

We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists

6,900

Open access books available

186,000

International authors and editors

200M

Downloads

Our authors are among the

154

Countries delivered to

TOP 1%

most cited scientists

12.2%

Contributors from top 500 universities



WEB OF SCIENCE™

Selection of our books indexed in the Book Citation Index
in Web of Science™ Core Collection (BKCI)

Interested in publishing with us?
Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected.
For more information visit www.intechopen.com



Gamma-Ray Spectrometry and the Investigation of Environmental and Food Samples

Markus R. Zehringer

Additional information is available at the end of the chapter

<http://dx.doi.org/10.5772/67099>

Abstract

Gamma radiation consists of high-energy photons and penetrates matter. This is an advantage for the detection of gamma rays, as gamma spectrometry does not need the elimination of the matrix. The disadvantage is the need of shielding to protect against this radiation. Gamma rays are everywhere: in the atmosphere; gamma nuclides are produced by radiation of the sun; in the Earth, the primordial radioactive nuclides thorium and uranium are sources for gamma and other radiation. The technical enrichment and use of radioisotopes led to the unscrupulous use of radioactive material and to the Cold War, with over 900 bomb tests from 1945 to 1990, combined with global fallout over the northern hemisphere. The friendly use of radiation in medicine and for the production of energy at nuclear power plants (NPPs) has caused further expositions with ionising radiation. This chapter describes in a practical manner the instrumentation for the detection of gamma radiation and some results of the use of these techniques in environmental and food investigations.

Keywords: gamma-ray spectrometry, neutron activation analysis, radioactive contamination, radiocaesium, radiostrontium

1. Gamma spectrometric equipment for the control of food and environmental samples

1.1. Theory of gamma spectrometry

Gamma rays are electromagnetic radiation and are part of photon radiation. They are produced when transitions between excited nuclear levels of a nucleus occur. Delayed gamma rays are emitted during the decay of the parent nucleus and often follow a Beta decay. There can be many transitions between energy levels of a nucleus, resulting in many gamma-ray lines. The typical wavelength is 10^{-7} to 10^{-13} m, corresponding to an energy range of 0.01–10 MeV.

Gamma rays can be detected through their interaction with matter. There are three main processes: photoelectric absorption, Compton scattering and pair production. The photoelectric effect occurs when a gamma ray interacts with an electron of an inner shell of an atom and a photoelectron is emitted. This is the most important effect for the detection of gamma rays with semiconductor detectors. The effect of Compton scattering describes the interaction of a gamma ray with matter when some of its energy is transferred to the recoil electron. The energy transmitted is a function of the scattering angle. Therefore, the Compton effect results in a broad range of gamma-ray energies, which gives a continuous background in the gamma spectrum. Pair production is the third effect when a gamma ray is absorbed by matter and loses energy to produce an electron/positron pair. This effect only occurs when gamma rays have more than 1.02 MeV energy, twice the rest mass energy of an electron (0.511 MeV) [1–4].

1.2. Semiconductor detectors

Until the mid-1970s, no germanium could be produced of the desired purity. The purity required for large-volume detectors could only be produced by doping germanium crystals with n-type impurities, such as lithium (Ge(Li)-detectors). Later on, pure germanium crystals became available in n-type or p-type form and of closed-end coaxial or planar geometry and as bore-hole crystals. n-type detectors cover an energy range from about 10 keV to 3 MeV, while p-type detectors range from 40 keV to 3 MeV. p-type detectors with a carbon fibre or beryllium window instead of the aluminium end cap are best to detect energies below 100 keV.

1.3. Requirements for proper gamma spectrometry

The minimum detectable activity (MDA) of a Ge-detector depends on its energy resolution, the efficiency of the crystal, peak/Compton-factor, background, measuring time, sample geometry, self-absorption and the emission probabilities of the gamma emission lines of the radionuclide. Information can be obtained from the homepages of the providers, such as Ortec, Canberra and others [5–7].

1.3.1. Detector calibration

Ge-detectors are calibrated for the energy response of the multi-channel analyser, peak resolution and counting efficiency. Normally, gamma spectroscopists use commercially available calibration sources containing a mix of gamma-nuclides, which cover the whole energy range. Such nuclides are ^{210}Pb or ^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co and ^{60}Co , ^{134}Cs or ^{137}Cs , ^{88}Y , ^{85}Sr . Such mixes cover an energy range from 46 keV (^{210}Pb) to 1836 keV (^{88}Y). The disadvantage is that some of the nuclides have short half lives (e.g. ^{85}Sr has a half-life of 65 days) and therefore such calibration mixes only can be used for a year. Sometimes it might be better to use a mix of a low-energy nuclide (e.g. ^{241}Am) and ^{152}Eu , which disintegrates slowly (half-life of 13.5 years) and shows a multiplicity of emission lines from 122 to 1528 keV. The disadvantage is the summing effects of ^{152}Eu , which require correction (e.g. using software based on Monte Carlo simulations). Furthermore, the calibration of peak resolution and efficiency of the sample geometry is necessary; this is performed with the same calibration sources. We use calibration sources with $^{241}\text{Am}/^{152}\text{Eu}$ of different sample geometries. They are solidified by gelation and have a

density of 1.0 g/mL. Such sources are available, e.g. at Czech Metrology Institute at Prague [7]. Peak resolution should be tested on a regular basis together with the energy calibration. The peak shape is close to a Poisson distribution. For more counts, the distribution is closer to a Gaussian shape. The peak resolution is given by the quotient of FWTM (full width at tenth maximum) versus FWHM (full width at half maximum). Ge-detectors show resolutions of typically 1–2.5 keV. The software for the recording and analysis of pulse high spectra is available from Canberra, Ortec products, Oxford instruments, etc. Interwinner software from ITEC is a commonly used software in Germany and Switzerland [8].

1.3.2. Background

It is absolutely necessary to know the background of the Ge-detector system. This depends on the shielding of the detector and the background of the laboratory. We use shielding with 10 cm of lead and an inner layer of copper 5-mm thick. Before our laboratory was built, radiation-poor materials for the construction of the walls, soil and ceiling were sought. We analysed different components, such as gravel, sand, white cement and additives from different producers. Our choice for gravel and sand was a local producer in the Swiss Alps. The cement was from Dyckerhoff in Denmark. With this effort, we could reduce the background of our counting laboratory from 70 to 20 nSv/h. Nevertheless, background is still present. Incoming cosmic muons are not suppressed. They can be reduced by building an anticoincidence chamber over the detector. Vojtyla et al. could reduce the background by a factor of 2.2 [9]. Another approach is described by Seo et al., using Marinelli beakers of aluminium and purging the surrounding air of the detector reduced the background [10]. The background has to be measured periodically for each geometry to this end; a sample container with deionised water is placed on the detector and counted over a weekend. The background depends on the counting geometry and the matrix.

1.3.3. Attenuation effects

Photons may be absorbed by the sample matrix and therefore do not reach the detector. This effect depends on the elemental composition of the sample and its density. The photon attenuation effect is not negligible for photons with lower energies and for high-volume sample geometries. It has to be taken into account with the Gamma spectrometry software or software based on Monte Carlo simulations.

1.3.4. Coincidence summing effect

This effect occurs for all radionuclides emitting at least two photons in sequence and is a function of the source-detector distance and the detector efficiency. With a 50% Ge-detector, more coincidence summing is recorded than with a 20% detector. To avoid this effect, the samples may be counted a certain distance away from the detector.

1.3.5. Dead time

Samples of high activities may lead to a loss of peak counts. This effect occurs when the pulse processing electronics are slower than the frequency of the incoming photons. This leads to a dead time of the detector. Normally, environmental and food samples do not show such

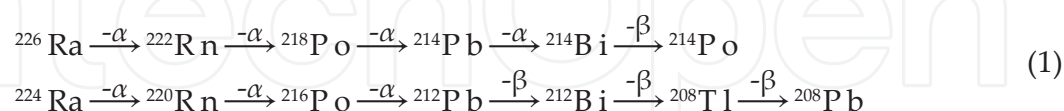
high activities that result in dead times of the detector. Dead time can be avoided by counting the sample at a well defined distance from the detector. Such constellations can be important when the samples have to be analysed for an emergency case.

1.4. Best sample geometries for gamma-ray analyses

MDA is a function of the detector efficiency and the sample weight. The best gamma-ray efficiencies are achieved with Marinelli beakers of 1 or 2 L, as the gamma rays of the sample interfere on top and on the sides with the Ge-crystal, gaining more efficiency. This geometry is best when large sample amounts of water, soil, vegetation, food, etc. are available. Marinelli geometries have to be calibrated carefully and coincidence summing has to be corrected. For small sample amounts, dishes with volumes of 32 and 77 mL (12 or 24 mm height and 6.5 cm in diameter) might be used. In small sample devices, the attenuation of gamma rays by the sample matrix is remarkably decreased. The disadvantage is the small sample load of 30–80 g. Other geometries commonly used are beakers of 250 and 500 mL volume. To enhance the sensitivity of the gamma-ray spectrometry, samples containing water may be freeze-dried. For milk samples, a concentration factor of eight can be achieved by freeze-drying. Soil, vegetation and food samples should also be dried (e.g. at 120°C). Soil samples are ground and sieved to eliminate large particles, such as stones or root parts. Further practical advice is given in other sources [11, 12].

