Procedure for determining the specific activities of radionuclides in fish by gamma spectrometry

G-γ-SPEKT-FISCH-01

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ISSN 1865-8725 Version January 2016 / verified March 2020

Procedures manual for monitoring of radioactive substances in the environment and of external radiation
(Messanleitungen für die „Überwachung radioaktiver Stoffe in der Umwelt und externer Strahlung")

Procedure for determining the specific activities of radionuclides in fish by gamma spectrometry

1 Scope

The procedure described in the following is suitable for gamma spectrometrical analysis of samples of fish that have to be monitored according to the German Radiation Protection Act (StrlSchG) in the IMIS-routine programme and the Guideline for the Monitoring of Emissions and Immissions of Nuclear Installations during Regular Operation.

The procedure is suitable for low-level measuring in the framework of radio-ecological studies due to its low limit of determination after dry-ashing of the sample.

2 Sampling

The sampling of fish – especially marine fish – relies to a wide extent on the co-operation of professionals in the respective industries (professional fishermen) and the product range available at fish markets.

It is in the responsibility of the person collecting the sample to ensure that a sample taken can be traced back to the sampled water body. The sampling position needs to be recorded as exact as possible. The name of the samples fish species is also an essential part of the sample label. In case of streams, the river kilometre has to be additionally reported, while for saltwater fish the FAO fishing areas or coordinates of the catch must be reported by the fishermen. According to an EU regulation, marketed fish must be labelled with its registered trade name and the fishing area (1).

The required sample size depends on the type of study to be carried out. For studies according to the Radiation Protection Act and the Guideline for the Monitoring of Emissions and Immissions of Nuclear Installations, at least 1,5 kg of fish flesh is required. This equals a minimum of 3 kg non-filleted original sample. If the samples are to be used for additional analyses like strontium or isotopes of plutonium and americium, the sample size should be enlarged to 5 kg of fish flesh. The fish flesh will usually be preferred for analysis, but some species, e. g. sprat, must be examined as a whole fish because of their small size. The determining factor for sample preparation is the typical consumption of this species. In case of individual fishes, the person collecting the sample must ensure that only marketable fish is sampled.

The freshly collected fish are killed quickly at the point of sampling, sorted by species, packed in plastic bags, and transported to the laboratory in ice-filled containers. The fish samples are stored deep-frozen at ca. -20 °C until they can be processed further in the laboratory.

3 Analysis

3.1 Principle of the procedure

The whole fish, or fish flesh obtained after filleting, is dried at 100 °C to 110 °C and then ashed at a maximum temperature of 420 °C. The specific activity of radionuclides in the homogenised ash are analysed using a germanium semiconductor detector.

The specific activities of iodine isotopes cannot be determined quantitatively because of the low boiling point of iodine (184 °C). The quantitative determination of iodine isotopes in fish requires the use of the procedure G-γ-SPEKT-FISCH-02.

3.2 Sample preparation

All items used for preparation of the ashes, e. g. stainless steel bowls, ceramic bowls, measuring beakers, etc. must be cleaned using cleaning solution prior each use.

During filleting of fresh or defrosted fish, attention needs to be paid to deboning them as completely as possible. The obtained fish flesh is roughly chopped up. Chopped fish flesh or whole fish samples are placed in suitably large stainless steel or ceramic bowls that are laid out with ash-free transparent paper. Frozen fillets are defrosted directly in prepared stainless steel bowls in order to include tissue fluids liberated during the defrosting process in the subsequent drying and ashing steps. In case of frozen whole fish, the cell liquids from defrosting of roundfish, e. g. herring, sprat or cod, and flatfish, e. g. dab, flounder or plaice, are treated differently. As the latter release large amounts of mucus during frosting and defrosting which may disturb a later radiochemical analysis, the liquid is discarded (see Table 1). The total fresh mass used is to be recorded.

