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Radioactivity in Food: Experiences of the Food Control Authority of Basel-City since the Chernobyl Accident

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Additional information is available at the end of the chapter

Abstract

The contamination of our environment and of food with artificial radionuclides originates from several sources. First, nuclear powers spread contamination all over the Northern Hemisphere by carrying out more than 600 atmospheric bomb tests from 1945 to 1963. The peaceful use of nuclear fission brought several accidents in nuclear installations [nuclear power plant (NPP)]. This began in the late 1940s and ended recently with the NPP's core meltings at Fukushima-Daiji in 2011. The catastrophe at the Chernobyl NPP in 1986 spread enormous fallout over most parts of Europe. Besides the artificial contamination, one has to mention the exposure to naturally occurring radionuclides from the uranium and thorium decay series. From 1980 on, the State Laboratory Basel-City began a monitoring programme of food. Special equipment for the analysis of α-, β-, and γ-emitting radionuclides had to be built. In 1986/1987, the laboratory had to manage thousands of samples according to the accident at Chernobyl. The Government estimated the dose of the mean Swiss population from the ingestion of contaminated food to be 1 to 2 mSv. Today, the contamination of food has lowered significantly. The Office of Public Health estimated the total ingested dose of about 0.3 to 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 the bomb fallout is less than 0.1 mSv/year.

Keywords: Chernobyl, dose estimation, food, radioactive contamination, radiocaesium, radiostrontium

1. Introduction

In the 1950s, Otto Huber and his team from the University of Fribourg started the regular monitoring of radioactivity in Switzerland [1]. Some years later, the federal government

initiated a countrywide monitoring programme. In 1969, the first nuclear power plant (NPP) of Beznau came on line to produce electric power. Today, five NPPs are producing about 38% of the electric power in Switzerland. In addition to the emission controls of the NPPs and other radiation-producing facilities, the monitoring of radioactive fallout from the atmospheric, nuclear bomb tests is of special concern. Over 600 bomb tests in the atmosphere led to a contamination of the Northern Hemisphere with long-lived radionuclides, such as radiocaesium, radiostrontium, and plutonium. The contamination situation was then aggravated by the reactor fire of the NPP of Chernobyl in late April of 1986. In Switzerland, the southern and eastern parts were more affected (in southern Switzerland, it rained on 3–5 May over 350 mm precipitation). On 5 May, the National Emergency Operations Centre (NAZ) started a monitoring programme for food with the help of the specialised laboratories in Dübendorf, Freiburg, Lausanne, Spiez, Würenlingen, and the State Laboratory Basel-City. In October 1986, a symposium on the measurements and their interpretation was held in Berne [2]. In 1987, other state laboratories took part in the monitoring programme. Five years later, when radiation levels were reduced considerably, most of the state laboratories reduced their monitoring programmes again; many state laboratories even cut off their survey activities. Until 2011, the radioactivity survey was mainly supported by the specialised laboratories. The core meltings of the Fukushima-Daiji NPPs gave a short increase in the survey activities for the years 2011/2012 in Switzerland.

2. Legislation of the radioactivity controls in Switzerland

In Switzerland, the radioactivity survey is a task of the Federal Office of Public Health (BAG), an office of the Federal Department of Home Affairs. One aspect is the monitoring of food, which is done in collaboration with the state laboratories of the cantons. In 1991, the Federal Assembly of the Swiss Confederation enforced the *Radiological Protection Act* [3] along with several Ordinances (e.g., the *Radiological Protection Ordinance*) [4]. The assessment of food is regulated in the *Ordinance on Contaminants and Constituents in Food* from 1994 (**Table 1**) [5]. This ordinance regulates the most important groups of radionuclides in a special way. For artificial radionuclides, two limits are set. The "tolerance values" should not be exceeded when food is grown or produced with good manufacturing practise. When values are over the tolerance limit, the food is considered as "reduced in its value". "Limit values" are of toxicological concern. When they are exceeded, the consumption of this food may lead to a dose of more than 1 mSv/year.

In emergency situations, when radioactive contamination may occur, special limit values are formulated. After the accident of Chernobyl, such special conditions were formulated for imports from East European countries [6]. Each importation of wild grown mushrooms has to be documented by an importation certificate, which documents a radiocaesium level below 600 Bq/kg. In 2011, the Federal Food Safety and Veterinary Office enforced precautions for the importation of Japanese food and feed origin from animals. Importations from specific prefectures have to be accompanied by a certificate of a radiocaesium analysis of the food [6, 7]. The Federal Office of Public Health will implement revised ordinances for Japan and

Chernobyl in the next few years. Even, the Government intends to annul most of the limit values for radionuclides in times of non-crisis but to implement special limit values for crises.

Radionuclides	Babyfood, infant formulas	Liquid food	Food of main importance	Food of minor importance
Radiostrontium $^{89+90}$Sr	1/75 75	1/125 125	1/750 750	1/7500
Radioiodine $^{131+132}$I	10/150 100	10/500 300	10/2000 2000	10/20,000
Radiocaesium $^{134+136+137}$Cs	10/400 200	10/1000 200	10/1250 500	10/20,000
Plutonium and trans-Pu-elements	0.1/1 1	0.1/20 1	0.1/80 10	0.1/800
Natural radionuclides 1 Σ ^{224}Ra, ^{228}Th, $^{234+235+238}$U	—/10	—/10	—/50	—/500
Natural radionuclides 2 Σ ^{210}Pb, ^{210}Po, $^{226+228}$Ra, $^{230+232}$Th, ^{231}Pa	—/1	—/1	—/5	—/50
Tritium	1000/3000	10/1000	1000/10,000	1000/100,000
Radiocarbon	200/1000	—	200/10,000	200/100,000
Others	10/400	10/1000	10/1250	10/12,500

Except for Brazil nuts (no limits).
Special tolerance and limit values for game: 600/1250 Bq/kg and wild berries: 100/1250 Bq/kg.
Special limit value for ^{210}Po in seafood: 150 Bq/kg.
All values in Bq/kg. First value of each row: tolerance value; second value: limit value. Second line: limits of the ordinance for imports of food from or origin of Japan (2011) [7].

Table 1. Ordinance on contaminants and constituents in food (1995). Appendix No. 6: "List of limit values for radionuclides" [5].

3. Radiation in the environment

Radioactive sources are in our body and everywhere outside in the environment. They can be divided in two groups of sources: naturally occurring radionuclides, which are part of naturally occurring radioactive materials (NORM), and technologically enhanced naturally occurring radioactive materials (TENORM). In addition, there are man-made (artificially produced) occurring radioactive materials.

Primordial radionuclides are radionuclides with long half-lives, even longer than the earth's age. They play an important role in the earth's radioactivity. The three natural decay series of

Uranium (^{235}U, ^{238}U) and thorium (^{232}Th) belong to them. Secondary radioactive elements are built by the decay of these primordial radionuclides and belong to the inventory of our soils. Some dose-relevant radionuclides of these decay chains are radium (^{224}Ra, ^{226}Ra, and ^{228}Ra), radon (^{220}Rn and ^{222}Rn), lead (^{210}Pb), polonium (^{210}Po), and thorium (^{228}Th, ^{230}Th, and ^{232}Th). Another ubiquitous radionuclide is potassium-40 (^{40}K), Which is a large part to the annual received dose. Cosmogenically produced radionuclides are, for example, beryllium-7 (^{7}Be), tritium (^{3}H), or radiocarbon (^{14}C) [8, 9].

Artificial radionuclides have been produced and released to the environment since the 1940s, when nuclear weapons development and tests began in the United States. These tests of nuclear fission in the United States, USSR, China, GB, and France led to fallout with a great number of radioactive fission products and activated radionuclides. Over 600 atmospheric bomb tests from 1945 to 1963 led to a contamination of the Northern Hemisphere with artificial radionuclides such as ^{14}C, ^{3}H, radiocaesium (^{134}Cs and ^{137}Cs), radiostrontium (^{90}Sr), and plutonium (^{239}Pu). [10]. It is because of scientists, such as Ernest Sternglass, who investigated and proved the negative effects of the bomb fallout on childhood mortality. Their warnings helped to enact the partial test ban treaty,[1] which most nuclear powers ratified [11]. After 1965, the fallout clearly decreased. One can clearly see this in sediment profiles of lakes (e.g., lake sediment investigations in Switzerland) [12].