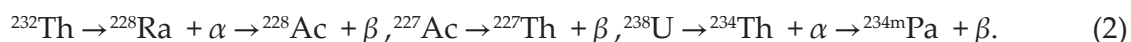
1.5. Interpretation of gamma spectrometry data of natural radionuclides

Dose-relevant radionuclides of the natural decay series of ^{238}U , ^{232}Th and ^{235}U are nuclides from uranium, radium, thorium, actinium, lead and polonium. Relevant criteria are the half-life and the dose coefficients of these radionuclides. With a few exceptions, these radionuclides can be detected via the gamma emissions of their daughter nuclides. The exceptions are ^7Be , ^{40}K , ^{223}Ra , ^{210}Pb , ^{231}Pa . The detection of ^{226}Ra and ^{224}Ra needs a secular equilibrium between the mother nuclide and its daughters. This can be reached when the sample is packed gas tight for at least the sevenfold half-life of the corresponding radon nuclide prior to the gamma analysis. This equates to 20 days in the case of ^{226}Ra , or 7 min for ^{224}Ra .



After reaching secular equilibrium, the activities of the daughters of ^{222}Rn and ^{226}Ra can be set equal to the activities of ^{214}Pb and ^{214}Bi . Often, the direct determination of ^{226}Ra is not possible due to the major interference with the gamma line of ^{235}U around 186 keV. Therefore, this is the best method to detect radium using gamma-ray spectrometry. The same applies for the system $^{228}\text{Th}/^{224}\text{Ra}$ and their daughters ^{212}Pb and ^{212}Bi . Here, equilibrium is reached within minutes due to the very short half-life of ^{220}Rn .

Other mother-daughter systems can be used for the determination of ^{232}Th , ^{228}Ra (a pure β -emitter), ^{227}Ac and ^{238}U .



Due to the very short half-lives of the daughters, these radionuclides are already at equilibrium. **Table 1** shows the adequate choice of gamma emission lines for relevant natural radionuclides. In natural uranium, the activity ratio of $^{238}\text{U}/^{235}\text{U}$ is 21.7.

Radionuclide		Energy (keV)	Emission probability ϵ (%)	Interferences
^7Be	Direct	477.61	10.3	
^{40}K	Direct	1460.8	10.67	
^{226}Ra	Direct	186.2	3.5	^{235}U (185.72 keV; 57.2%)
	^{214}Pb	295.21	18.2	^{211}Bi (351.06 keV; 12.91%)
	^{214}Pb	351.92	35.8	
	^{214}Bi	609.32	44.6	
	^{214}Bi	1120.3	14.8	
	^{214}Bi	1764.5	15.4	
^{228}Ra	^{228}Ac	338.32	11.3	
	^{228}Ac	911.21	26.6	
	^{228}Ac	968.97	15.8	
^{228}Th	^{224}Ra	240.99	4.0	^{214}Pb (241.98; 7.12%)
	^{212}Pb	238.63	43.3	^{228}Ac (583.41 keV; 0.114%)
	^{212}Pb	300.09	3.3	
	^{208}Tl	277.36	2.3	
	^{208}Tl	583.17	30.5	
	^{208}Tl	860.56	4.5	
^{227}Ac	^{227}Th	235.97	12.1, 11.2	
	^{227}Th	256.5	7.0	
	^{223}Ra	269.5	13.7	
^{235}U	Direct	143.76	10.96	^{226}Ra (186.1; 3.51%)
		163.33	5.08	^{228}Ac (204.10; 0.171%)
		185.72	57.2	
		205.31	5.01	
^{238}U	^{234}Th	63.28	4.3	^{232}Th (63.81; 0.267%)
	^{234}Th	92.37	2.5	Weak line
	^{234}Th	92.79	2.4	Weak line
	$^{234\text{m}}\text{Pa}$	766.37	0.21	
	$^{234\text{m}}\text{Pa}$	1001.03	0.84	
^{210}Pb	Direct	46.54	4.2	Weak line
^{223}Ra	Direct	154.21	5.6	
	^{211}Bi	269.46	13.7	
	^{219}Rn	351.07	12.9	
	^{219}Rn	271.23	10.5	
	^{219}Rn	401.81	6.5	
^{231}Pa	Direct	300.07	2.5	
		302.67	2.2	

Table 1. Common used emission lines for the detection of natural radionuclides with gamma-ray spectrometry. Emission probabilities are mean values from different sources [13–17].

When analysing for natural radionuclides, it is very important to know and to reconsider the background of the gamma system. Prominent radionuclides in the background are radon and its daughter nuclides from underground radiation. The background has to be determined for each geometry and has to be subtracted from the sample. We determine the background radiation using gamma spectrometry with sample containers containing deionised water, to take into account that the sample matrix also absorbs a part of the background radiation.

2. Instrumental neutron activation analysis

2.1. Principle

Instrumental neutron activation analysis (INAA) is based on the production of short-lived radionuclides by nuclear reactions. Most frequently, reactor neutrons (i.e. thermal neutrons) are used to activate many nuclides to produce radioactive nuclides. The efficiency of the irradiation process depends on the flux density of the neutrons and the cross section of the nuclear reaction of the irradiated nucleus. Typically, the thermal neutrons required for INAA are generated in a nuclear reactor [18, 19]. For our experiments, we used the reactor at the University of Basel (AGN-211-P), which is a light water-moderated swimming pool reactor. The compact core contained 2.2 kg of highly enriched uranium and a graphite reflector around the core. This uranium gave a thermal neutron flux of 3.8×10^{10} n/cm²/s at a power of 2 kW. The insertion of samples into the core was possible over a cannula (brown cylinder above the reactor in **Figure 1**) through the so-called glory hole. The glory hole consists of an air-filled pipe of a diameter of 1" (2.5 cm), which goes from above the pool down and through the centre of the reactor.



Figure 1. Equipment for NAA. Swimming pool reactor (left) gamma-ray spectrometer with lead shielding (middle) gamma-ray spectrum (above right) curry sample and sample device for the irradiation [4].

2.2. Operational procedure

The detection limit of gamma spectrometry is given by the half-life of the activated nuclides and the underlying Compton background from highly activated nuclides, such as sodium or chloride. Gold foils are used as the internal standard for each sample and are set on top of each sample. A sample series of 12 samples each of 1–2 g material was irradiated for 30 min at a power of 2 kW. Each irradiation place in the neutron field of the reactor was calibrated with coagulated salt solutions containing a known amount of the analyte and a corresponding gold foil. The response factors of each analyte to its gold foil for each place in the neutron field were then calculated (comparator method).

2.3. Common applications of INAA

2.3.1. Determination of total bromine content in food samples

The total bromine content of food, such as tea, coffee, dried mushrooms, vegetables and spices, gives information about the use of methyl bromide, a fumigant. The application of methyl bromide results in residues of bromide. This bromide can be activated to the gamma-active compound ^{82}Br (half-life of 35 h) by neutrons and analysed with gamma spectrometry. After irradiation of 30 min, the samples have to be cooled down for several hours (for the disintegration of activated sodium and chloride nuclides). The gamma analysis takes 15 min.



According to **Table 2**, many objections had to be executed for spices and dried mushrooms, which were treated with methyl bromide. Our last investigation of tea resulted in one objection. Since some years, the use of methyl bromide as a fumigant has been rare. Other fumigants, such as sulfuryl fluoride, hydrogen cyanide and phosphines, have become more important [20].

2.3.2. Determination of total iodine content in food

Algae and other food samples rich in iodine were irradiated to determinate the total content of iodine. Iodine is essential for the production of thyroid hormones and prevents goitre. In most European countries, people suffer from an iodine deficiency. The iodine level can be increased by the consumption of iodine-enriched food and dietary supplements. However, high levels of iodine (i.e. over 500 $\mu\text{g/kg}$) can result in hyperthyreosis. Therefore, the range of tolerance for iodine is narrow and it is important to declare the correct iodine content for food.

About 1 g of sample can be activated with reactor neutrons (30 min, 2 kW). The radioactive product ^{128}I is analysed directly using a gamma spectrometer



The half-life of ^{128}I is only 25 min; therefore, the samples had to be counted immediately after the activation. This is unfavourable regarding the background of other activated ions and results in higher detection limits. The total iodine content of fish, seafood, algae and dietary supplements can be analysed [22, 23].

Year/food	Spices	Dried mushrooms	Tea	Coffee	Chocolate	Rice
1988	10 (172)					
1989	4 (34)	7 (48)				
1990					0 (30)	
1991	5 (28)		0 (27)			
1992	2 (57)			0 (5)		
1994	0 (30)					0 (24)
1995	0 (33)					
1996		1 (33)				
1998		0 (15)				
2001	1 (26)					
2002			2 (33)			
2006			0 (17)			
2009			1 (40)			

Objections, which prove a use of the fumigant methyl bromide, show a bromine content over the limit. Limit values are 50 mg/kg (coffee, tea) and 100 mg/kg (spices mushrooms) according to the Swiss Ordinance on contaminants and constituents in Food [21]. The number of investigated samples is given in brackets.