Tab. 1: Consideration of tissue fluids arising from the defrosting process of frozen fish samples in the analysis

	fillet	whole fish	
		roundfish	flatfish
Addition of tissue fluids arising from the defrosting process to the sample	+	+	–

Afterwards, fish samples are dried until constant mass is reached, i. e. about one day or two days at a temperature of about 110 °C. Thereafter, the dry mass (TM) is determined. The ratio between fresh mass and dry mass in fish meat is about 5.

The time to reach the maximum ashing temperature depends on the lipid content of the sample; for normal fish it takes 45 hours. If fish with high lipid content, e. g. eel, mackerel, herring or sprat are to be ashed, this time needs to be extended in order to avoid the ignition of the sample. In order to achieve a complete ashing of the sample, the maximum temperature is held for another 96 to 154 hours.

The following temperature program is commonly used for fish samples with average lipid content:

Step 1: Linear increase of the temperature to 230 °C within 3 hours;

Step 2: Linear increase to the maximum temperature 420 °C within 42 hours;

Step 3: Hold the maximum temperature for at least 96 hours, optimal 154 hours.

The final temperature of 420 °C is selected to avoid the volatilisation of caesium. Experiments to this effect, using the temperature program listed above, revealed ashing-induced losses of about 1 %.

Note
The danger of ignition during the ashing process rises, when the sample consists of material high in lipid content, e. g. cod liver. Therefore, the bowls must contain only thin layer of these samples. The maximum temperature must be selected lower than 380 °C in these instances.

Once ashing has been completed and the sample has cooled down, the mass of the ash is recorded. For fish meat, a ratio of fresh mass to ash mass of about 80 (range 65 to 95) has usually to be expected; in whole fish, it may be lower by a factor of 2. Carbon residues still contained in the ash will not affect the results of the gamma spectrometric analysis. The ash is homogenised using a mortar, filled into a suitably large measuring beaker, and then carefully manually compressed with a stamp. The obtained filling height is recorded. The measurement geometry is calculated from the mass and filling height of the sample and the diameter of the measuring beaker. The compressed density is commonly around 0,5 $g \cdot cm^{-3}$.

3.3 Radiochemical separation

A radiochemical separation is not required.

4 Measuring the activity

4.1 General

The basics of gamma spectrometry are discussed in the basic chapter γ-SPEKT/GRUNDL of this procedures manual and in the literature (2, 3, 4).

4.2 Calibration

4.2.1 Determination of the detection efficiency

For determining the energy-dependency of the detection efficiency, reference is made to the basic chapter γ-SPEKT/GRUNDL of this procedures manual.

A calibration using a calibration source is described here. A suitable calibration source is e. g. an aqueous solution of a traceable standard or a resin containing traceable amounts of radioactive compounds. Mathematical calibration of the detection efficiency may also be used if the required premises are fulfilled.

The relative standard uncertainties of the activities used should be less than 1 %. The relative standard uncertainty of Pb-210 may be larger due to conditions of manufacturing. The measurement duration of the calibration has to be selected to ensure standard uncertainties of the net count rates to be below 1%.

An example of a calibration using an aqueous solution with a density of 1,03 g·cm^{-3} (hydrochloric acid, 2 mol·l^{-1}) is described in the following.

Two alternatives are available which differ in the selection of radionuclides and depend on the consideration of coincidence summation.

4.2.1.1 Calibration using single-line radionuclides

A calibration solution containing sufficient activity of a gamma emitting radionuclide with only one emission line in the energy range between 100 keV and 1200 keV, e. g. Sr-85, Cs-137, Mn-54 or Zn-65, respectively, is used. To extend this calibration curve to energies smaller than 100 keV, a second solution containing e. g. Pb-210, Am-241, Cd-109 und Co-57 is required. A curve is fitted to the values obtained by equation (1):

$$\varepsilon_{\mathrm{w}}(E_{\mathrm{r},i}) = \frac{R_{\mathrm{n,r},i}}{A_{\mathrm{r}} \cdot p_{\gamma}(E_{\mathrm{r},i})} \cdot f_1(E_{\mathrm{r},i}) \cdot f_2(E_{\mathrm{r},i}) \cdot f_{4,\mathrm{r}} \qquad (1)$$