NPPs are also emitters of artificial radionuclides. Several accidents caused the release of large quantities of radioactive fallout to the environment. Accidents were the burn of one pile at the Windscale reactor in Sellafield in 1957, the partial core melting of the Three Mile Island reactor in Harrisburg in 1979, the nuclear catastrophe of the Chernobyl NPP in 1986, and the core meltings of the NPPs of Fukushima-Daiji in Japan 2011. The catastrophe of Chernobyl affected many European countries, many thousands of miles away from the NPP ground.

The use of radionuclides in diagnoses and therapies against cancer leads to the release of short-lived radionuclides, such as iodine (^{131}I), technetium (^{99m}Tc), indium (^{111}In), lutetium (^{177}Lu), yttrium (^{90}Y), and others. They do not enter the food chain because of their short half-lives and are therefore of minor concern.

4. Radiation detection

The following overview of radioactivity laboratory equipment for food control is only a brief summary of the required equipment. For information about the theory of radiation detection and measurements, technical details, or special applications, we refer to the standard literature [13–16].

4.1 Dose rate monitors

Measurement devices to count the dose rate of radioactive sources are not sensitive enough for the precise contamination measurements of food. However, they can be adequate for

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[1] Treaty banning of nuclear weapon tests in the atmosphere, in outer space, and under water. 5 August 1963.

screening analysis. In 2013 and 2014, the State Laboratory Ticino and the Federal Office of Public Health used dose rate monitors to screen wild boars shot in the southern parts of Switzerland. They only sampled animals over a certain dose rate for precise γ-spectrometric analysis for radiocaesium [17]. This kind of measurement equipment is not sensitive enough to detect contamination in the case of food imports from Ukraine, Japan, and other contaminated countries. Therefore, one cannot save the counting on a γ-spectrometer.

4.2. γ-Spectrometry

It is of primary importance to be equipped with a γ-ray detector. γ-Spectrometry can be operated with inorganic scintillators, such as pure crystals of sodium iodide (NaI) and caesium iodide (CsI) or doped crystals, such as NaI(Tl) or CsI(Tl). NaI(Tl) is the most used scintillator and has excellent light yield and a good linear response, but energy resolution is quite limited. Today, the best choice is semiconductor detectors. Crystals of silicon, germanium (Ge), cadmium-telluride, and others are the detector materials. Ge detectors have been widespread since 1980, when the production of high-purity Ge monocrystals became possible. They have to be kept under vacuum and cooled with liquid nitrogen. The detectors have an excellent energy resolution. Therefore, even complex spectra can be analysed without prior chemical separation steps. Two criteria are of importance: resolution and sensitivity. Energy resolution of Ge detectors is excellent: 1.5 to 2.5 keV at 1.33 MeV, below 100 keV, less than 1 keV FWHM (full width at half maximum). The efficiency of the detector (relative efficiency compared to a NaI detector at 1.33 MeV) and the relation of the peak to Compton background are the most important factors for the sensitivity. Today, Ge detectors with efficiencies of 25% and higher are available. The peak/Compton quotient is more than 46. In our laboratory, we use Ge detectors with 50% efficiency. Important factors for quantitative γ-spectrometry are the shielding of the detector and the efficient suppression of the electronic noise of the amplifier system. γ-Spectrometry has the advantage that γ-radiation can be measured without the elimination of the matrix. Therefore, sample preparation takes only a short time. γ-Spectrometers need an exact calibration over the whole energy range (e.g., 50–2000 keV). Calibrations with certified radioactive sources are necessary for every counting geometry used (volume and shape of the sample, and distance from the detector) Calibration solutions consist of a mix of short-lived radionuclide with emission lines over the whole energy range (e.g., ^{109}Cd, ^{57}Co, ^{113}Sn, ^{137}Cs, ^{88}Y, and ^{60}Co). After 1 year, the short-lived radionuclides are partly disintegrated; therefore, the calibration mix cannot be used anymore. This can be overcome using mixtures of ^{152}Eu (half-life of 13.5 years) in combination with a low-energy γ-nuclide, such as ^{210}Pb or ^{241}Am. When using ^{152}Eu, summation effects have to be corrected properly.

There are other important factors to consider besides the counting geometry. The sample matrix itself absorbs γ-rays before they arrive at the detector. These self-absorption effects are a function of the sample geometry and the elemental composition and the density of the sample matrix. Normally, calibrations and efficiency curves are produced with calibration standards in water or gels of density 1. Density corrections can be calculated for every material and geometry with means of a software based on Monte Carlo simulations. The background radiation has to be considered. Some background radiation remains, even with good shielding

with lead and copper. This background consists of radionuclides of the natural decay series, such as [214]Pb and [214]Bi and others. For every counting geometry, the background has to be measured with water-filled containers of the needed counting geometries. The γ-spectra have to be subtracted by the specific background spectrum. Radionuclides with cascade emissions show coincidence summing effects (e.g., [134]Cs and [152]Eu). Spectra have to be corrected or the measurement must be repeated with a distance between sample and detector. For short-lived radionuclides, their partial decay has to be corrected to the reference date (e.g., the date of the sampling). Further advice and descriptions over quantitative γ-spectrometry are given in the literature [18].

4.3. β-Spectrometry

For the counting of β-rays, the sample matrix has to be eliminated. An exception is water samples (e.g., the measurement of ^{3}H needs not much sample preparation, only the mix of the water sample with a scintillator cocktail). Some important β-nuclides, such as ^{3}H, ^{14}C, ^{89}Sr, ^{90}Sr, and ^{90}Y, can be analysed with scintillation counting. Commercially available scintillation counters can detect α- and β-decays. This widens the spectrum of radionuclides (e.g., [222]Rn can be analysed in water samples or in charcoal air samples). When samples are analysed directly, the sensitivity is given by the small sample amount of typically some millilitres.

Radiostrontium, ^{89}Sr and ^{90}Sr, are important fission products. One possibility is to extract the ^{90}Sr with the use of specific crown ethers from the sample. In our laboratory, we have developed a fast analysis scheme for water samples [19]. Another possibility is to clean up extracts over a column filled with crown ethers. These methods are suitable for activity concentrations higher than 1 Bq/kg. For sensitive analyses, the β-spectrometers of choice are gas flow proportional counters. We use this technique for the analyses of ^{90}Sr traces in food, human, and environmental samples. The method is based on the counting of the daughter nuclide, [90]Y. Before the counting starts, a rigorous elimination of the matrix and disturbing β-nuclides, such as [40]K, is necessary. With an oxalate precipitation step, most of the [40]K is eliminated. Then, ^{90}Y is separated from ^{90}Sr by precipitation as hydroxide. The $Y(OH)_3$ is dissolved and precipitated as $Y_2(oxalate)_3$. These β-sources are pure enough for counting. Counting is performed in 10 consecutive runs, as ^{90}Y decays during the counting (half-life is 64 h). A good-quality criterion for the purity is the measured decay of the source. Decay should be near 64 h. When decay is slower, impurities are present. The conserved ^{90}Sr solution may be prepared and analysed again after 20 days (the built-up ^{90}Y will then arrive equilibrium with ^{90}Sr). Quite sensitive analyses may be performed down to 10 mBq/kg. Counting time is 3 days. Therefore, several detectors should be available. Our Canberra α/β-counter LB 4100 can take up to four drawers with four sample holders each [20].

4.4. α-Spectrometry

Like β-spectrometry, α-spectrometry requires an elimination of the sample matrix. Only water samples need a minimal preparation. Two counting techniques are common today: scintillation counting and passivated implanted planar silicon (PIPS) detectors. We use liquid scintillation counters in our laboratory for the analyses of uranium, thorium, radium, and polonium.