Table 2. INAA analyses of food samples for total bromine content.

2.3.3. Determination of flame-retarding agents in plastics

INAA can be used as a screening analysis for flame-retarding agents in plastic materials, such as decabromo-bis-phenylether or tetrabromo-bisphenol A. The activation and decay process are the same as for the bromine analysis in food samples. The INAA gives information about the total content of brominated flame-retarding agents. We used INAA as a screening analysis and samples containing a high amount of bromine were detected and then analysed with gas chromatography to determine the amount of different flame-retarding compounds [24–26].

2.3.4. Determination of U and Th in suspended matter, sediment and soil samples

About 1 g samples of dried and ground material (e.g. freeze-dried suspended matter) were irradiated for 30 min at a power of 2 kW. After a cooling period of 2 h, the samples were analysed with a gamleast 30 min [27]

Activation processes : $^{238}\text{U} + n \rightarrow ^{239}\text{U} + \beta + \gamma$ and $^{232}\text{Th} + n \rightarrow ^{233}\text{Th}$
Decay processes : $^{239}\text{U} \rightarrow ^{239}\text{Np} + \beta + \gamma \rightarrow ^{239}\text{Pu} + \beta + \gamma$
and $^{233}\text{Th} \rightarrow ^{233}\text{Pa} + \beta + \gamma \rightarrow ^{233}\text{U} + \beta + \gamma$ (5)

3. Gamma-ray sources in the environment

Materials that emit radioactive rays are called radioactive sources. We distinguish between naturally occurring radioactive material (NORM) and technologically enriched naturally occurring radioactive material (TENORM) on the one hand and artificially produced radioactive sources on the other. Radionuclides may emit different rays, such as alpha, beta and gamma rays. Most α - and β -decays are accompanied by γ -rays. There are only a few important exceptions, such as ^{210}Po , ^{63}Ni , or ^{90}Sr , which are pure α - or β -emitters. Therefore, many important β -nuclides can be detected with gamma spectrometry.

3.1. First use of natural radioactivity

Soon after the discovery of radioactivity by Henry Becquerel, Pierre and Marie Curie and others, when radium became available, the production and the commercial use of TENORM began. Radium and thorium were used as remedies to cure many diseases. Underwear and wool, soap, lipstick, hair shampoo, toothpaste, suppositories, soda drinks, butter, etc. were spiked with NORM or TENORM. The negative effects of TENORM went visible only decades later. Quacks, such as William Bailey, earned their money by dealing with radioactive sources as medicinal drugs. It was therapy with radithor (a mixture of ^{226}Ra and ^{228}Ra) that led to the tragic death of Eben McBurney Byers [28]. Another tragedy was the “radium girls” from New Jersey. Many young women became ill or died from painting watch dials with radium. These and many more cases became public and led to the decline of the popularity of radioactivity [29]. Today, radon therapy in radon water, inhalation of radon air in tunnels or drinking of radon water remain the few existing applications of NORM for health cures against chronic diseases, rheumatic diseases and Morbus Bechterew. The health effects of radon are well described, but are not fully understood [30]. Such dubious items of the past are sometimes still present in households (see Section 3.3).

3.2. Natural radioactivity in food

Some natural radionuclides from the natural decay series of uranium and thorium enter the food chain. The alpha nuclide polonium-210 (^{210}Po), a product of the decay series of uranium-238 (^{238}U), is enriched in the intestinal tract of mussels and fish. Lead-210 (^{210}Pb), radium and thorium nuclides are present in cereals. In addition, spices and salt may contain elevated levels of radium and potassium-40 (^{40}K). Generally, potassium-rich food is also rich in ^{40}K (e.g. tea, vegetables). A special case is Brazil nuts, which are enriched in radium from soil. This is well described in [31]. Tap water may contain uranium, radium and their daughter nuclides depending on the local geological situation [32].

3.3. Radioactive sources in consumer products

Remnants from the application of natural radionuclides in the past century may be present in households even today. Our laboratory maintains a collection of radioactive objects that people brought in for investigation or disposal.

The use of thorium in flame detectors is widespread: for instance, in dials with radium in watches or on dial-plates for military use, coloured glass pearls or drinking glasses containing uranium oxides, wall tiles with uranium oxides, etc (Table 3). The finders of such items are encouraged to bring them to a specialised laboratory or to a collecting point for radioactive materials. The included radioactive material may be harmful.

Consumer product	Radionuclide(s)	Radionuclide content range
Radio luminous timepieces	^3H ^{147}Pm ^{226}Ra	4–930 MBq 0.4–4 MBq 0.07–170 kBq
Marine compass	^3H ^{226}Ra	28 MBq 15 kBq
Aircraft luminous safety devices	^3H ^{147}Pm	10 kBq 300 kBq
Static eliminators	^{210}Po	1–19 MBq
Dental products	$^{\text{nat}}\text{U}$	up to 4 Bq
Gas mantles	^{232}Th	1–2 kBq
Welding rods	^{232}Th	0.2–1.2 kBq
Optical glasses Ophthalmic lenses [35]	^{232}Th	5–75 Bq
Glassware: vaseline glass, canary flint glass	$^{\text{nat}}\text{U}$	100 kBq
Lamp starters	^{85}Kr	0.6 kBq
Smoke detectors	^{241}Am	37 kBq
Electron capture detectors	^{63}Ni	370 kBq
Drinking devices “Radium Drinkkur”	^{226}Ra , (^{222}Rn)	100 MBq
Wall tiles, ceramics	$^{\text{nat}}\text{UO}_3$	50–500 kBq
Granitic surfaces	$^{\text{nat}}\text{U}$	5–10 kBq/kg
Cardiac pacemaker [36]	^{239}Pu	113 GBq

Table 3. Consumer products containing radioactive materials (modified after Ref. [37]).

Incandescent gas mantles are in use without the knowledge of any possible danger. They contain ^{232}Th -oxides used to produce a bright light, which may be inhaled when the gas mantle disintegrates. In Germany and Switzerland, these gas mantles have been banned from the market. Attention has to be given to imports of products from the Far East. They may still contain thorium oxides.

A special case is the “Radium Drinkkur” (radium drinking device). It was used at the start of the twentieth century. The “Drinkkur” contained a small piece of pitch blend as a radium source (e.g. 100 MBq). The idea was to enrich drinking water with radon by emanation. Radon is said to be a remedy against rheumatics. Our own experiments have shown that a 2 month application of such a drinking therapy gave a yearly dose of 34 mSv, only from the radon. Unfortunately,

radium was also released when the source was immersed into the water. Therefore, an even higher dose with additionally washed-out radium could be incorporated [33].

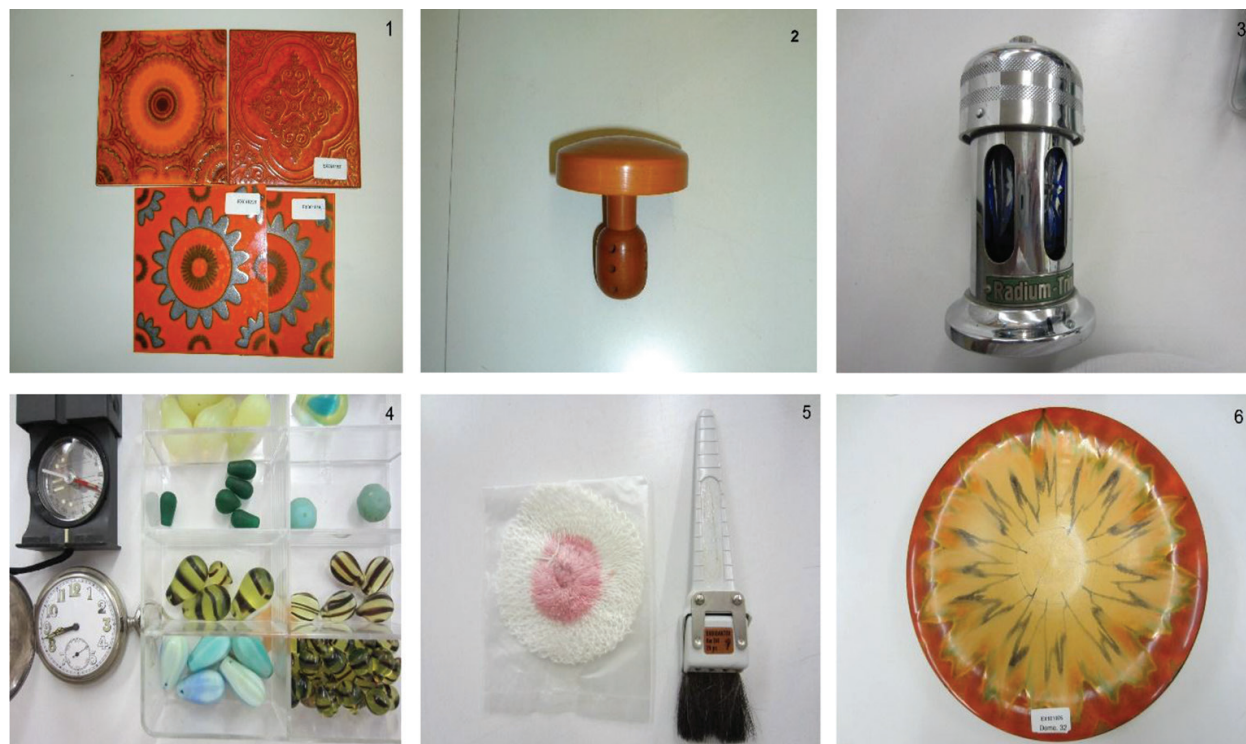


Figure 2. (1) Wall tiles; (2) pitch-blend source for radium drinking device (3); (4) watch, compass, glass pearls; (5) gas mantle and static eliminator; (6) bowl with paintings.