Herein are:

A_{r} activity of the radionuclide r used for the calibration at the reference date, in Bq;

$E_{\mathrm{r},i}$ energy of the gamma line *i* of radionuclide r, in keV;

$\varepsilon_{\mathrm{w}}(E_{\mathrm{r},i})$ detection efficiency for water, depending on energy and fill level, in Bq^{-1}·s^{-1};

$R_{\mathrm{n,r},i}$ net count rate of line *i* of radionuclide r, in s^{-1};

$f_2(E_{\mathrm{r},i})$ self-absorption correction factor in water relative to hydrochloric acid (2 mol·l^{-1}):

$$f_2(E_{\mathrm{r},i}) = \frac{\varepsilon_{\mathrm{MC(H_2O)}}}{\varepsilon_{\mathrm{MC(HCl)}}} \ ;$$

$\varepsilon_{\mathrm{MC}}$ detection efficiency, calculated by Monte Carlo (MC)-simulation, in Bq^{-1}·s^{-1};

$f_{4,\mathrm{r}}$ decay correction factor of radionuclide r

$$f_{4,\mathrm{r}} = \frac{\lambda_{\mathrm{r}} \cdot t_{\mathrm{m}}}{(1-\mathrm{e}^{-\lambda_{\mathrm{r}} \cdot t_{\mathrm{m}}})} \cdot \mathrm{e}^{-\lambda_{\mathrm{r}} \cdot t_{\mathrm{A}}} \ ;$$

t_{A} time period between commencement of measuring and reference date of the standard, in s;

t_{m} duration of measurement, in s;

$p_{\gamma}(E_{\mathrm{r},i})$ probability of gamma intensity of line *i* from radionuclide r.

K-40 is the only available single-line emitting radionuclide in the energy range greater than 1200 keV, but it requires large periods of measurement. Therefore, the multi-line gamma emitting nuclide Y-88 is used instead, but afterwards a correction for coincidence summing effects is needed to avoid a bias in the calibration curve (see section 4.2.1.2).

4.2.1.2 Calibration using multi-line radionuclides

The experimental effort may be reduced when corrections for coincidence summation can considered already during calibration, because then the energy range between 47 keV and 1836 keV may be covered by a single solution containing a mixture of radionuclides. This mixture consists of the radionuclides for the lower energy range mentioned in section 4.2.1.1 and additional multi-line radionuclides like Te-123m, Cr-51, Sn-113, Cs-134, Co-60 and Y-88.

A calibration curve is fitted to values calculated by equation (2):

$$\varepsilon_{\mathrm{W}}(E_{\mathrm{r},i}) = \frac{R_{\mathrm{n,r},i}}{A_{\mathrm{r}} \cdot p_{\gamma}(E_{\mathrm{r},i})} \cdot f_1(E_{\mathrm{r},i}) \cdot f_2(E_{\mathrm{r},i}) \cdot f_{4,\mathrm{r}} \qquad (2)$$

An additional symbol used here is:

$f_1(E_{\mathrm{r},i})$ correction factor for coincidence summation of line *i* emitted by the radionuclide r.

4.2.1.3 Filling height dependent calibration curves

The calibration must be carried out for different filling heights. The filling heights of the measuring preparations are selected according to the range of filling heights expected in samples. Four energy-dependent calibration curves for different filling heights are shown in Figure 1.