According to the methods published by W. Jack McDowell, the water sample is extracted once with some millilitres of a nuclide selective extractant, which contains the scintillator cocktail for the α-analysis. Very low activity concentrations (5–10 mBq/L) can be achieved [21]. The disadvantage is the poor energy resolution. Also, α/β-discrimination has to be set carefully. Analysis time is 24 hours for water samples containing 100 mBq.

Analyses with PIPS detectors show good energy resolution and efficiencies from 20% to 30%. Counting has to be performed under vacuum and very thin layer α-sources are needed. The sample matrix has to be destroyed (by ashing, etc.) following a clean-up (specific nuclide extraction, scavenging, etc.) and a preparation of a thin-layer source by means of electroplating or coprecipitation. Many actinides, such as uranium, thorium, and plutonium, may be analysed (well described in [22]). Some elements such as [210]Po and radium nuclides may be auto-deposited onto special surfaces. [210]Po is auto-deposited under reductive conditions onto silver or copper disks after a microwave digestion of the sample [23, 24]. Radium nuclides ([224]Ra and [226]Ra) are auto-deposited onto MnO_2 surfaces at pH 8 as has been shown by Surbeck [25, 26]. These methods are suitable for drinking water and mineral water analyses. Some new developments were done for uranium and thorium analyses in honey and spices [27, 28] (**Figure 1**).
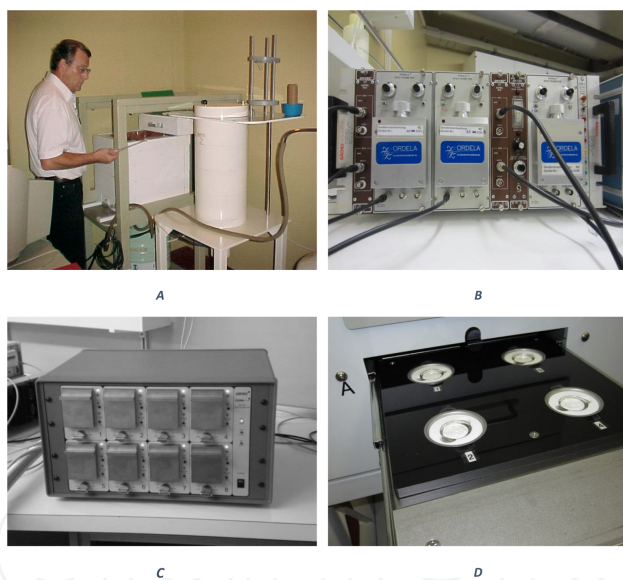

Figure 1. Equipment of a radioactivity laboratory: (A) γ-spectrometry; (B) α-liquid scintillation (PERALS); (C) α-Spectrometry (PIPS detectors); and (D) β-spectrometry (gas proportional counter), drawer with ^{90}Y oxalate sources.

4.5. Neutron activation analysis (NAA)

INAA (instrumental neutron activation analysis) is based on the production of radionuclides by nuclear reactions. Thermal neutrons can activate many elements. The efficiency of this irradiation process depends on the flux density of the neutrons and the cross-section of the nuclear reaction. The thermal neutrons are generated in a nuclear reactor, for example, at the University of Basel (AGN-211-P, a light water moderated swimming pool reactor).

The samples (1–5 g material) are inserted into the core over a cannula through the so-called glory hole and irradiated for 30 min with a power rate of 2 kW. After a cooling time of some hours, the samples are counted on an HPGe detector.

We used INAA for the analysis of total bromine content. Bromide is built by the decay of the fumigant methyl bromide. We used this technique for many years to determine the total bromine in spices, tea, and dried mushrooms. Another application is the determination of the total bromine content as a screening analysis for flame-retardants in plastic materials [29]. ^{238}U and ^{232}Th can be determined by INAA in suspended matter and sediments [30]. In addition, total iodine content in iodine rich food, such as algae, can be determined over the activation of ^{127}I to ^{128}I, which decays to ^{128}Xe [31].

We mentioned the use of INAA as a completion of the possibilities of γ-spectrometry. These applications will not be discussed further, because the analytes are not radionuclides.

5. Results from 35 years of food control

In Basel, the Government decided to buy the equipment for the monitoring of β- and γ-nuclides in 1980 because of the NPP accident at Harrisburg in 1979. Therefore, we were the only state laboratory that was prepared when the accident at Chernobyl happened. In 1986 and 1987, thousands of samples were analysed.

The fallout from bomb tests and the NPP accident of Chernobyl resulted in the ubiquitous contamination of the landscapes of the Northern Hemisphere. The core meltings at the NPP of Fukushima-Daiji also reached Europe, but the fallout was considerably lower than from Chernobyl. The situation after the Chernobyl fallout in Switzerland is well described in some papers [31–33, 98].

The contamination of farmland leads to contaminated food, such as milk or vegetables. These matrices are an important part of the Swiss survey programme. The fallout from Chernobyl affected the regions of southern Switzerland the most (total rain of 350 mm). This has to be compared to the washout in eastern Switzerland with 150 mm and the regions of Basel and Jura with 50 mm rain.

5.1. Milk and milk products

In 1981, the State Laboratory Basel-City started the first regular radioanalyses in Basel. Milk samples from local milk production centres in northwest Switzerland of the states (cantons) of Aargau, Basel-Campaign, Basel-City, Solothurn, and Jura were analysed with β- and γ-spectrometry. On 2 May 1986, after the accident in Chernobyl, the frequency of the survey was intensified. On 4 and 5 May, radioiodine activity concentrations between 220 and 650 Bq/L ^{131}I were measured. The milk from the mountainous region of the state of Jura showed lower values than in the states localised in the plain: 178 Bq/L ^{131}I (4 May). In the milk distribution centres of Basel-City, the milk from the different regions had to be mixed in such a way that the population received milk with activities below 350 Bq/L. Due to its short half-life, the

activity of radioiodine fell under the detection limit (0.1 Bq/L) in July. The activities of caesium and strontium were quite lower but resisted longer. The activity line of total caesium in the milk of the state of Jura (**Figure 2**) shows a maximum value of 127 Bq/L. In the following 3 years, smaller peak values could be observed due to the fact that the cows were fed with contaminated hay from the year before. After 1990, the total caesium level fell below 1 Bq/L, except for some farms in southern Switzerland, where, even in 2013, the radiocaesium level of the milk of one farm was over the tolerance limit of 10 Bq/kg.
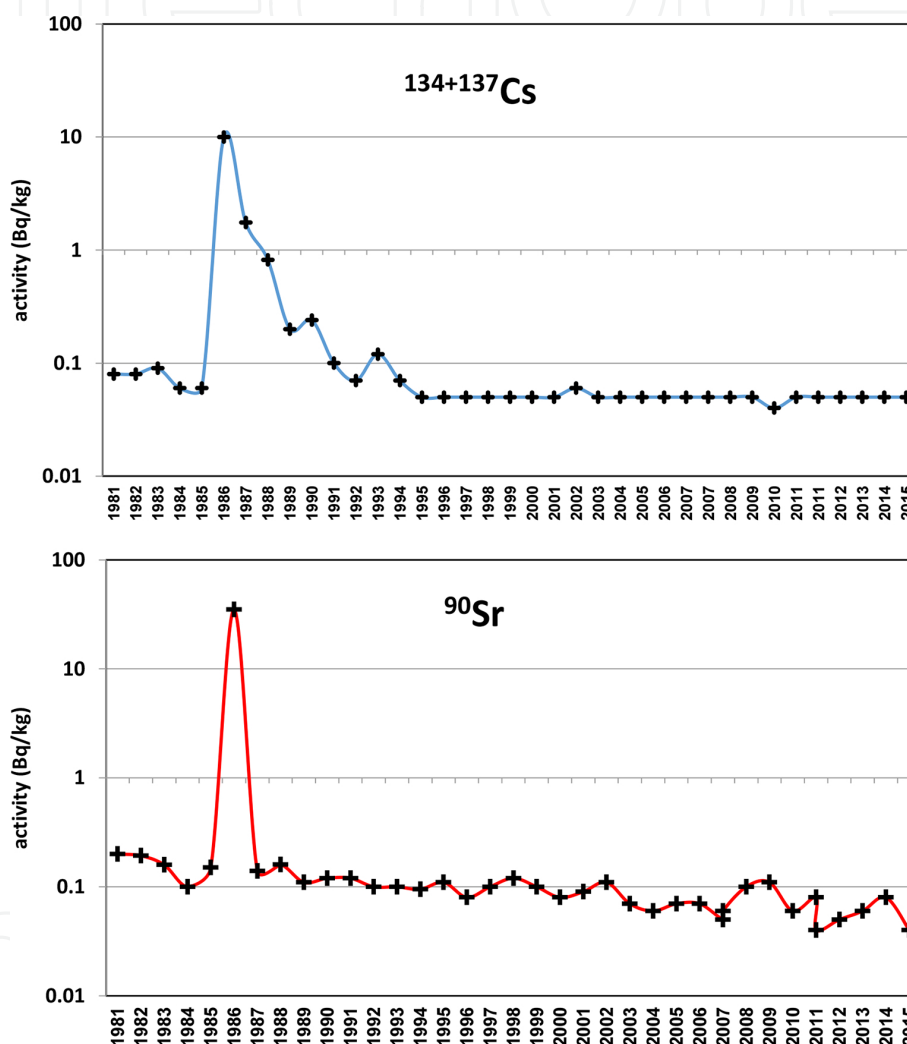