In the 1960s, radioactive wall tiles were discovered in Swiss households. They were produced with uranium oxide to obtain a brilliant red colour. Radiation from the walls of kitchens and toilets was of minor concern (low gamma energies), but a certain risk existed when the tiles were removed. The unavoidable dust contained uranium oxides; its inhalation had to be avoided. The Federal Office of Public Health regulated the professional drawbacks and disposal of the radioactive tiles [34] (**Figure 2**).

4. Gamma nuclides in the environment

Radioactive fallout is the main source for artificial radionuclides in the environment. In the following section, the application of gamma-ray spectrometry in Swiss environmental monitoring programs will be presented with examples [38].

4.1. Swiss monitoring programme

The Federal Office of Public Health (BAG) publishes a yearly report on the radioactivity in the environment and on radiation doses of the Swiss public. Several institutions, such as

Labor Spiez (LS), Institut de Radiophysique Appliqué (IRA), Paul Scherrer Institut of ETH Zurich (PSI), the Swiss Federal Institute of Aquatic Science and Technology (EAWAG), the National Emergency Operations Centre (NAZ), the Swiss Accident Insurance Fund (SUVA), the Swiss Federal Nuclear Safety Inspectorate (ENSI), the BAG, the European Organization for Nuclear Research (CERN), laboratories of the NPPs and some of the state laboratories of Switzerland, analyse different compartments with different techniques [39]. The main content of the reports lies in the supervision of emissions from NPPs and other industry using and producing radionuclides, the emissions from wastewater treatment plants and waste incineration plants. The report shows the results of the yearly survey of a grid of environmental sampling points, such as farms, sampling points in the vicinity of NPPs, water and air monitoring stations. Data from monitoring stations are collected and interpreted. These are the NADAM-net of the NAZ (66 automatic radiation dose meters [40]), the MADUK-net, operated by the ENSI (radiation dose meters [41]), RADAIR (air monitoring stations) and URANET (Automatic River monitoring detectors) both operated by BAG [42, 43]. These results are completed with the investigation of human tissues, such as the investigation of teeth and bones by the IRA or whole-body counting at the university hospital in Geneva and investigation of special radionuclides, e.g. ^{14}C in tree leaf samples in the vicinity of NPP's and chemical industries by the University of Berne. Based on these results, the radiation exposure of the public is estimated annually.

From the beginning, the state laboratory of Basel-City took part. The Office of Public Health chose for us three sampling points (farms) in Ticino, one farm in Basel-Country and a milk processing centre in the Canton of Jura for the yearly analysis of soil (upper 5-cm layer), grass and milk. Milk from milk distribution centres and other sites are analysed for radiostrontium as a supplement. Also, the survey of suspended matter of the River Rhine in Basel was delegated to our laboratory. Our laboratory also controls the local emissions of radioactivity. These are the wastewater of the local hospitals, the waste water treatment plant (WWTP) of Basel, ProRhen, and the incineration Plant of Basel, KVA Basel. Monitoring was started in 1993.

4.1.1. Environmental monitoring

In 1986, the southern parts of Switzerland were the most heavily affected by the fallout from Chernobyl. This can clearly be demonstrated by the time series of the investigated soil, grass and milk samples. There and also in elevated sampling points, such as in the Jura mountains, a remarkable increase in the radio-strontium and radio-caesium levels from global fallout was observed from 1986 to 1988. Other sampling points, such as Grangeneuve, the vicinity of the NPP's of Gösgen and Mühleberg and Leibstadt showed the normal decreasing trend of the global fallout with only slight additional fallout from Chernobyl [44]. For radiostrontium, they did not observe an increase in the contamination. Corcho et al. [45] recently published trend curves for three alpine investigated sites and two sampling sites in Swiss Mittelland. They analysed radioactive data of soil, grass and milk samples from 1994 to 2013. The effective half-lives did not depend on the altitude of the site. Radiostrontium showed quite a shorter half-life than the physical half-life. This can be explained by its migration to deeper soil layers and therefore less being available for plants. On the contrary, the effective half-life

of radiocaesium is similar to its physical half-life. This is because it is fixed on clay particles in the soil and moves only slowly into the deeper soil layers [45].

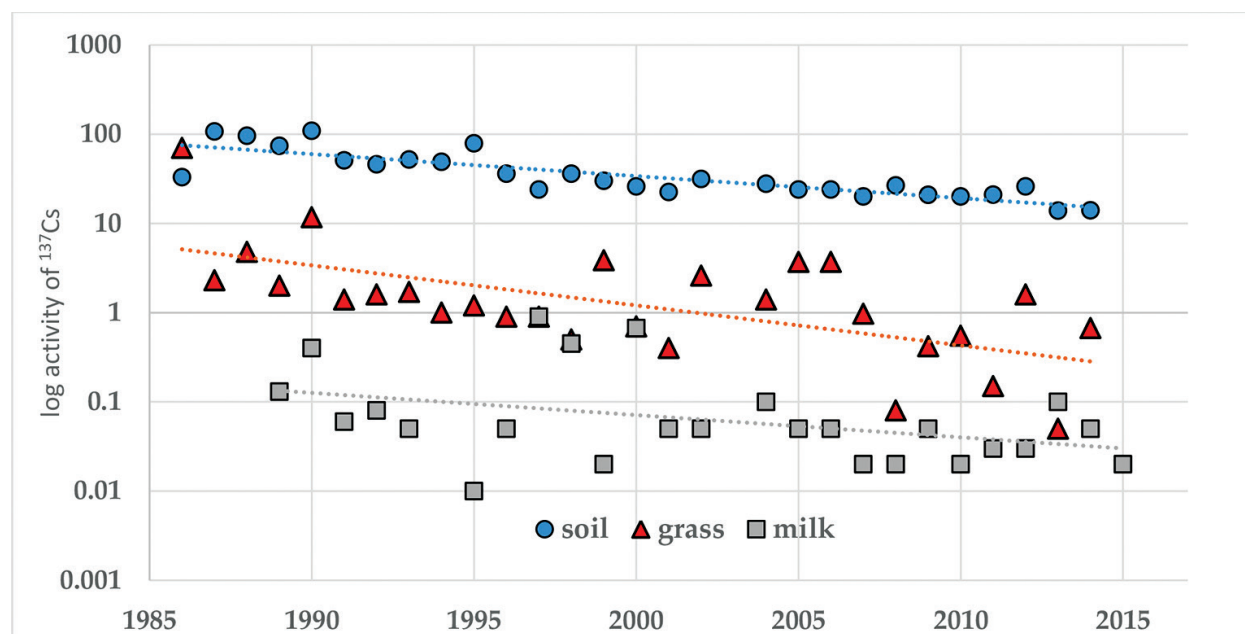


Figure 3. Activity trends of radiocaesium in soil, vegetation and cow's milk on a farm in Basel-Country. We calculated the effective half-life to 12.9 years (soil), 11.6 years (grass) and 12.0 years (cow's milk).

One of our regular sampling points is a farm in Basel-Country (450 m altitude). Data from 1986 until today are available. The data analysis of ^{137}Cs shows the following trends (**Figure 3**). The effective half-life of radiocaesium is lower than the physical one. We explain this fact by movement of the radionuclide into deeper soil layers. Therefore, the radiocaesium becomes progressively less available for the grass roots.

Such calculations are of interest when trends in the behaviour of radionuclides for food and feed are sought. They provide important information about the contamination of the food chain. The Chernobyl impact resulted in an increase in the contamination in cow's milk for about 6 years.

4.1.2. Monitoring of local emissions of radionuclides

In Basel, the main emission sources of radioactive material are the chemical/pharmaceutical industry, hospitals and some minor industries using radioactive sources. Emissions from these sources are deposited in the environment via wastewater and the air. The waste water is cleaned at the local WWTP ProRheno and two waste incineration plants, the city's incineration plant, KVA Basel and an incineration plant of the chemical industry for hazardous wastes, RSMVA of Valorec.