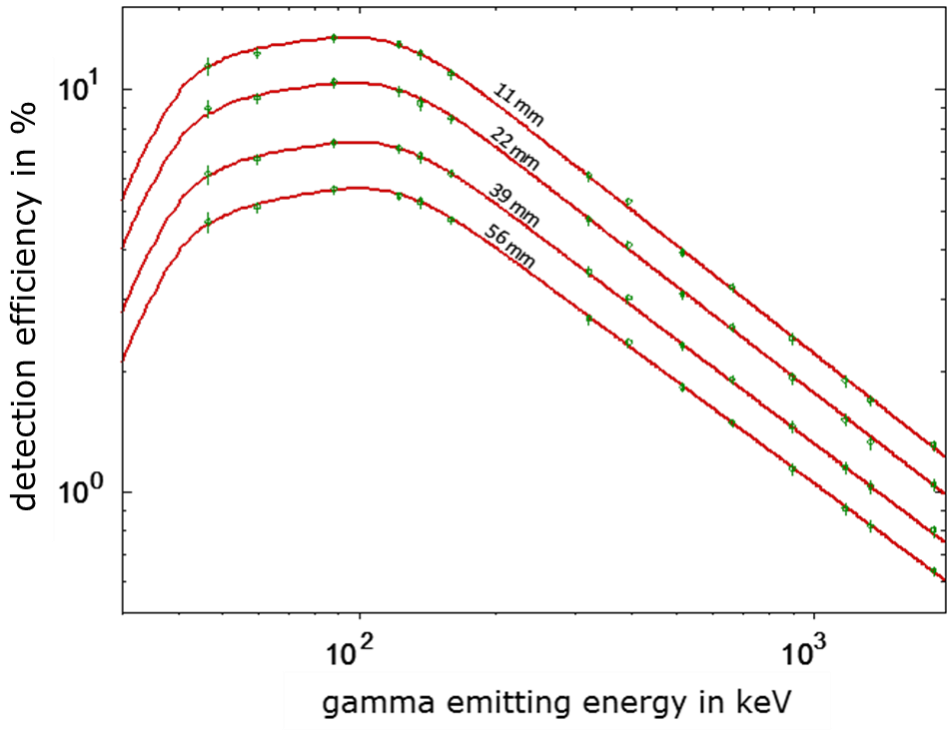


Fig. 2: Determined detection efficiency in % and fitted calibration curves for different filling heights between 11 mm and 56 mm, ordered from top to bottom; the calibration was carried out on an n-type detector with aluminum endcap.

4.2.2 Correction for self-attenuation

The self-attenuation of a sample depends on its density, its elemental composition and the energy-dependent mass attenuation coefficients of the single elements.

A self-attenuation correction factor $f_5(E)$ is used to correct the detection efficiency for water as obtained in section 4.2.1 to those of fish-ash. This correction factor depends on the gamma-ray energy, the density of the preparation, the elemental composition of the ash and the beaker and finally the filling height, respectively. Software-tools are essential for its calculation. Therein, an approximate knowledge of the detector's geometric dimensions is sufficient.

The elemental composition listed in Table 2 may be used to calculate the self-attenuation correction factor of ash from fish flesh as weighted mean of the mass-attenuation factor from single elements (5, 12).

Tab. 1: Typical composition of ash from fish flesh

Element resp. compound	relative mass fraction
K	0,245
PO_4	0,460
Cl	0,109
Na	0,067
ZnO	0,0576
CaO	0,0308
MgO	0,0305

The following equation is used to calculate the energy depending self-attenuation factor $f_5(E)$ in the energy range larger than 45 keV:

$$f_5(E) = a_1 \cdot E^{a_2} + a_3 \cdot E^{a_4} = \frac{\varepsilon_{\mathrm{MC}}(\mathrm{H_2O})}{\varepsilon_{\mathrm{MC}}(\mathrm{Ash})} \qquad (3)$$

with

$f_5(E)$ self-attenuation correction factor for ash relative to water;

E gamma-ray energy, in keV;

a_k parameter obtained through curve fitting (k = 1, … ,4).

Figure 2 shows four curves fitted to calculated values of the energy dependent self-attenuation factor obtained for ash from fish flesh and hydrochloric acid (2 mol·l^{-1}). It is obvious that $f_5(E)$ strongly increases in the energy range below 45 keV. For energies significantly below 40 keV it might be necessary to include a third power function term in equation (3).

