (^{238}U)	0.12	0.068	0.045
Uranium-234 (^{234}U)	0.13	0.074	0.049

Table 6.
Ingestion factors of the Swiss radiological protection ordinance [78] based on the ICRP concept. All data in μSv/Bq.

Radionuclide		Mean ± s.d.	Min	Max	n
^{234}U	Bq/L	0.02 ± 0.02	0.002	0.14	45
^{238}U	Bq/L	0.02 ± 0.03	0.002	0.13	45
^{nat}U	μg/L	1.9 ± 2.0	0.1	10.6	45
^{224}Ra	Bq/L	0.003 ± 0.02	0.002	0.11	2
^{226}Ra	Bq/L	0.04 ± 0.21	0.002	1.4	23
^{228}Ra	Bq/L	0.07 ± 0.05	0.05	0.44	11
^{222}Rn	Bq/L	1.3 ± 1.4	0.4	4.4	27
^{210}Po	Bq/L	0.04 ± 0.06	0.01	0.23	14
^{210}Pb	Bq/L	0.10 ± 0.06	0.05	0.27	14
^{228}Th	Bq/L	0.003 ± 0.002	0.002	0.01	22
^{230}Th	Bq/L	0.002 ± 0.002	0.002	0.01	12
^{232}Th	Bq/L	0.003 ± 0.004	0.002	0.02	20

s.d., standard deviation; n, number of positive samples from a total of 46 samples.

Table 7.
Results from the monitoring of mineral water in Switzerland.

The mean was 0.10 ± 0.06 Bq/L. The *Pedras Salgadas* mineral water contained 0.3 Bq/L and therefore was over the guidance value of 0.2 Bq/L. Fourteen mineral water samples contained [210]Po in low activities (mean: 0.04 ± 0.06 Bq/L). In an Italian mineral water, the guidance limit of 0.1 Bq/L was overridden (0.23 Bq/L).

Thorium nuclides were found in low activities but in almost all mineral waters. The mean sum of the three nuclides was 0.01 Bq/L. These low activities are explained with the insolubility of thorium in water. Thorium species bind mainly on particles or co-precipitate with minerals and are therefore removed from the water phase.

[222]Rn was found in 27 of the 46 samples with a low, mean activity of 1.3 ± 1.4 Bq/L (0.4–4.4 Bq/L). Most of the radon is lost during the production and transport of the mineral waters, except the Portuguese *Pedras Salgadas*, where radon is constantly produced by the disintegration of [226]Ra.

Artificial radionuclides, such as ^{3}H, ^{90}Sr and ^{137}Cs, were detectable in traces. Eight samples contained 0.01 ± 0.01 Bq/L [137]Cs. One sample showed traces of [90]Sr (0.05 Bq/L). [3]H was detectable in only two samples 1.2 ± 0.9 Bq/L. Radiocesium and radiostrontium are the main components of the global fallout and fallout from NPP accidents (Chernobyl). Their migration into the soil is a slow process. There-fore, disintegration may be fast enough to reduce the activities to amounts below the detection limits in the groundwater phase.

The calculated internal doses of the mineral waters (indicative doses) were 0.06 ± 0.60 mSv/a. Most samples fulfilled the guidance value of 0.1 mSv/a. The indicative dose of five mineral waters was over the guidance value. The highest dose was calculated for the Portuguese product *Pedras Salgadas.* We calculated a dose of 0.46 mSv/a for adult persons. Four other mineral waters showed doses between 0.12 and 0.27 mSv/a. Elevated activities of [226]Ra, [228]Ra and ^{210}Pb were the cause for these higher doses (**Figure 3**).

6.2 Analysis of healing earths

Siliceous earths are widely used in the food industry as a food supplement. They are deposits of the silica shells of diatoms (main constituent of marine phytoplankton). These layers are extracted in mines. Siliceous earths incorporate foreign atoms in the crystal lattice, such as radionuclides of the natural decay series
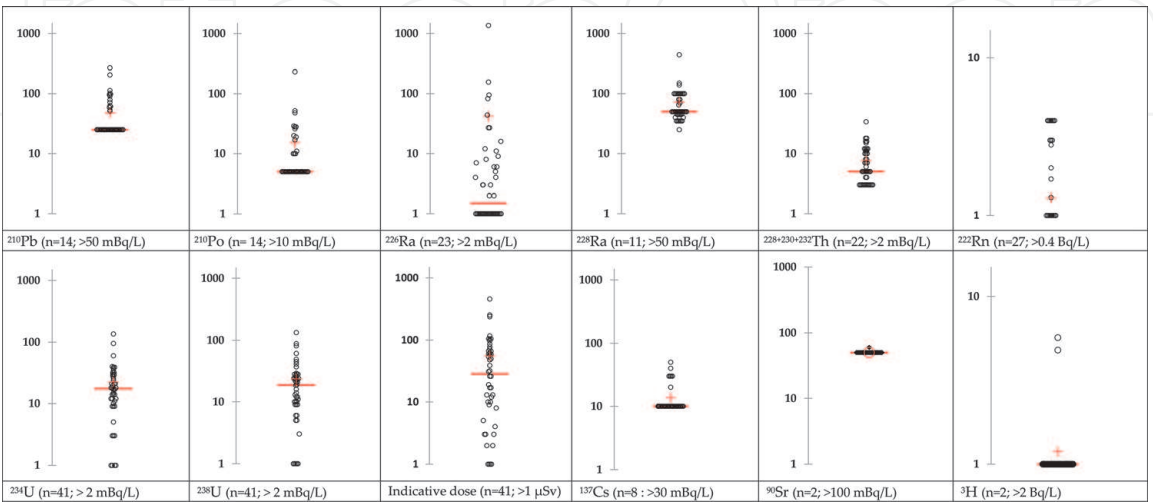