Figure 2. Results of the monitoring of milk from northwest Switzerland. Notice the sharp peaks due to the Chernobyl fallout. The ^{90}Sr activity concentration shows a steady decline from 1981 to 2015.

We found much lower activity of ^{90}Sr with a maximum value of 4.4 Bq/L on 11 March 1987 in Jura. In the Swiss Alps, the level reached 35 Bq/L. After 1988, the contamination of the milk from the canton of Jura reached a value of approximately 0.1 Bq/L. The present contamination of Swiss milk is highest in the Alpine regions of the states of Grison and Ticino, where the radiostrontium level is a factor 10 higher than in the rest of Switzerland (0.1–0.4 Bq/L), and radiocaesium reaches 9 Bq/L for the highest value (0.3–9 Bq/L) [34, 35].

Many milk products were analysed in 1986: Swiss cheese, milk powder, butter, yogurt, and cream [34, 36]. These investigations showed the same trends in somewhat lower activity concentrations. Two drinks also showed higher levels of radiocaesium (22–96 Bq/L in 1987), as they contained milk serum. The same contaminated milk from 1986 was used to produce milk powder. This milk powder was used for the production of chocolates. Milk chocolate contains 20 to 25 g milk powder (160–200 mL milk) per 100 g chocolate. We analysed the first samples in autumn 1986 for a local chocolate producer. Soon, we noticed higher levels when chocolates contained hazelnuts. These chocolate samples were in the range of 55±13 Bq/kg radiocaesium. In contrast, chocolates without nut ingredients showed lower activities (12±3 Bq/kg). These investigations showed that the contamination level of chocolates is even more from the use of hazelnuts (60%) than from the milk (~40%). In the following year, we found even higher radiocaesium values: 710 Bq/kg in chocolates without hazelnuts and 1.3 kBq/kg in chocolates with hazelnuts. Even chocolates without hazelnuts showed higher values. We explained this with higher values in milk. Hence, we started a special survey programme for hazelnuts.

5.2. Wild-grown vegetables

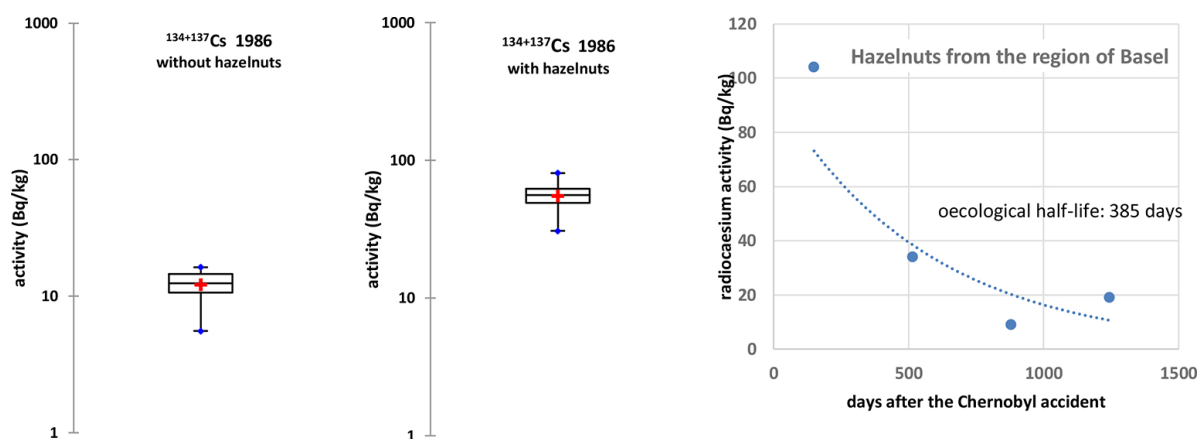### 5.2.1. Hazelnuts and other nuts

Figure 3. Activity concentrations of radiocaesium in hazelnuts. Plots to the left: Comparison of the radiocaesium level in chocolates with and without hazelnuts (1986). Plot to the right: Development of the activity in hazelnuts from Basel.

The investigation of chocolates containing hazelnuts showed that the latter were part of the contamination. In 1986 and 1987, more than 180 hazelnut samples from a chocolate producer of the region were analysed for radiocontaminants. In 1987, the contamination level reached a total radiocaesium activity of up to 17 kBq/kg. Most hazelnuts were imported from Turkey, a country that was seriously contaminated with fallout from Chernobyl. Since 1989, we also investigated different nuts from other countries. Hazelnuts from Turkey remained the only contaminated nut species. Even in 2007, 21 years after the catastrophe of Chernobyl, one hazelnut sample had to be rejected. It did not reach the limit value but was over the tolerance

value of 10 Bq/kg. After 1990, the contamination of hazelnuts in Switzerland and abroad remains under 10 Bq/kg; meanwhile, in Turkey, the contamination level was reduced much more slowly due to the higher contaminated soils (**Figure 3**).

5.2.2. Brazil nuts

Brazil nuts (*Bertholletia excelsa*) are known to accumulate earth alkaline metals from soil. First, this was shown with barium. Then, Penna-Franca detected the radium nuclides ^{226}Ra and ^{228}Ra, both from the natural decay series of uranium and thorium, in amounts of 50 Bq/kg. In Switzerland, imported Brazil nuts showed 61 to 112 Bq/kg total radium. This means 1000-fold of the radium content of the total diet in Europe. The consummation of Brazil nuts is estimated to be 0.1 g/day and person. Therefore, the consummation of Brazil nuts is not relevant. In literature, it is advised to consume Brazil nuts to enhance the selenium level. Two Brazil nuts lead to a yearly dose of 160 μSv, which is relevant [8].

Another important alkaline earth metal is strontium. Its most important radioactive nuclide is ^{90}Sr. Froidevaux et al. [37] measured 11 to 15 Bq/kg in imported Brazil nuts. As Brazil nuts are often part of nut mixtures, the measured natural radioactivity is mainly produced by the Brazil nuts contained in the mixture.

5.2.3. Mushrooms

Mushrooms are known to be organisms that can accumulate heavy metals from soil. Therefore, mushrooms from abroad and from Switzerland were of concern in 1987. More than 200 samples were analysed in our laboratory for radiocaesium. 24 samples had to be rejected. They showed a contamination level of more than 600 Bq/kg. The range of all analysed mushrooms was from 107 to more than 8000 Bq/kg. Swiss mushrooms and mushrooms from abroad then did not show any significant difference in their contamination level [38]. From 1989 on, mushrooms were regularly analysed, but violations became rare.