An important task of the state laboratory is the monitoring of the local wastewater effluents. These monitoring programmes started in 1988 and included the parameters ^3H (mainly used

by the local chemical/pharmaceutical industries) and short-lived radionuclides used in hospitals, such as ^{131}I , $^{99\text{m}}\text{Tc}$, ^{90}Y , ^{111}In , ^{177}Lu , ^{186}Re , ^{153}Sm , ^{67}Ga and others. The university hospital of Basel is specialised towards DOTATOC therapies, where mainly ^{90}Y and ^{177}Lu are used. Wastewater from the patients is collected in cool-down tanks for some weeks before being discharged to the wastewater treatment plant. Monitoring results are published yearly [46].

Tritium emissions are regularly detected in the washing water of the air filters of KVA Basel. Several times during the last 25 years, tritium was above the permitted emission limits of 6000 Bq/L and 60,000 Bq/month. However, these emissions contribute very little to the ^3H -level of the River Rhine. Here, the main ^3H sources are the NPPs and tritium producing industry in Switzerland upstream [47].

The main contamination factor of air is radiocarbon, which are used mainly by the chemical/pharmaceutical industries. In Basel, radiocarbon is emitted when waste is burned at RSMVA. Incineration takes place mainly in the night, to lower the uptake of radiocarbon in plants by photosynthesis. These emissions are controlled annually by the University of Berne. The monitoring programme of the University of Berne includes other emission sources in Switzerland, such as incineration plants, ZWILAG and NPPs [39].

4.1.3. Behaviour of radionuclides in a WWTP

In 2014, the influents and effluents of the WWTP ProRheno were investigated, to obtain a balance of short-lived radionuclides for medical use. The main fraction of ^{131}I was dissolved in wastewater and over 90% of the input was emitted with the treated wastewater into the River Rhine. Only a small amount was emitted via air when the sewage sludge is incinerated. For ^{177}Lu , we observed that about 60% of the incoming activity was eliminated in the WWTP, mainly by adsorption on the sewage sludge. Finally, it remained in the sludge ash, which is deposited on a nearby landfill disposal site. It disintegrates there rapidly. About 40% of the activity is emitted with the treated wastewater to the River Rhine and deposited on suspended matter and river sediment [48, 49]. The investigation of sewer sludge in the local sewage water system of Basel clearly shows that a part of the activities found at the influent of the WWTP originates from patients treated in ambulances [50].

Some of the waste from the hospitals is burnt at the incineration plant. This is proven by the activities found in the washing water of the air filters. While ^{131}I is found permanently in the low Becquerel level, other nuclides only are found sporadically. The main contamination factor at KVA Basel is ^3H . The sources of these emissions are not known. The Swiss Accident Insurance Fund, SUVA, supposes the source to be the accidental burning of ^3H -containing watches with the daily-delivered waste from households and industry.

4.1.4. Suspended matter of the River Rhine

Many contaminants, such as organics, metals and radionuclides, adsorb onto clay particles and are transported in a river as suspended matter. After quite a long distance or sections where the river water stands still, e.g. behind dams, the suspended particles settle onto the river sediment. Radionuclides released from NPPs are monitored by EAWAG and our

laboratory at defined sampling points downstream. Suspended matter is collected either continuously by a special particle-settling chamber, or by the use of a centrifuge (i.e. non-continuous monitoring). At the river monitoring station Weil, downstream of Basel, suspended matter is collected monthly with a centrifuge. The freeze-dried and ground material is then analysed with a Ge-detector. Beside NPP-specific radionuclides, such as ^{60}Co , ^{54}Mn or ^{65}Zn , radionuclides from medicinal applications (^{131}I , ^{177}Lu and others), natural radionuclides from the decay series of U and Th and from fallout (^{137}Cs) can be detected [39, 51].

4.2. Special applications/projects

We now describe our own investigations to find representative organisms for radioactivity monitoring of the environment. Therefore, we analysed possible sample types, such as mosses, soil, grass, dust, water and wood.

4.2.1. Behaviour of radionuclides in soil filters of local drinking water production

The filtration of river water through forest soils is the most important step of drinking water production in Basel. In the context of an emergency concept for drinking water production, the question arose as to how the radioactive contaminants behave when entering the soil filter. Is there a danger of contamination of the drinking water after a nuclear accident? We analysed soil cores in one of the filtering fields for global fallout. As described in the literature, radiocaesium and plutonium remain in the upper soil layer. Radiostrontium moved deeper into the soil. For radiocaesium, we estimate two main sources: global fallout and fallout from Chernobyl. We estimated that in 1986, only about 65% of the total caesium reached the infiltration site with the infiltrated river water. The rest was bound to the suspended matter and removed by sand filtration before the infiltration step. Plutonium and perhaps radiostrontium, only originates from global fallout. We suppose that radionuclides are retained in the soil even at higher charges. For radiostrontium, the retention was lower and could have reached the groundwater. Further investigations are necessary (Figure 4) [52].

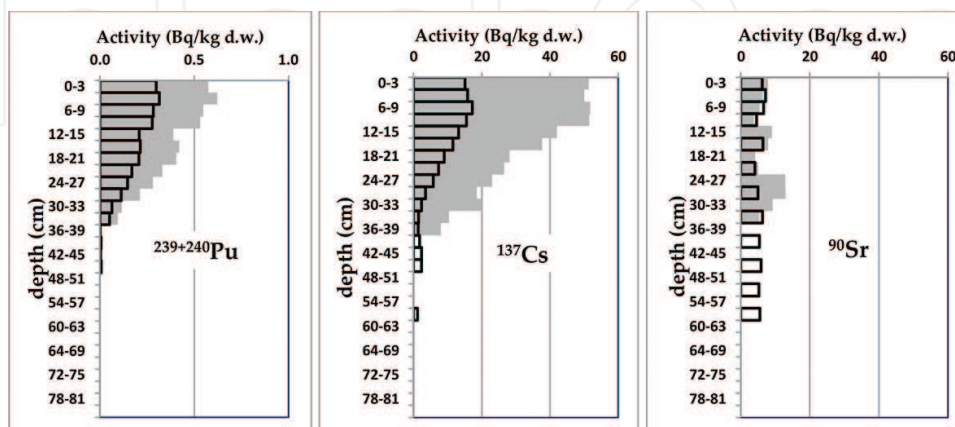


Figure 4. Soil profiles from a soil filtration site of the drinking water producer of Basel. Bars in grey are cores from the filtration site compared to a reference site outside (bars in white). From Ref. [52].

4.2.2. *Fallout monitoring at Basel and vicinity*

During the annual emergency exercises for an A-impact, soil samples were collected (the upper 5-cm layer) over 50 different sampling points in the city of Basel and surroundings at different altitudes (250–360 m). The analysis of over 200 soil samples with gamma-ray spectrometry resulted in an overall range of 10.6 ± 6.4 Bq/kg of ^{137}Cs .

A second project was focused on the analysis of mosses. Mosses were analysed with beta and gamma spectrometry. The analysis with gamma-ray spectrometry resulted in an overall range 2.2 ± 2.6 Bq/kg for ^{134}Cs ($n=3$) and 24 ± 42 Bq/kg ($n=87$) for ^{137}Cs with a maximum of over 300 Bq/kg. Radiostrontium was found in 67 samples: 5.2 ± 4.5 Bq/kg of ^{90}Sr [53].

Recently, our focus was on tree bark monitoring. The gamma-ray analysis of the tree bark of 26 different trees gave a mean of 6.7 ± 18 Bq/kg ^{137}Cs .

Compared to the undisturbed situation on a country site, variability in soil and vegetation in a city and surroundings is quite dominant. Nonetheless, we think that tree bark monitoring is comparable with soil monitoring and can give relevant contamination data for emergency cases. The uptake mechanism for radionuclides in mosses is quite different to that of trees and of the deposition on soils. Mosses do not have roots; they incorporate contamination mainly through the air. Contamination is deposited on tree leaves. Trees accumulate contamination through their roots and also later by the deposited leaves (litter-fall) [54].

Despite their great variability in moss species and the difficulties in the determination of age, mosses can be monitoring plants for radioactive fallout, when carefully normalised (**Figure 5**). In Basel, radiocaesium and radioiodine (^{131}I) were detectable in the first rainfall in April 2011, after the catastrophe at Fukushima-Dai-ichi. Here, over 9500 km away from Japan, the fallout could also be detected in moss, grass and soil and even in cow's milk [55].