Figure 3.
Results from the investigation of mineral waters on the Swiss market 2018. All data in mBq/L, except indicative dose (μSv). n: Number of samples with activities over the detection limit (given as ">x mBq/L").

Radionuclide	Mean ± s.d.	Samples[*]	Min	Max
^{224}Ra	25.3 ± 21.5	30/32	1.7	63
^{226}Ra	42.1 ± 33.6	30/32	2.9	133
^{228}Ra	27.2 ± 23.5	30/32	2.0	67
^{234}U	56.9 ± 29.1	18/26	15.4	115
^{235}U	4.0 ± 2.1	07/26	1.5	6.9
^{238}U	75.4 ± 56.7	22/26	6.5	227
^{227}Th	2.6 ± 1.8	17/29	0.7	6.0
^{210}Pb	183 ± 152	6/24	49	428
^{210}Po	11.0 ± 9.0	22/24	2.0	42

s.d.: standard deviation.
[*]*Number of positive samples/total of samples.*
All data in Bq/kg dry weight and from [78, 79].

Table 8.
Investigation of healing earths on the Swiss market.

of uranium and thorium. In 2008, the state laboratory of Basel City analyzed siliceous earth products on the Swiss market with α- and γ-spectrometry. In two products, the limit values for ^{226}Ra and ^{210}Po were exceeded (>50 Bq/kg). Furthermore, the annual dose by regular consummation of one product from California, USA, reached 0.5 mSv, half of the permitted yearly dose (1 mSv). Consequently, this product was withdrawn from the Swiss market [79]. In 2010, a second inspection of the healing earths on the Swiss market showed that two products from German production slightly exceeded the limit values of ^{226}Ra and ^{228}Ra. The calculated, annual doses of these products when regularly consumed reached 0.1 mSv/year [80]. Finally, we noted that healing earths can lead to doses up to 0.5 mSv. At last, materials based on silica for industrial use (e.g. as adsorbents or filter media in a chemical laboratories) can lead to the contamination of the environment when such materials are disposed or burnt (**Table 8**).

6.3 Radiological investigation of spices

Radionuclides from the uranium and thorium series may be enriched in plants and therefore also in spices. Until 2017, Swiss legislation included limit values for natural radionuclides in spices. For radionuclides of the group I (^{224}Ra, ^{228}Th, ^{234}U, ^{235}U, ^{238}U), a cumulative limit value of 500 Bq/kg was given in the Ordinance of Contaminants and Constituents in Food [3]. For group II (^{210}Pb, ^{210}Po, ^{226}Ra, ^{228}Ra, ^{230}Th, ^{232}Th and ^{231}Pa), a cumulative limit value of 50 Bq/kg was defined.

A total of 50 spice samples from Spain, South Africa, Asian countries, Turkey and India were collected on the Swiss market and analyzed with gamma-ray spectrometry. After reaching equilibrium between ^{226}Ra and ^{222}Rn (20 days), ^{226}Ra was determined via its daughter nuclides ^{214}Bi and ^{214}Pb. ^{224}Ra is in equilibrium with its daughters ^{212}Pb and ^{212}Bi. ^{228}Ra was analyzed via its daughter nuclide ^{228}Ac. The thorium nuclides ^{228}Th, ^{230}Th and ^{232}Th were analyzed with alpha spectrometry using PIPS detectors. The sources were prepared by deposition of the thorium species from acid microwave extracts onto steel disks with electrodeposition.