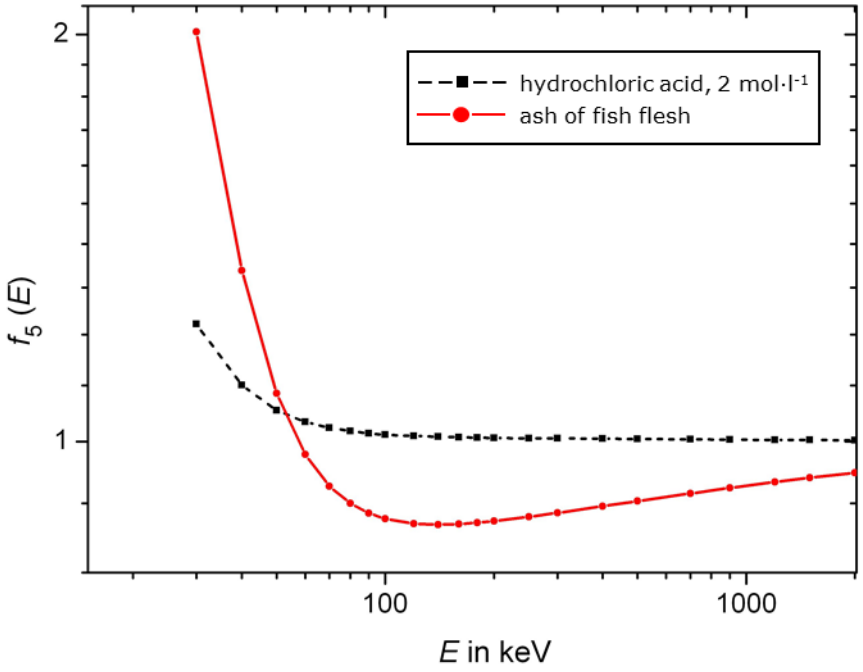


Fig. 3: Calculated self-attenuation correction factors as function of the gamma-ray energy after calibration using hydrochloric acid (2 mol·l^{-1}) and in ash from fish flesh; the filling-height are 40 mm, the density of the ash is 0,47 g·cm^{-3} and that of hydrochloric acid 1,03 g·cm^{-3}.

Values of the relative standard uncertainty of $f_5(E)$ for photon energies above 100 keV have been determined to be around 1,5 %; the influence of measurement geometry and elemental composition of the ash increases for lower photon energies, which leads to a strong increase in relative standard uncertainty.

4.2.3 Detection efficiency in ash

The analysis for specific activities of gamma-ray emitting radionuclides in the sample requires to determine the detection efficiency in fish ash $\varepsilon_A(E)$. It is calculated as the quotient of the detection efficiency in water $\varepsilon_W(E)$ and the self-attenuation correction factor $f_5(E)$.

$$\varepsilon_A(E) = \frac{\varepsilon_W(E)}{f_5(E)} \tag{4}$$

In case that the standard uncertainty of the activities used for calibration is less than 1 %, values of the relative combined standard uncertainty of the detection efficiency in ash $\varepsilon_A(E)$ in the range of 2 % to 4 % for photon energies larger than 100 keV are achievable; they may be much larger for photon energies below 100 keV.

4.3 Background

For determining the background, reference is made to the basic chapter γ-SPEKT/NULLEF of this procedures manual.

Background spectra need to be recorded at regular intervals to check the gamma spectrometer for contaminations. Measuring periods for this purpose should be at least 1,5 days to 3 days. It is recommended to obtain a mean value and standard deviation of the net count rate of each individual background gamma-ray energy from two or three background spectra. This will typically encompass lines relating to Ra-226 decay products which may exhibit some count rate fluctuation depending on the ventilation of the room. They may be reduced by e. g. the supply of gaseous nitrogen escaping from the dewar used for cooling of the detector to the measurement chamber.

For samples of fish, reliably determining the K-40 background count rate of the background effect is important in order to be able to use the K-40 activity determined in fish for verifying the correctness of the subsequent sample spectrum analysis.