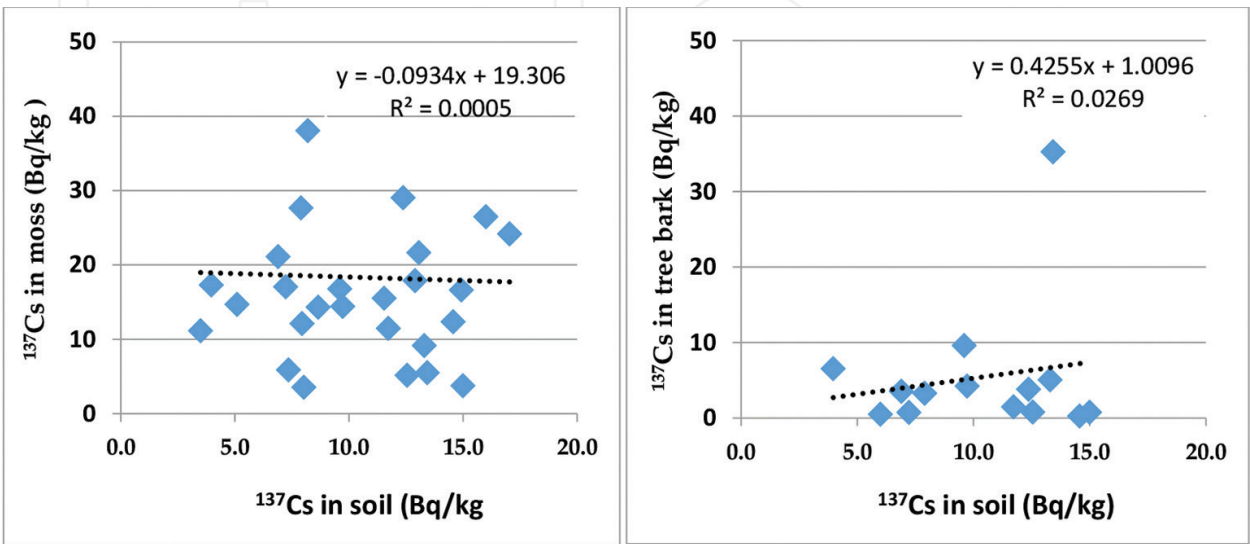


Figure 5. Correlations of radiocaesium between moss, tree bark and the corresponding soil activity.

5. Gamma nuclides in food

5.1. Radio contamination of food

Whereas food can be contaminated just after its release with fallout of short-lived radionuclides for a short period, contamination with long-lived radionuclides from global fallout and the Chernobyl catastrophe remain. The contamination of food by the Chernobyl fallout was reduced within 2 years concerning the short-lived radionuclides, such as ^{131}I , ^{132}I , ^{134}Cs , whereas long-lived radionuclides still persist (see **Figure 3**) in the soil and are transferred to crops and grass (feed for cows). The typical tracer food for this is milk. We recently published a review of radioactivity monitoring in Switzerland over the last 35 years [56]. We compared the contamination level of food categories with artificial radionuclides. In the time span 1990–2015, some moderate contamination of some food categories was noted. Special cases were hazelnuts and tea from Turkey. As some regions of that country were affected by the fallout from Chernobyl, food imports may contain higher levels of Radiocaesium (0.1–30 Bq/kg). Tea contained up to 100 Bq/kg radiocaesium and 2–40 Bq/kg of radiostrontium. The latter may also originate from global fallout. Until the present, most affected food from the fallout of Chernobyl concerns wild grown mushrooms, wild grown berries and game (especially wild boars). Even today, violations are noted for wild boars from Bavaria, Southern Germany and Southern Switzerland [57] (**Table 4**).

Food category	Origin	Radiocaesium	Radiostrontium
Hazelnuts (2005–2013)	Turkey	0.1–16 (n = 96)	n.a.
Milk (2004–2016)	Switzerland	<0.05–22 (n = 177)	0.01–26 (n = 194)
Baby food (2006–2016)	Switzerland, Europe	<0.05–0.5 (n = 71)	<0.1–0.33 (n = 71)
Mushrooms	Europe	<1–1'320 (n = 248)	n.a.
Vegetables (2006–2014)	Europe	<0.1–0.5 (n = 88)	<0.1–0.5
Wild grown berries ¹	East Europe	0.1–170 (n = 123)	<0.05–60 (n = 57)
Game	Europe	<1–1250	n.a.
Wild boar (2013–2014)	South Switzerland	1250 (n = 41)	n.a.
Wild boar (2014) [58]	Canton of Zurich	<0.2–388 (n = 28)	n.a.
Fish (2011–2015)	Pacific Ocean	<0.1–0.7 (n = 52) ²	0.1–0.4 (n = 7)
Flour, bred (2006–2011)	Switzerland	<0.1–5	n.a.
Honey (2004–2012)	Europe	<0.2–51.4	<0.05–2
Tea ²	Japan, other countries	<0.5–258.2	<0.1–57

¹ Including blue berries and chest nuts.

² Fish and Japanese tea contains also ^{134}Cs from local fallout of the Fukushima-Daiji accidents.
n.a.: not analysed.

Table 4. Some food categories which are still contaminated with global fallout and/or fallout from NPP's accidents in Chernobyl and Fukushima.

The Office of Public Health estimates the total ingested dose to about 0.3–0.4 mSv/year. The main contribution comes from potassium-40 (^{40}K ; 0.2 mSv/year) and from natural radionuclides of the uranium and thorium decay series. The remaining contamination from bomb fallout is less than 0.1 mSv/year [39].

5.2. Healing earths

Siliceous earths are widely used in the food industries as a food supplement. They incorporate foreign atoms in the crystal lattice, such as radionuclides of the natural decay series of uranium and thorium.

In 2008, siliceous earth products on the Swiss market were analysed with γ -spectrometry. In two products, the threshold value for natural radionuclides of group 2¹ was exceeded (>50 Bq/kg). Furthermore, by regular consumption of one product from California, USA, the annual dose would reach half of the permitted yearly dose of 1 mSv. Consequently, this product was withdrawn from the Swiss market. In 2010, the reinspection of healing earths showed that two products from one producer in Germany slightly exceeded the limit value according to higher levels of ²²⁶Ra and ²²⁸Ra. The annual dose from the consumption of these products would lead to 0.1 mSv/year. Therefore, healing earths and silica-based chemicals used in food industry and in chemical laboratories remain a source of natural radionuclides [59].

6. Gamma spectrometry as an important analytical tool for emergency cases with ionising radiation

The instrumentation of an emergency A-Laboratory depends on the required detection devices for the analysis of the fallout from nuclear bombs or from nuclear power plants. **Table 5** gives a short survey of some expected fission and activation products. In used reactor fuel, more than 200 radionuclides are present [60] (**Figure 6**). The Institut de Radiophysique Appliqué (IRA) prepared simulated gamma spectra 2 and 11 days after release from an NPP. After 2 days, the gamma analysis was very complex and contained more than 270 gamma emission lines. At day 11 after the release, the number of gamma lines was reduced remarkably. Not all expected radionuclides were detectable due to low activity. For mother/daughter nuclide pairs, attention has to be paid to when the half-life of the daughter is shorter than that of the mother. After seven half-lives of the daughter, the two nuclides are in equilibrium, so one has to calculate the activity of the daughter using the half-life of the mother nuclide (e.g. ¹³²Te (77.5 h) and daughter ¹³²I (2.3 h)) [61].

As we see, gamma spectrometry is the most important instrumentation for an emergency case with ionising radiation. Only for a few radionuclides α -spectrometry (Pu-isotopes) and β -spectrometry (³H, ¹⁴C, ⁸⁹Sr, ⁹⁰Sr) have to be available.

The main task is to analyse environmental samples, such as soil, vegetation, fallout, air and food samples, such as vegetables and fruit grown outdoors. The pathways are air/fallout, rain/wash-out and water (rivers, lakes). Later on, collection and analysis of sediment, grass and soil samples will follow. The most important/affected food is milk and milk products, baby food, outdoor-grown vegetables, meat and game, fruit (including hazelnuts) and cereals. According to our experience in 1986, vegetables were the most affected (by radioiodine and radiocaesium). Special focus should be set on baby food and human milk (including analysis for radiostrontium) [55].

¹Ordinance on Contaminants and Constituents in Food: natural radionuclides group 2: the sum of activities of ²²⁶Ra, ²²⁸Ra, ²³⁰Th, ²³²Th and ²³¹Pa.