Pepper samples contained increased amounts of the radium and thorium nuclides. The cumulated activity of the radionuclides from group I, especially ^{224}Ra,

Radionuclide	Mean ± s.d.	Samples[*]	Min	Max
Pepper				
^{224}Ra	8.2 ± 6.0	5/8	2.0	16
^{226}Ra	3.4 ± 0.89	5/8	2.0	4.0
^{228}Ra	9.7 ± 7.1	6/8	3.3	23
^{228}Th	0.9 ± 0.37	3/3	0.51	1.25
^{230}Th	3.0 ± 0.72	1/3	3.0	3.0
^{232}Th	3.0 ± 3.1	2/3	0.77	5.3
Paprika				
^{224}Ra	2.0 ± 0.5	1/12	2.0	2.0
^{226}Ra	2.1 ± 0.61	3/12	1.6	2.8
^{228}Ra	<2			
^{228}Th	1.1 ± 0.85	5/5	0.27	2.4
^{230}Th	0.3 ± 0.08	1/5	0.33	0.33
^{232}Th	1.1 ± 0.79	5/5	0.37	2.5
Curries				
^{224}Ra	<2			
^{226}Ra	3.1 ± 1.7	3/11	2.0	5.0
^{228}Ra	3.3 ± 1.7	1/11	3.3	3.3
^{228}Th	1.3 ± 0.8	6/11	0.32	2.5
^{230}Th	0.4 ± 0.17	3/11	0.21	0.52
^{232}Th	1.0 ± 0.52	6/11	0.23	1.8

s.d.: Standard deviation.
[*]*Number of positive samples/total of samples*
All data from [53, 80].

Table 9.
Investigation of spices on the Swiss market.

reached 10–32% of the limit value (500 Bq/kg). The cumulated activity of the radionuclides of group II, especially ^{226}Ra and ^{228}Ra, reached 10–26% of the limit value (50 Bq/kg) (**Table 9**). The consummation of 100 g of pepper in a year leads to a dose of about 1–2 μSv [54, 81].

6.4 Analysis of seafood and fish

Natural radionuclides cause the main radio contamination of fish and seafood. Mussels and molluscs may enrich ^{210}Po in the intestinal tract, whereas the mother nuclide ^{210}Pb is not enriched [82]. Activity concentrations of ^{210}Po range from 20 to 100 Bq/kg. In fish, the ^{210}Po level is much lower (1–20 Bq/kg) [83].

In 1998, we investigated the contamination of ^{210}Po in sea fish and mussels. The ^{210}Po was extracted with acid and microwaves at temperatures below 200°C. Then, the analytes (^{210}Po and internal standard ^{209}Po) were adsorbed onto silver disks by autodeposition in an alkaline milieu. The disks were counted with alpha PIPS detectors for 24 hours (**Table 10**).

We had to declare objections for 12 mussel and 2 fish samples (sardines). Sardines showed elevated activity of ^{210}Po. They are consumed as the whole fish, the

Radionuclide	Mean ± s.d.	Samples[*]	Min	Max
Fish				
^{210}Po	0.004 ± 0.005	21/	0.002	0.02
^{226}Ra	3.2 ± 6.3	54/140	0.20	43
^{228}Ra	1.3 ± 0.87	22/100	0.40	4.5
Sardines, anchovies				
^{210}Po	0.02 ± 0.03	18/19	0.001	0.11
^{226}Ra	1.7 ± 1.4	5/19	0.62	4.1
^{228}Ra	0.71 ± 0.44	6/19	0.30	1.3
Seafood				
^{210}Po	0.005 ± 0.013	11/27	0.001	0.05
^{226}Ra	2.0 ± 1.7	12/27	0.30	5.4
^{228}Ra	1.2 ± 0.10	3/27	1.1	1.2

s.d.: Standard deviation.
[*]*Number of positive samples/total of samples.*
All data from [83, 84].

Table 10.
Investigation of fish and seafood.

intestinal tract included, similar to the consummation of mussels. This explains the higher contamination level in sardines and anchovies. In 2010, the state laboratory of Basel City undertook a second investigation with similar results. Because since 1990 the limit value for ^{210}Po in fish and seafood was raised from 10 to 150 Bq/kg (indeed, the rate of fish and seafood consummation in Switzerland is of minor relevance), no more objections had to be raised [85, 86].

7. Conclusions

In the radiation laboratory, where food and environmental samples are investigated, alpha spectrometry is a mandatory part of the instrumentation. Well-established analytical procedures exist for the analysis of polonium, radium, uranium, thorium and transuranium nuclides. Suitable radioactive sources are prepared either by co-precipitation, selective extraction or adsorption onto active surfaces. The alpha spectrometric equipment at choice are PIPS detectors, gas proportional detectors and liquid scintillation counters. Drinking water is the most important food. Periodical survey for radioactive contaminants is important to guarantee a secure consummation. Because drinking water sources may underlie seasonal variations of their activity concentrations, they must be reexamined from time to time. The dose-relevant radionuclides in our investigation of mineral waters and tap water were ^{210}Pb, ^{226}Ra and ^{228}Ra.

Additionally, there are some food categories, which may show elevated activities of alpha nuclides. Mussels and fish may enrich ^{210}Po in the gastrointestinal tract. When consuming whole fish, such as sardines or anchovies, higher amounts of ^{210}Po are taken up. This can lead to relevant doses, especially in countries where consummation of fish and seafood is a main part of the nutrition. Healing earths may contain higher amounts of ^{226}Ra and ^{228}Ra. In spices, radium species are dominant, while pepper can contain higher amounts of thorium nuclides.

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