A quite different situation could be observed in South Bavaria, Germany. At the end of April 1986, much Chernobyl fallout was washed out in southern Bavaria over several days. In Munich, the local dose rate increased up to 1.1 μSv/h (10-fold over the normal value). Therefore, the contamination level in wild-grown mushrooms and berries reached high values (five-fold the normal radiocaesium activities due to the bomb fallout). Mushrooms with mycorrhiza, such as boletus species (*Hydnum repandum* and *Boletus badius*), blueberries, mosses, and lichens, were most affected [39–41]. Other mushrooms, such as porcini, champignons, and chanterelles, did not show this effect. In 2002, bay boletes (*B. badius*) showed radiocaesium levels of 33 to 23,200 Bq/kg and porcini from 6 to 10,000 Bq/kg. In 2015, activities were reduced remarkably in boletes 13 to 900 Bq/kg and porcini 4 to 108 Bq/kg [39]. Similar values were reported in Austria [42] and Switzerland [43]. Even in 2015, bay boletes were found with maximum activities of more than 2600 Bq/kg, whereas porcini showed a reduced contamination level with a maximum of 416 Bq/kg. The reduction was slow compared to West European countries. For example, in Spain, values ranged from 1 to 122 Bq/kg for radiocaesium and from 0.2 to

3.5 Bq/kg for radiostrontium. Mushrooms of a special region in the canton of Aargau (Switzerland) show a continuous fall of the activity from 1986 to 2014 [44].

Wood soils build their own biosphere: dead leaves, pins, etc., build the humic fraction of the soil. The plants take up the radioactivity from the soil. The radioactivity is recycled to the soil through the fall of the leaves and pins.

5.2.4. Wild-grown berries

In 1986, the State Laboratory Basel-City analysed approximately 30 berry samples from Switzerland. Strawberries showed a quite low activity (2.7 Bq/kg radiocaesium), whereas currants, raspberries, elderberries, and gooseberries showed significantly higher activities (51±27 Bq/kg). The reason is that strawberries are cultivated in greenhouses, in the shelter. Only in 2009 was our focus set on blueberries and other wild-grown berries. These products are imported in big charges of tons from East European countries, such as Ukraine, Poland, Russia or Hungary. From 16 analysed samples, we registered eight violations because of too high activities of radiostrontium. In the following years, we analysed more than 100 samples with 10 further violations for products from Austria, Ukraine, or Poland. The products were blueberries and blueberry products, such as marmalades. Violations were mostly because of radiostrontium (>1 Bq/kg) and radiocaesium (>100 Bq/kg) [45]. Wild berries grow in woods. The soil is more acid and promotes the uptake of the contaminants. Because of the cycle soil-plant-soil, the residues in the plants are reduced only slowly compared to berries grown on farmland.

5.3. Vegetables and fruit

5.3.1. Vegetables

Leafy vegetables were, besides milk, the most contaminated food. Depositions from washouts on spinach and salad cultures happened just before harvest. Radioiodine level reached almost 4000 Bq/kg in spinach. Therefore, it was advised that small children, pregnant women, and nursing women should not consume this kind of food. The investigation of mother's milk showed considerable amounts of radioiodine (up to 35 Bq/L). We tried to reduce the contamination on salads with washings but with only poor success. In May 1986, leafy vegetables were most affected by radioiodine (^{131}I and ^{132}I): radioiodine was 65% of the measured total dose. Radiocaesium (^{134}Cs and ^{137}Cs) was 25% and other short-lived radionuclides, such as ^{103}Ru, ^{140}Ba, ^{140}La, and ^{99}Mo, gave approximately 10% to the total dose. We noticed many violations of the limit values for radioiodine and radiocaesium (24 and 10 violations for baby food from a total of 40 samples analysed). This survey focused on food with higher radiation levels as a screening before the γ-analyses were done. The investigation of deep-frozen vegetables did not show any elevated radiation. These products were produced before 1986 [46]. After 2010, we started up again with controls of leafy and root vegetables. The contamination level was low and caused by the fallout from the bombs. Radiocaesium is normally below the detection limit of 0.1 Bq/kg. Radiostrontium is detectable in small amounts of 0.05 to 0.5 Bq/kg [47].

5.3.2. Herbs

Herbs of 1986/1987 showed the highest contamination with radiocaesium of all investigated food. In 1986, 35 of 42 herbs contained up to 3.200 Bq/kg. In the following year, the contamination level was even higher: 12.300±32.000 Bq/kg [38].

5.3.3. Fruit

In 1986, fruit was less contaminated than vegetables. When harvesting time arrived, the radioiodine had already disintegrated. The radiocaesium level was below 100 Bq/kg. In 1990, no more contamination was found (<2 Bq/kg radiocaesium in dried fruit) [46].

5.3.4. Chestnuts

Chestnuts, the fruits from the chestnut tree, *Castanea sativa*, are cultivated in northern Italy and southern Switzerland. At harvest time, in autumn, they are roasted and eaten or processed to crèmes or purees. These were also the regions with high fallout and washout from Chernobyl. The soils and vegetation had elevated activities of radiocaesium. As a consequence, chestnuts contained elevated radiocaesium levels. In 1986, a level of 265 Bq/kg was reached. In the following years, the contamination was reduced but only slowly. Even in 1988, one sample violated the tolerance value of 10 Bq/kg. In 2005 and 2015, we again investigated chestnuts and chestnut products. The radiocaesium level was reduced by a factor of 10. The resulting level was approximately 1 to 4 Bq/kg. No more violations were noticed [48].

5.4. Flour, bread, and biscuits

One year after Chernobyl, corn, grains, bread, shortbreads, and pasta were seriously contaminated with radiocaesium. In 1987, cereals from 1986 were taken into production. Seven of 17 breads and shortbreads and one pasta were over the limit value of 1250 Bq/kg and had to be withdrawn. Twenty-one more samples were over the tolerance level of 100 Bq/kg. As a consequence, the control of flour and flour products was intensified over the next years. However, with the exception of one sample in 1988, no more violations of the tolerance value were noticed. In 2011, the radiation level fell down to approximately 0.5 Bq/kg radiocaesium in cereals.

Natural radionuclides are present in cereals (radium 1–5 Bq/kg and thorium 0.1–1 Bq/kg). The levels of radium are near the limit value of 5 Bq/kg.

5.5. Meat

Our own γ-analyses of meat in 1986 showed values in two categories: for cows and calves, the level was low (57±71 Bq/kg), whereas meat from sheep and lamb was quite higher contaminated (645±744 Bq/kg radiocaesium). In the following year, the radiocaesium levels were elevated to the same level: sheep and lamb 550±198 Bq/kg and cow and calf 417±384 Bq/kg. During 1986, the Federal Food Safety and Veterinary Office (former Federal Office for Veterinary Affairs) analysed more than 1700 meat samples of sheep, goat, cow, pig, and game

mainly from eastern and southern Switzerland. The contamination levels in the meat were higher in regions with higher depositions of fallout. In southern Switzerland, the radiocaesium activity concentrations varied from 40 to 4400 Bq/kg. The highest value was found in a goat. In contrast to this, in eastern Switzerland, values were from 40 to 1300 Bq/kg. In the other parts of Switzerland, even lower values were found. Almost 2000 samples from imported meat were analysed. Here, 251 samples had to be rejected due to values more than 600 Bq/kg. During the year 1986, a reduction of the contamination level was observed, with the exception of southern Switzerland (cantons of Ticino and Grison). The biological half-life for radiocaesium was calculated to be approximately 50 days for sheep and goat and approximately 30 days for game. Pigs showed a half-life of 90 days [49].

5.6. Game

Game became of interest when high radioactive contaminations of reindeer were reported in Norway and Sweden. The northern European countries were more highly contaminated with radioactive fallout from Chernobyl than other European countries. Wild berries, mushrooms, and lichens are the main food of reindeers. These were seriously contaminated with this fallout. Thousands of animals had to be burnt because of a violation of radiocaesium values that were too high. The survey of game in Switzerland began in autumn 1986. We analysed meat from five roe deer and deer with γ-ray spectrometry. The activities were not high. Nevertheless, in 1987, the contamination of the game meat showed higher values up to 7 kBq/kg radiocaesium. From 1986 on, game was investigated yearly. In 1990, four objections had to be executed according to our measurements. It seemed that chamois were the most contaminated game in Switzerland. After 1995, the monitoring programme was reduced. The detected radioactivity was under the tolerance limit of 600 Bq/kg.