Radionuclide	Half-lives	detection	Radionuclide	Half-lives	detection
Noble gases			Non-volatile nuclides		
^{85m} Kr	4.4 h	γ	⁶⁰ Co	5.3 a	Γ
⁸⁷ Kr	1.3 h	γ	⁵⁴ Mn	312 d	γ
⁸⁸ Kr/ ⁸⁸ Rb	2.8 h/50 d	γ	⁶⁵ Zn	244 d	γ
¹³³ Xe	125 h	γ	⁹⁷ Zr/ ⁹⁷ Nb/ ^{97m} Nb	17 h/1.2 h/53 s	γ
^{133m} Xe	53 h	γ	⁸⁹ Sr	50 d	β ¹
Halogens			⁹⁰ Sr/ ⁹⁰ Y	28.6 a/64 h	β ¹
			⁹¹ Sr/ ⁹¹ Y	9.5 h/59 d	γ
			⁹² Sr/ ⁹² Y	2.7 h/3.5 h	γ
			⁹⁵ Zr/ ⁹⁵ Nb	65 d/35 d	γ
			⁶⁰ Co	5.3 a	γ
			⁹⁹ Mo/ ^{99m} Tc	66 h/6 h	γ
¹³¹ I	192 h	γ	⁹⁷ Zr/ ⁹⁷ Nb	17 h/1.2 h	γ
¹³² I	2.3 h	γ	¹²⁷ Sb/ ¹²⁷ Te	3.9 d/9.4 h	γ
¹³³ I/ ¹³³ Xe	21 h	γ	¹²⁹ Sb/ ¹²⁹ Te	4.4 h/70 m	γ
¹³⁴ I	0.9 h	γ	¹²⁷ Sn/ ¹²⁷ Sb	2.1h/3.9d	γ
¹³⁵ I/ ^{135m} Xe	6.6 h/0.3 h	γ	¹⁴⁰ Ba/ ¹⁴⁰ La	306 h/40 h	γ
⁸² Br	35 h	γ	¹⁴¹ Ce	33 d	γ
Volatile nuclides			¹⁴³ Ce/ ¹⁴³ Pr	33 h/14 d	γ
			¹⁴⁴ Ce/ ¹⁴⁴ Pr	285 d/17 m	γ
			¹⁴⁹ Pm	53 h	γ
			²³⁵ U/ ²³¹ Th etc.	7 × 10 ⁸ a	γ
			²⁴¹ Pu/ ²⁴¹ Am	14.35 a/432 a	γ
			²³⁹ Np/ ²³⁹ Pu etc.	2.4 d	γ
			²⁴² Cm/ ²³⁸ Pu etc.	163 d	α
			²³⁸ Pu/ ²³⁴ U etc.	88 a	α
			²³⁹ Pu/ ²³⁵ U etc.	2.4 × 10 ⁴ a	γ
			²⁴⁰ Pu/ ²³⁶ U etc.	6.6 × 10 ³ a	α
¹²⁹ Te/ ¹²⁹ I	70 m/12.4 h	γ	²⁴² Pu/ ²³⁸ U etc.	3.7 × 10 ⁵ a	α
^{129m} Te/ ¹²⁹ Te	802 h/1.2 h	γ			
^{131m} Te/ ¹³¹ Te	30 h/0.4 h	γ			
¹³² Te/ ¹³² I	78 h/2.3 h	γ			
¹³⁴ Te/ ¹³⁴ I	42 m/52 m	γ			
^{133m} Te/ ¹³³ I	55 m/21 h	γ			
¹⁰³ Ru/ ^{103m} Rh	941 h/56 m	γ			
¹⁰⁶ Ru/ ¹⁰⁶ Rh	374 d/30 s	γ			
¹³⁴ Cs	2.1 a	γ			
¹³⁶ Cs/ ^{136m} Ba	316 h/	γ			
¹³⁷ Cs/ ^{137m} Ba	30a/2.5 m	γ			
³ H	12.4 a	β ¹			
¹⁴ C	5730 a	β ¹			

After Refs. [60–62].

¹Analysis has to be performed with β-spectrometry.

Table 5. Extract of possible fission and activation products released at an NPP accident or from a bomb.

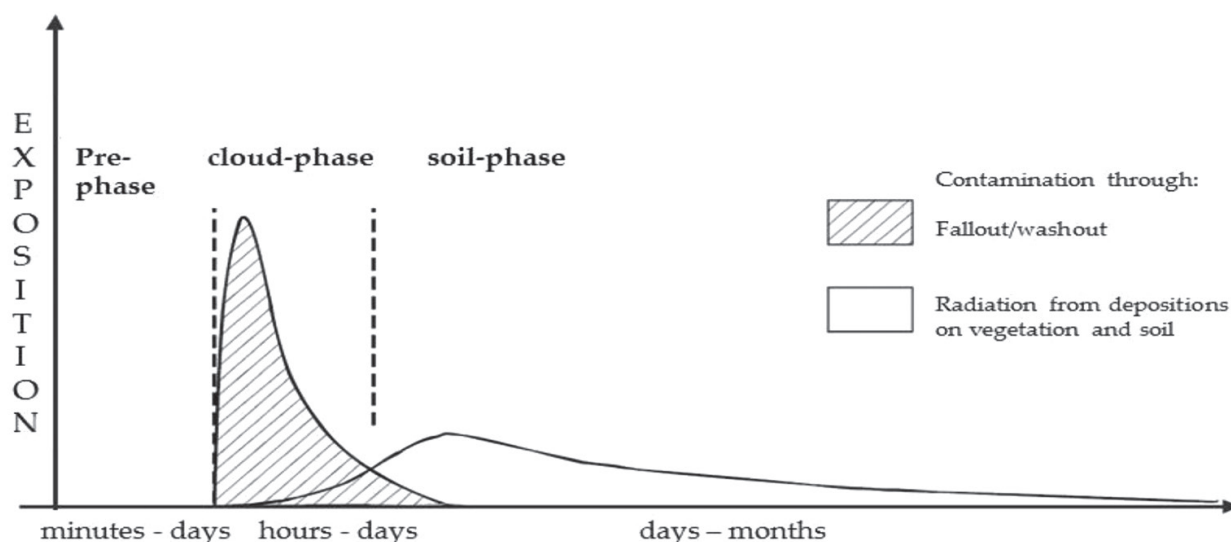


Figure 6. Process of a release of radioactive material to the environment by an accident or bomb (after Ref. [64]).

In Switzerland, the National Emergency Operations Centre (NEOC) coordinates these emergency investigations. They give the order to collect samples for the estimation of the contamination/radiation level outdoor and for controlling the level of the contamination of food. The local authorities then take action (e.g. banning severely contaminated food categories). An initial focus is set on the drinking water quality. In Basel, drinking water is produced from groundwater, which is enriched with river water by soil filtration. It is important to prevent the entry of contaminated river water into this filtration system. After an earthquake, when the drinking water production site is no longer operable, there exist plans for emergency supply of the public with pumped water from the ground and from rivers. This requires efficient and rapid analysis systems. With a gamma analysis of a 1 L water sample in a Marinelli-beaker, it is possible to restrict the analysis time to 15 min for the examination of threshold values. ^3H and ^{90}Sr have to be analysed by β -spectrometry. With an extraction/scintillation-method, we are able to obtain results for both radionuclides within 2 hours [63].

Acknowledgements

My special thanks go to my former collaborator, Matthias Stöckli. I benefited greatly from his great expertise in radiation detection, especially in Neutron Activation Analysis. Furthermore, I express my thanks to my collaborators Franziska Kammerer and Michael Wagmann for their permanent support. Finally, I wish to thank Major Franz Näf from the civil protection of Basel City for his engagement during the annual emergency exercises that are mentioned in Section 6.

Author details

Markus R. Zehring

Address all correspondence to: markus.zehring@bs.ch

Head of Radiation Laboratory, State-Laboratory of Basel-City, Basel, Switzerland

References

- [1] Knoll G. Radiation Detection and Measurement. 3rd ed. New York: John Wiley & Sons; 1999. ISBN 0-471-81504-7, p. 387ff.
- [2] Gilmore G, Hemingway J. Practical Gamma-ray Spectrometry. New York: John Wiley & Sons; 1995. ISBN: 0-471-95150-1.
- [3] L'Annunziata M, editor. Handbook of Radioactivity Analysis. 2nd ed. San Diego: Academic Press; 2003. ISBN 0-12-436603-1, p. 239–294.
- [4] Debertin K, Helmer R. Gamma- and X-Ray Spectrometry with Semiconductor Detectors. New York: Elsevier; 1988. ISBN: 0-444-871071.
- [5] Ortec. [Internet]; Available at: <http://www.ortec-online.com/>
- [6] Canberra. [Internet]; Available at: <http://www.canberra.com/products/detectors/> [Accessed: 2016-08-02].
- [7] Institute of Metrology Cze [Internet]; Available at: <https://www.cmi.cz/?language=en> [Accessed: 2016-08-02].
- [8] Interwinner by ITECH INSTRUMENTS [Internet]; Available at: <http://www.itech-instruments.com/> [Accessed: 2016-08-02].
- [9] Vojtyla P, Beer J, Stavina, P. Experimental and simulated cosmic muon background of a Ge spectrometer equipped with a top side anticoincidence proportional chamber. Nucl Instr Methods Phys Res B. 1994; **86**: 380–386.
- [10] Seo B, Lee K, Yoon Y, Lee D. Direct and precise determination of environmental radionuclides in solid materials using a modified Marinelli beaker and a HPGe detector. Fresenius J Anal Chem. 2001; **370**:264–269.
- [11] Valkovic V. Determination of radionuclides in environmental samples. In: Barcelo D, editor. Environmental Analysis: Techniques, Applications and Quality. Amsterdam: Elsevier Science Publishers; 1993, p. 311–356.
- [12] Wallbrink P, Walling D, He Q. Radionuclide measurement using HPGE gamma spectrometry. In: Zapata F, editor. Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides. Amsterdam: Springer; 2003. ISBN 978-0-306-48054-6. p. 67–87.
- [13] Lederer M, Shirley V. Table of Isotopes. 7th ed. New York: Wiley & Sons; 1978. ISBN: 0-471-04179-3.
- [14] Erdtmann G and Soyka W. The Gamma Rays of the Radionuclides. Weinheim: Verlag Chemie; 1979. ISBN: 3-527-25816-7.
- [15] Wahl W. α — β — γ -Table of Commonly Used Radionuclides, Version 2.1; 1995. D83722 Schliersee.