4.4 Measurement

The determination of the specific activity of gamma emitting radionuclides in fish ash does not necessarily require a low-level gamma spectrometer, because a significant input to the pulse height spectrum originates from the Compton spectrum of K-40. This input may increase the background of the sample compared to the background checks by a factor of three. Generally, the specific activity concentration of K-40 in fish flesh is around 110 $Bq \cdot kg^{-1}$ FM and in whole fish around 80 $Bq \cdot kg^{-1}$ FM.

Cylindrical polyvinylchloride (PVC) beakers with plane bottom are used for the analysis.

Note
Compared to other plastic materials, PVC has got the advantage that the detection efficiency for X-ray energies is reduced due to the higher absorption of smaller energies. This may influence the summing of coincidences (see section 4.2.1.2 and basic chapter γ-SPEKT/GRUNDL of this procedures manual).

The measurement is normally carried out over night. The measuring period may be enlarged, depending on the scope of the analysis or the required detection limit (see section 5.2).

A correction of coincidence summing effects according to section 4.2.1.2 in γ-SPEKT/GRUNDL has to be applied, if nuclides with the emission of multiple gamma-ray energies are to be determined; see also (6, 7, 8).

5 Calculation of the results

5.1 Equations

When determining specific activities of radionuclides in ash from fish flesh, interferences due to gamma lines of different radionuclides are scarcely occurring. Therefore, specific activities of radionuclides may be calculated either from a single gamma line or, in the case of multiple-line radionuclides, by using a weighted mean.

If interferences occur, reference is made to the basic chapter γ-SPEKT/INTERF of this procedures manual.

5.1.1 Equations for calculating the specific activity from a single gamma line

If a net count rate $R_{\mathrm{n,r}}$ at the gamma-ray energy of the radionuclide r has been detected, the resulting specific activity a_{r}, relative to fresh mass (FM) and the date and time of sampling, is calculated according to equation (5):

$$a_{\mathrm{r}} = \varphi \cdot R_{\mathrm{n,r}} = \frac{f_1 \cdot f_3}{\varepsilon_{\mathrm{A}} \cdot p_{\gamma} \cdot m_{\mathrm{A}} \cdot q_{\mathrm{F}}} \cdot \mathrm{e}^{\lambda_{\mathrm{r}} \cdot t_{\mathrm{A}}} \cdot R_{\mathrm{n,r}} \qquad (5)$$

with:

$$f_3 = \frac{\lambda_{\mathrm{r}} \cdot t_{\mathrm{m}}}{1 - \mathrm{e}^{-\lambda_{\mathrm{r}} \cdot t_{\mathrm{m}}}}$$

Herein are:

$R_{\mathrm{n,r}}$ net count rate of the line of the radionuclide r, in s^{-1};

φ procedural calibration factor, in $Bq \cdot s \cdot kg^{-1}$;

f_1 correction factor for coincidence summing;

f_3 correction factor for the decay of the activity of the radionuclide r during the measurement;

ε_{A} detection efficiency in ash, in $Bq^{-1} \cdot s^{-1}$ (see section 4.2.3);

p_{γ} emission intensity of the gamma line of the radionuclide r;

m_{A} mass of the ash used for the measurement, in kg;

q_{F} ratio of fresh mass to ash mass;

t_{A} time period between sampling and beginning of the measurement, in s;

t_{m} duration of measurement, in s;

λ_{r} decay constant of the radionuclide r, in s^{-1}.

The net count rate of the gamma line of the radionuclide r is calculated according to equation (6):

$$R_{\mathrm{n,r}} = R_{\mathrm{b,r}} - R_{\mathrm{T,r}} - R_{0,\mathrm{r}} \qquad (6)$$

Herein are:

$R_{\mathrm{b,r}}$ gross count rate of the line of the radionuclide r, in s^{-1};

$R_{\mathrm{T,r}}$ background count rate underneath the line of the radionuclide r, e. g. as count rate of a trapezoidal background, in s^{-1};

$R_{0,\mathrm{r}}$ net count rate at the line of the radionuclide r in a background spectrum, in s^{-1}.