Wild boars are an exception. The southern parts of Switzerland, such as in Bavaria (Germany), are more contaminated landscapes. Here, the contamination of wild boar remains a problem up to today. Wild boars search for their food on the ground. Elaphomyces, a truffle species, grow underground and are able to enrich radiocaesium from soil. It is estimated that these fungi can be up to 20% to 30% of the food of wild boars. In 2012, in southern Bavaria and the Bavarian Wood, the contamination levels were up to 9.8 kBq/kg wild boar and up to 430 Bq/kg in roe deer [50]. In Switzerland, the State of Ticino, in collaboration with the Federal Office of Public Health, investigated wild boars in 2013 and 2014. In 2013, 28 wild boars of a total of 470 animals violated the limit value of 1250 Bq/kg. In 2014, they found 13 such contaminated animals. These animals had to be confiscated [17]. In contrast, in the State of Zurich, the State Laboratory Zurich found no violations when they analysed 80 wild boars. The mean activity was low with 28 Bq/kg radiocaesium [51]. In 2014, the Umweltinstitut München reported from their monitoring programmes for wild-grown vegetables and game. More than 2000 samples showed contamination levels over the limit value of 600 Bq/kg, and 141 samples showed radiocaesium activities of more than 10,000 Bq/kg. Ten samples contained more than 16,000 Bq/kg. The maximum value was 27,800 Bq/kg [52]. Therefore, the serious contamination rests a problem in Bavaria.

Wild boars have to be surveyed over the coming years. Fortunately, they are not a widely consumed game. More important are deer and roe deer, which show considerably lower contaminations [53, 54].

5.7. Seafood and fish

5.7.1. Fish

In Switzerland, we analysed fish from the Rhine River at Basel for radioactive contamination in 1986. Fifteen species caught by local fishermen contained 22 to 707 Bq/kg radiocaesium. These activities were not alarming, as fish are not an important part of the daily food consumption in Switzerland. Again, the most affected region was the southern part of Switzerland. The mean activity of 70 fish species caught in the lake of Lugano was 1.09±0.6 kBq/kg radiocaesium with a maximum value of more than 4.4 kBq/kg, approximately five times higher than in fish from other lakes of Switzerland. This lake has no major confluences and effluents such as the lake of Maggiore where the contamination level of fish was quite lower [49, 55]. After 1987, the contamination with radiocaesium from the Chernobyl fallout was reduced and reached a level of approximately 0.2 Bq/kg for ^{137}Cs [56].

The accidents at the Fukushima-Daiji NPPs gave us cause to investigate fish importation from the Pacific Ocean. Approximately 90% of the released fallout reached the sea (4–90 PBq ^{137}Cs). Radiocaesium levels in fish reached 200 kBq/kg. In 2011, the Japanese Government banned fishing in the coastal waters near Fukushima NPP and the fishing of fish species in some prefectures, which are severely contaminated. Local, private associations of fishermen voluntarily imposed a limit value for radiocaesium of 50 Bq/kg [57]. Also in territorial waters, such as lakes, ponds, and rivers, fish accumulated radioactivity from fallout. Here, radioactivity levels reached approximately 10 kBq/kg fish. In 2012, an intensified monitoring programme of Pacific blue tunas off the Californian coast showed a slight contamination of the fish (0.7±0.2 Bq/kg ^{134}Cs and 2.0±0.5 Bq/kg ^{137}Cs). The presence of the short-lived ^{134}Cs proves the contamination from the Fukushima fallout [58–61]. A received dose of 1 mSv/year was estimated from the consumption of 50 kg fish caught within a zone 3 km away from the Fukushima NPP. Our own investigations of imported fish from the Pacific show a relatively low contamination level below 1 Bq/kg radiocaesium, with a mean value of approximately 0.3 Bq/kg. Some fish samples also contained the short-lived caesium-nuclide ^{134}Cs [62]. Only 14% of the total dose of the consumption of fish and sea food originates from artificial radionuclides, and 86% is from natural radionuclides, such as polonium (^{210}Po).

5.7.2. Natural radionuclides

Natural radionuclides cause the main radiocontamination of fish and seafood. Mussels and molluscs are known to enrich ^{210}Po in the intestinal tract, whereas the mother nuclide lead-210 (^{210}Pb) is not enriched. Cherry and Shannon [63] published an excellent review. Both radionuclides are part of the natural decay series of uranium and are built at the end of the decay chain. ^{210}Po is a powerful α-emitter with an energy of 5,500 keV and a half-life of 183 days. ^{210}Pb is a

β-emitter with a half-life of 23 years and acts as a reservoir for ^{210}Po. Activity concentrations range from 20 to 100 Bq/kg. In fish, the ^{210}Po level is much lower (1–20 Bq/kg) [64].

Our own investigations on imported seafood in 1998 resulted in 12 objections in mussels and 2 objections in sardines concerning too high levels of ^{210}Po. For food producers and food distribution agents, this was surprising. ^{210}Po was never seen as a problem. One consumes sardines as a whole fish, the intestinal tract included [65, 66]. This explains the higher contamination level of sardines and anchovies. A second survey in 2010 showed values equal to those in 1998. Since 1990, the limit value for ^{210}Po in fish was raised from 10 to 150 Bq/kg (the rate of fish and seafood consumed in Switzerland is of minor relevance). Therefore, since 1998, no more objections had to be raised [56]. A survey of the ^{210}Pb and ^{210}Po contamination of seafood in France over the last 15 years reports the same contamination levels [67]. Even higher ^{210}Po levels were found in anchovy from local fishers at the Turkish coast of the Aegean Sea. The annual dose by ingestion was calculated to be 15 μSv [68]. Low activity concentrations were found in fish caught in Swiss lakes. A mean value of 87 samples was 0.4±0.3 mBq/kg ^{210}Po. Such low values are not astonishing. Only the edible parts of the fish, without the intestinal tract and entrails, were analysed. Measurements of entrails of 34 fish samples showed a mean activity concentration of 25 Bq/kg [69].

5.8. Baby food

Baby food is infant follow-on formula that is industrially produced from cow's milk or soybeans. It is given to children up to 4 months after birth. For this kind of food, more restricted limit values are regulated concerning radionuclides. The given limit values are calculated to the final reconstructed constitution of the food (**table 1**). Radioactive contaminants are introduced through the milk into the products. Therefore, radiostrontium and radium are of special interest.

In 1987, an investigation of 56 samples of follow-on formulas showed a severe contamination with radiocaesium. Four samples exceeded the limit value of 400 Bq/kg; the highest value was more than 5.000 Bq/kg. Ten further samples contained radiocaesium in amounts greater than today's tolerance limit of 10 Bq/kg. However, no radiostrontium was analysed, so it is unknown how more violations were present concerning too high activities of ^{90}Sr. In 2007 and 2012, we analysed baby food for both radionuclides. Whereas radiocaesium levels were quite low (<0.05 up to 0.5 Bq/kg), the radiostrontium contamination reached almost the same values (0.3 Bq/kg). Radium belongs to the same element group of the earth alkaline metals as calcium and strontium. Therefore, it is not surprising to find contaminations with radium (^{226}Ra and ^{228}Ra) in infant formulas (0.1–0.8 Bq/kg) [70, 71].

5.9. Spices and salt

In 1986/1987, spices were of no special concern. They figure as a food of minor relevance, because the consumption rate of spices is relatively low in Switzerland. A second reason is the fact that spices are imported from the Middle and Far East, where they were not at all affected by the fallout from Chernobyl. Over the last 10 years, the radiocaesium content in spices was

stable: 1 to 5 Bq/kg for 200 samples analysed. Higher activity concentrations were found for natural radionuclides from the uranium and thorium decay series. For example, white and black pepper and cinnamon contain considerable amounts of thorium and radium (up to 40 Bq/kg). This is near the Swiss limit value of 50 Bq/kg [72].

Salts belong to the spices. Either they are produced from evaporation of seawater or are yielded from mines or salt fields. A survey of 23 products gave the following results: artificial radionuclides, such as radiocaesium, are not present. A major contaminant of salts is potassium chloride. Therefore, it is not surprising that ^{40}K activity concentrations are relatively high in salts: 330±30 Bq/kg. One salt from Persia contained 6 kBq/kg ^{40}K. Some radionuclides from the decay series, such as radium, are present in small amounts: 1.2±1.1 Bq/kg $^{226+228}$Ra [73].