- [16] Firestone R, Shirley V. Table of Isotopes. 8th ed. New York: Wiley & Sons; 1999. ISBN: 978-0-471-35633-2
- [17] Legrand J, Perolat J, Lagourtine F, Le Gallic Y. Table of Radionuclides. Atomic Energy Agency, Gif-sur-Yvettes, France 1957.
- [18] Amiel S (Ed.). Nondestructive Activation Analysis, Amsterdam: Elsevier; 1981. ISBN: 0-444-41942-X.
- [19] Parry S. Handbook of Neutron Activation Analysis. Woking: Viridian Publishing; 2003. ISBN: 0-9544891-1-X.
- [20] Zehring M, Stöckli M. Determination of total bromine residue in food and non-food samples. *Chimia* 2004;**59**:112.
- [21] The Federal Department of Home Affairs. Ordinance on Contaminants and Constituents in Food; 1995.
- [22] Zehring M, Testa G, Jourdan J. Determination of total Iodine content of food with means of Neutron Activation Analysis (NAA). Proceedings of the Swiss Food Science Meeting (SFSM '13); 2013; Neuchatel.
- [23] Bhagat P et al. Estimation of iodine in food, food products and salt using ENAA. *Food Chem.* 2009; **115**: 706–710.
- [24] Frey T. Detection and quantification of brominated flame retarding agents in plastic materials. [Master's thesis]. Aarau; 1999.
- [25] Kuhn E, Frey T, Arnet R, Känzig A. Brominated flame retarding agents in plastic products on the Swiss market. In: Federal Office of Environment (Ed.) *Umweltmaterialien*; 2004;**189**.
- [26] Wegmann L, Werfeli M, Bachmann R, Tremp J, Figueiredo V. Brominated flame retarding agents in plastics. A tentative investigation on the Swiss Market, Amt für Umweltschutz und Energie. Liestal; 1999.
- [27] Zehring M et al. Neutron Activation Analysis (NAA)—another approach to uranium and thorium analysis in environmental samples. *Chimia* 2013; **67**:828.
- [28] Mahaffey J. Atomic Accidents. A History of Nuclear Meltdowns and Disasters. New York: Pegasus Books; 2014. ISBN: 978-1-60598-680-7.
- [29] Valerius-Mahler C. Radiation—the two faces of radioactivity. Pharmazie-Historisches Museum der Universität Basel; 2014, 67–87.
- [30] Pratzel H, Deetjen P. Radon Application at Sanitariums. Geretsried: I.S.M.H Verlag; 1997.
- [31] Pöschl M, Noll L, editors. Radionuclide Concentrations in Food and the Environment. New York: CRC Taylor & Francis; 2007. ISBN: 0-8493-3594-9.
- [32] Bünger T et al. Radioactivity in water—an actual overview. *Strahlenschutzpraxis*. 2014;**1**: 3–28.

- [33] Zehring M. Radiological investigation of a radium drinking device. In: annual report of the state-laboratory Basel-City. Basel; 2006. p. 173–175.
- [34] Zehring M. Wall tiles and other radioactive objects. In: Annual Report of the State-Laboratory Basel-City. Basel; 2006. p. 165–167.
- [35] Marzocchi O. Analysis of a radioactive lens and of the correlated UV-light bleaching properties. *Strahlenschutzpraxis* 2015;2: 46–49.
- [36] Marfeld A. Modern Electronics and Practice of Electro Technique. Berlin: Safari-Verlag. 1971. p. 210.
- [37] Buckley D, Belanger R, Martin P, Nicholaw K, Swenson J (Eds.). Environmental assessment of consumer products containing radioactive material. Report of the U.S. nuclear regulatory commission NUREG/CR-1775; 1980.
- [38] Izrael Y. Radioactive Fallout after Nuclear Explosions and Accidents. New York: Elsevier; 2002, eBook ISBN: 9780080540238. p. 102.
- [39] Federal Office of Public Health. Environmental Radioactivity and Radiation Doses in Switzerland. Bern: BAG [internet]; Available from: <http://www.bag.admin.ch/themen/strahlung/12128/12242/index.html?lang=de> [Accessed: 2016-08-02].
- [40] National Emergency Operations Centre (NAZ) [Internet]; Available at: <https://www.naz.ch/de/aktuell/messwerte.html> [Accessed: 2016-08-02].
- [41] Swiss Federal Nuclear Safety Inspectorate (ENSI) [Internet]; Available at: <https://www.ensi.ch/de/notfallschutz/messnetz-maduk/> [Accessed: 2016-08-02].
- [42] Federal Office of Public Health (BAG) [Internet]; Available at: <http://www.bag.admin.ch/themen/strahlung/12128/12266/12268/index.html?lang=de> [Accessed: 2016-08-02].
- [43] Federal Office of Public Health (BAG) [Internet]; Available at: <http://wp.radenviro.ch/?p=891&lang=de> [Accessed: 2016-08-02].
- [44] Geering J, Friedli C, Carlone F, Lerch P. Analysis of strontium-90. In: Federal Office of Public Health, editor. Environmental Radioactivity and Radiation Doses in Switzerland. Bern: BAG; 1991. ch.B.3.6.7.
- [45] Corcho-Alvarado, J et al. Long-term behaviour of ^{90}Sr and ^{137}Cs in the environment. Case studies in Switzerland. *J Environ. Rad.* 2016;160: 54–63.
- [46] Zehring M. Waste water monitoring of the ARA Basel. In: annual report of the state-laboratory Basel-City Basel; 2015. p. 129–131.
- [47] Zehring M. Waste water monitoring at KVA Basel. In: annual report of the state-laboratory Basel-City. Basel; 2015. p. 132–134.
- [48] Rumpel N. Balancing of short-lived radionuclides for medical use in Basel-City [scholarly paper]. Basel: state-laboratory Basel-City. Basel 2015.

- [49] Kammer F, Rumpel N, Wagmann M, Zehringer M. Monitoring of Radionuclides for medical use in Basel. Proceedings of the GDCH-Wissenschaftsforum, Dresden; 2015.
- [50] Rumpel N, Kammerer F, Wagmann M, Zehringer M. Gamma ray spectrometry of sewer sludge—a useful tool for the identification of emissions sources in a city. *Chimia* 2015;**69**: 301.
- [51] Zehringer M. Radioactivity monitoring of the river Rhine. In: Annual Report of the State-Laboratory Basel-City. Basel; 2015. p. 134–138.
- [52] Abraham J. Study of the behavior of radionuclides at an infiltration site for drinking water production. Basel: State-Laboratory Basel-City. Basel; 2016.
- [53] Meyer J. Monitoring of moss in Basel-City for the investigation of radioactive fallout [scholarly paper]. Oberwil: State Grammar School Oberwil; 2014.
- [54] Agapkina G et al. Dynamics of Chernobyl-fallout radionuclides in soil solutions of forest ecosystems. *Chemosphere* 1998;**36**: 1125–1130.
- [55] Zehringer M. Radioactivity in food and other objects from Japan. In: Annual Report of the State-Laboratory Basel-City. Basel; 2011. p. 29–31.
- [56] Zehringer M. Radioactivity in Food: Experiences of the Food Control Authority of Basel since the Chernobyl Accident. In: Monteiro W. editor. Radiation Effects in Materials. InTech Open. 2016. ISBN: 978-953-51-2438-2.
- [57] Palacios M, Estier S, Ferreri G. On the trace of ^{137}Cs in wild boars in Ticino In: Federal Office of Public Health, editor. Environmental Radioactivity and Radiation Doses in Switzerland. Bern: BAG; 2013. p. 100–101.
- [58] Swiss wild boar meat: radioactivity and heavy metal content. In: Annual Report of the State-Laboratory of Zurich. Zurich; 2014. p. 71–72.
- [59] Zehringer M. Radionuclides in siliceous and healing earths. In: Annual Report of the State-Laboratory Basel-City. Basel; 2008. p. 79–82.
- [60] Choppin G, Rydberg J. Nuclear Chemistry—Theory and Applications. New York: Pergamon Press; 1980. ISBN: 0-08-023826-2. p.593 Appendix H.
- [61] Buchillier T, Baillat C, Laedermann J, Leupin A. Rapport sur l'exercice d'analyse de spectre de centrale nucléaire 2013; Rapport interne de l'Institut de Radiophysique Appliqué, Lausanne, Switzerland, 2015.
- [62] Demongeot S et al. A practical guide for laboratories measuring radionuclides at post-accidental situations. IRSN (Institut de Radioprotection et de Sûreté Nucléaire), Rapport No 2011-02, 2011.

- [63] Zehringer M, Abraham J, Kammerer F, Sylva V, Wagmann M. Fast survey of Radiostrontium after an Emergency Incident involving Ionizing Radiation. *Chimia* 2016;**70**: 816.
- [64] Federal Commission for ABC-protection. Concept for emergency protection in the vicinity of NPPs, KomABC 2006-03-D; Berne: 2006.

All references concerning the reports of the State Laboratory Basel City are on the internet, available at: <http://www.kantonslabor.bs.ch/berichte.html>.