5.10. Honey

In 1986, 40 Swiss honey samples were investigated with γ-spectrometry. They contained ^{131}I with a mean of 40.3 Bq/kg (6 samples) and radiocaesium with 54±47 Bq/kg (35 samples). The highest value was 192 Bq/kg of ^{137}Cs. Six samples contained radioiodine over the tolerance limit of 10 Bq/kg, and six samples were over the tolerance level for radiocaesium of 10 Bq/kg. No violation of the limit values was observed [74].

Honey is considered as food of minor importance. Therefore, its survey was stopped in 2004. From 2005 on, we analysed more than 150 honey samples. They can be divided into two groups: honey from flowers and forest honeys (included chestnut honeys). Honey from flowers show only small amounts of contamination: 0.2 to 5 Bq/kg radiocaesium. In forest honey, one can find up to 25 Bq/kg ^{137}Cs. Sporadically, we found violation of the tolerance values for radiocaesium and radiostrontium. One honey from Austria contained 1.6 Bq/kg ^{90}Sr and 176 Bq/kg ^{137}Cs. These products, and especially products from East European countries, contain elevated contaminations even 30 years after the accident at Chernobyl [75, 76].

5.11. Tea

After the Chernobyl accident, we analysed tea with γ-ray spectrometry. From 21 samples, 12 teas exceeded the tolerance value of 500 Bq/kg. The mean activity found was 12.4±10 kBq/kg radiocaesium. One tea showed 429 Bq/kg, a clearly elevated contamination. In the following year, no sample exceeded 500 Bq/kg radiocaesium. After some years, the contamination levels were reduced to below 5 Bq Cs/kg with one exception. Contamination in black tea from Turkey was only slowly declining. Even in 2015, almost 30 years after the Chernobyl accident, the radiocaesium level reached 50 Bq/kg, and we found radiostrontium in amounts of 38 Bq/kg maximum. In 2011, our focus was set on imported tea from Japan. Until the end of 2015, we analysed more than 150 tea samples coming from different prefectures in Japan. The γ-analyses proved the contamination of green tea from the Fukushima-Daiji NPP's accident. At least, part of the measured radiocaesium originates from the fallout of the NPP's accident. This is proven by the presence of the short-lived radionuclide ^{134}Cs (2.1 years). In 30 of 157 investigated tea samples, ^{134}Cs was present in amounts of 13.4±24.8 Bq/kg. Besides tea, other food categories that are imported from Japan were analysed. Over the last 5 years, we analysed more than 350

food samples (**Table 2**). As can be noticed, the radiostrontium level of tea is approximately 5±7 Bq/kg and could be found in every tea sample analysed. This contamination mainly comes from the bomb's fallout. Teas from other countries of the Far East also contain radiostrontium [77, 78].

Food category	^{134}Cs	^{137}Cs	$^{134+137}$Cs	^{90}Sr
Tea (157)	13.4±24.8 (30)	9.9±28.6 (86)	8.0±33 (90)	5.2±7.4 (94)
	<0.5–87	<0.5–171	<0.5–258	<0.1–57
Soups, miso, (44)	<0.5	0.34±0.24 (9)	0.34±0.24 (9)	N/A
		<0.5–0.7	<0.5–0.7	
Algae (60)	<0.2	0.6±0.7 (9)	0.6±0.7 (9)	0.56±0.31 (12)
		<0.2–2.4	<0.2–2.4	<0.01–1.0
Rice and rice products (24)	<0.2	0.31±0.04 (2)	0.31±0.04 (2)	N/A
		<0.2–0.3	<0.2–0.3	
Soja and soja products (7)	<0.1	0.5±0.2 (1)	0.5±0.2 (1)	N/A
Cereals and cereal products (26)	<0.2	6.3±8.1 (2)	6.3±8.1 (2)	N/A
		<0.2–12	<0.2–12	
Vegetables, fruits (15)	3.9 (1)	5.0±6.9 (2)	6.9±9.6 (2)	0.5±0.4 (2)
	<0.5–3.9	<0.5–9.8	<0.5–14	
Fish and fish products (4)	<0.2	0.3±0.2 (1)	0.3±0.2 (1)	N/A
		<0.2–0.3	<0.2–0.3	
Divers (28)	<0.2	0.52±0.54 (2)	0.52±0.54 (2)	N/A
		<0.2–0.14	<0.2–0.14	

All values in Bq/kg. First line: mean ± standard deviation of the activity concentrations; the number of samples with values over the detection limit is bracketed. The total of analysed samples per food category is set in brackets after the food category name. Second line: activity concentration range of all samples. N/A, not analysed.

Table 2. Overview of investigated food imports from Japan from 2011 to 2015.

5.12. Mineral and tap waters

5.12.1. Artificial radionuclides

1981, when our laboratory started with the radioactivity survey of food, water, besides milk, was the first food category to be monitored. Before 1986, no bomb fallout was detectable in the drinking water of Basel (<0.01 Bq/L radiocaesium). Just after the accident at Chernobyl, radioiodine and radiocaesium were detectable in small amounts of 59±77 and 9±9 Bq/L in some drinking water reservoirs of the state of Jura. The production of drinking water of the city of Basel was never affected.

5.12.2. Natural radionuclides

After 1995, the focus was set on the natural radionuclides from the uranium and thorium decay series. First, uranium and radium were analysed in tap and mineral waters of Switzerland and of abroad. Uranium and radium were found in activity concentrations from <10 to 250 mBq/L and from <10 to 200 mBq/L, respectively. In 2005, the Federal Office of Health analysed more than 5,500 water samples for their uranium content [79]. The World Health Organisation (WHO) considers uranium as relevant in drinking water because of its toxicity as a heavy metal. The WHO recommends a limit value of 30 μg/L (376 mBq/L) for drinking water [80]. In Germany, the tap water from more than 500 drinking water plants were analysed for their natural radionuclides. According to this study, the drinking water of 10% of all plants was over the limit dose of 0.1 mSv/year [81]. One possible input of uranium is supposed to come from the use of phosphate fertilisers in agriculture. These fertilisers may contain uranium up to 50 mg U/kg P_2O_5, which was shown by a market survey in Basel [82]. Our investigation gave cause for a national investigation of the Federal Office of Agriculture. The uranium is relatively soluble and washed from the fertiliser into the soil. From there, it is transferred into groundwater. Surbeck [83] estimated that the use of fertilisers results in the increase of the uranium concentration in groundwater from <0.1 to 3 μg/kg. In 2014, we analysed the tap water of all villages of the states of Basel-Campaign and Basel-City. The spectrum of the relevant radionuclides was expanded with ^{222}Rn and ^{210}Po. For uranium and radium, we found 12±16 mBq/L (n=120) and 17±27 mBq/L (n=54). Radon was present in all samples in the Becquerel range (5±6 Bq/L). Also, ^{210}Po was present in 57 samples in the low mBq range of 26±30 mBq/L [84]. In the alpine regions of southern Switzerland, the activity concentrations were somewhat higher due to the geological underground [85].

5.13. Healing earths

Minerals and sediments consisting mainly of silicon dioxide (quartz) are known as siliceous earths. These fine, floury mineral mixtures are deposits of the silica shells of diatoms, the main constituent of marine phytoplankton. The dead cells sink to the ocean floor and form sediments. These layers of sediment are extracted in numerous mines all over the world. Siliceous earths have a wide variety of uses in, for example, the pharmaceutical and food industries (e.g., as a food supplement). Due to their special structural properties, foreign atoms and ions are incorporated during sediment formation, such as radionuclides of the natural decay series of uranium and thorium [86].

In 2008, we collected some siliceous earth products on the Swiss market and analysed them with γ-spectrometry. In two products, the limit value for natural radionuclides was exceeded (50 Bq/kg). Furthermore, the annual dose by regular consumption of one product reached half of the permitted yearly dose of 1 mSv. The Federal Office for Health Products Control, Swiss Medic, complained about these products. The company involved then withdrew the product from the market [86]. An inspection of the products in 2010 showed that two products from one producer in Germany slightly exceeded the limit value. Higher levels of ^{226}Ra and ^{228}Ra were the reason for this. The estimation of the received annual dose from the consumption of the product according to recommended amount per day as advised on the information leaflet

enclosed would lead to 0.1 mSv/year. Healing earths, and also silica-based chemicals of chemical laboratories, remain a source for natural radionuclides [87].

5.14. Charcoal and briquettes

In 2009, more than 10,000 tons of wood pellets had to be withdrawn from the Italian market. The product was from Lithuania and was contaminated with radiocaesium (300 Bq/kg). The fact that such products and barbecue coals are imported from countries such as Ukraine or Poland motivated us to conduct this investigation. Charcoal is produced either by charcoal burning of wood (possibly contaminated by the fallout of Chernobyl) or from coal mining. The survey of barbecue coals over the last 6 years showed that there is some contamination with radiocaesium (13±20 Bq/kg). We could not verify the high values from Italy; instead, one also has to consider some radiation of the coals derived from natural radionuclides, such as radium, uranium, thorium, and lead (^{210}Pb). The latter is present in activities of around 65±68 Bq/kg, which is over the limit of 10 Bq/kg of the ordinance of radioprotection. The thorium activity concentrations also reach the permitted limit of 6 Bq/kg [88, 89]. Barbeque experiments with steaks grilled over charcoal showed only a slightly contamination of the meat. The main activity remains in the barbeque ashes [90].

5.15. Estimation of internal doses by the consumption of contaminated food in 1986/1987

Based on the results presented from our own investigations in 1986/1987, we estimated a received dose by ingestion of 4.6 mSv. A main contribution came from contaminated vegetables. It is not clear if the population followed an appeal and the recommendations by the government to avoid the consumption of such contaminated vegetables. If so, the dose would have been reduced to approximately 2.4 mSv. Our estimations seem to be too high. The basis of our calculations was dominantly on the first months after the Chernobyl accident. Thus, our mean values are not representative of the whole years 1986 and 1987. **Table 3** gives an overview of these investigations.

	Origin	Activity levels 1986/1987			Activity levels 1990/2015	
		$^{131+132}$I	$^{134+137}$Cs	^{90}Sr	$^{134+137}$Cs	^{90}Sr
Milk, CH	Cow	<1	<-1025	35	<0.05–0.2	0.04–0.2
	Sheep	<1–30	<1–624	<1–22	N/A	N/A
	Mother	<1–30	<1	N/A	N/A	N/A
Milk powder	CH	<1	<1–19,000	<1–22	N/A	N/A
Baby food	CH	<1	<1–5400	N/A	<0.05–0.5	0.03–0.3
Milk products	CH	0.4–2	1–2	0.05–1.1		
Chocolate	CH, D	<1	6–80		N/A	N/A
Nuts	TR	<1	100–17,100		0.1–30	N/A

	Origin	Activity levels 1986/1987			Activity levels 1990/2015	
		$^{131+132}$I	$^{134+137}$Cs	^{90}Sr	$^{134+137}$Cs	^{90}Sr
	CH	<1	0.6–120		N/A	
Mushrooms, wild	CH and imports	<1	107–8100	N/A	<1–1300	N/A
Fruit, dried	CH	<1	<1–1600		<1–1	
Vegetables	Leafy	<1–5500	<1–1100		<0.1–1	0.1–0.4
	Root				<0.05–0.5	
Herbs	CH	<1–4	<5–123,000			
Chest nuts	CH, IT	<1	6–270		<0.17	N/A
Wild-grown berries	CH	N/A	<1–110	N/A	0.1–170	0.2–6
	East Europe		N/A			
Tea	Imp.	<2	<2–30,000	430	<0.5–100	2–40
Meat	CH, cow	<1–30	<1–1200			
	CH, sheep, lamb	<1–75	13–2200			
Game	CH, roe deer	<1	150		<1–400	
	CH, deer	<1	20		<1–600	
	CH, wild boar	<1	600		<1–>1250	
	Reindeer imp.	<1	3600		40–600	
Fish	Fish, Basel (CH)	<	22–710	N/A	0.32±0.16	0.17±0.10
	Fish, Ticino (CH)	<1	40–4400	N/A		
	Sea fish, imp.	<1	N/A	N/A		
Flour	CH	<1	20±37		<1–9	
Bread	CH	<1	30–1200		0.1–10	
Flakes, muesli	CH	<1	N/A		<0.2–5	
Honey	CH and abroad	<2–110	<2–192		<0.2–24	<0.05–2
Spices	Abroad	N/A	N/A	N/A	<0.5–5	N/A
Drinking water	CH, tap water, mineral water	<1–200	0.4–22	N/A	<0.05	N/A
	CH and abroad		N/A		<0.05	

Table 3. Summary of the artificial radioactivity levels found in different food of 1986/1987 compared to the data of 1990/2015. All data were generated at the State-Laboratory Basel-City.

No dose estimation was possible for ^{90}Sr. There were not sufficient ^{90}Sr data available for most of the food categories. Most Swiss agencies believe that ^{90}Sr activity was low in the fallout from Chernobyl.

The Federal Office of Public Health estimated the dose commitment for the Swiss population to be 0.2 mSv/year in 1986. The estimation was based on the results of the food categories milk, vegetables, and meat only. Other food categories were not taken into account (e.g., cereals or ^{90}Sr data of milk). According to the estimated dose from radioiodine in milk, they estimated that, for children, 75 new cases of thyroid cancer with seven or eight fatal cases would occur. For adults, there was no risk seen [91]. The genetically based radiation risk, which causes genetically anomalies, was estimated at 2 to 22 new cases between 1986 and 2086 [92]. The Swiss Federal Nuclear Safety Inspectorate (former HSK) published a more profound study. They came to the following conclusions: for children of age 1 year, children up to 10 years, and adults, a whole body dose of 0.6 to 1.6, 1.0, and 1.1 mSv was calculated, respectively [93]. The Association for Radioprotection of Germany and Switzerland calculated somewhat lower doses (0.4–1.0 mSv for children and 0.4–0.8 mSv for adults) [94]. The Swiss Federal Office of Public Health compared the calculated doses to the whole-body countings of Swiss people. They calculated 10-fold lower doses according to the whole-body countings [95].

In 2014, the Federal Office of Public Health estimated the mean dose through the consumption of food to be 0.35 mSv. The main contribution came from ^{40}K (0.2 mSv/year) and from natural radionuclides of the uranium and thorium series. The remaining contamination from the bomb fallout was less than 0.1 mSv/year [96].

The listed dose estimations fluctuate because of the use of different radioactivity concentrations in food and different assumptions of the consumption rates. All these estimations are based on the dose coefficients for inhalation and ingestion of the International Commission for Radiation Protection (ICRP). However, there are other commissions such as the European Committee on Radiation Risk (ECRR), which conducts more assessments on the effects of low doses. Their dose coefficients are different from ICRP data for some important radionuclides. The ingestion dose factors of the ECRR for ^{137}Cs and ^{90}Sr are 5- and 320-fold higher than the ICRP factors. The ECRR attaches more importance to aspects such as DNA damage by radionuclides [97]. The application of these dose factors would result in higher doses by ingestion of ^{137}Cs and ^{90}Sr. Furthermore, by respecting these dose factors, the limit values for radionuclides in food should be considerably lower for ^{90}Sr and ^{137}Cs.

Different countries have different limit values, even using the same dose coefficients from ICRP. This led to confusion in 1986 and was also a problem in 2011. Policymakers and the general public do not understand such differences. Why are there different threshold values for the same radionuclide in the same food? In Switzerland, the Government has the intention to let fall most limit values for radionuclides in food in "times of non-crisis". In times of crisis, the Government should enact ad hoc specific limit values, as was done in 1986 and 2011.

It is not clear how the state laboratories can sustain their activities and conserve their know-how in the field of radioactivity monitoring of food without limit values. However, we must always be prepared for emergency cases.

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