The generalized expression to calculate the uncertainty of the net count rate is given in equation (7), with the coefficients μ_k are to be determined according to equation (8):

$$u^2(R_{\mathrm{n,r}}) = \mu_0 \cdot R_{\mathrm{n,r}}^2 + \mu_1 \cdot R_{\mathrm{n,r}} + \mu_2 \tag{7}$$

With the coefficients:

$$\mu_0 = 0 \qquad \mu_1 = \frac{1}{t_{\mathrm{m}}} \qquad \mu_2 = \frac{R_{\mathrm{T,r}} + R_{0,\mathrm{r}}}{t_{\mathrm{m}}} + u^2(R_{\mathrm{T,r}}) + u^2(R_{0,\mathrm{r}}) \tag{8}$$

Using the trapezoidal method (assuming a linear background continuum) the coefficient μ_2 is given by equation (9):

$$\mu_2 = \frac{R_{\mathrm{T,r}}}{t_{\mathrm{m}}} \cdot \left(1 + \frac{b}{2 \cdot L}\right) + R_{0,\mathrm{r}} \cdot \left(\frac{1}{t_{\mathrm{m}}} + \frac{1}{t_0}\right) + \frac{R_{\mathrm{T},0,\mathrm{r}}}{t_0} \cdot \left(1 + \frac{b_0}{2 \cdot L_0}\right) \tag{9}$$

Herein are:

t_0 duration of the background measurement, in s;

b line widths of sample spectra at the respective peak baselines, in channels;

b_0 line widths of background spectra at the respective peak baselines, in channels;

L numbers of channels for the sample spectrum, respectively, over which the background continuum to the left and to the right of the peaks are estimated;

L_0 numbers of channels for the background spectrum, respectively, over which the background continuum to the left and to the right of the peaks are estimated;

$R_{\mathrm{T},0,\mathrm{r}}$ background continuum count rate at the line of the radionuclide r within the background spectrum, e. g. as a trapezoidal background count rate, in s^{-1}.

Equation (9) is also a sufficient approximation for a background estimated by an empirical background step function.

The standard uncertainty of the net count rate $u(R_{\mathrm{n,r}})$ calculated by equations (7) to (9) is given by:

$$\begin{aligned} u^2(R_{\mathrm{n,r}}) &= \frac{R_{\mathrm{n,r}}}{t_m} + \mu_2 = \\ &= \frac{R_{\mathrm{n,r}}}{t_m} + \frac{R_{\mathrm{T,r}}}{t_{\mathrm{m}}} \cdot \left(1 + \frac{b}{2\,L}\right) + R_{0,\mathrm{r}} \cdot \left(\frac{1}{t_m} + \frac{1}{t_0}\right) + \frac{R_{\mathrm{T},0,\mathrm{r}}}{t_0} \cdot \left(1 + \frac{b_0}{2\,L_0}\right) \end{aligned} \tag{10}$$

When no count rate occurs at corresponding gamma-ray energy of background spectrum equation (10) is reduced by the last two terms.

The relative standard uncertainty of the procedural calibration factor is determined according to equation (11), where the uncertainty of the decay correction may be neglected:

$$u_{\mathrm{rel}}(\varphi) = \sqrt{u_{\mathrm{rel}}^2(f_1) + u_{\mathrm{rel}}^2(f_3) + u_{\mathrm{rel}}^2(\varepsilon_{\mathrm{A}}) + u_{\mathrm{rel}}^2(p_{\gamma}) + u_{\mathrm{rel}}^2(m_{\mathrm{A}}) + u_{\mathrm{rel}}^2(q_{\mathrm{F}})} \qquad (11)$$

The combined standard uncertainty of the specific activity $u(a_{\mathrm{r}})$ using equation (5) is calculated as:

$$u(a_{\mathrm{r}}) = a_{\mathrm{r}} \cdot \sqrt{u_{\mathrm{rel}}^2(\varphi) + u_{\mathrm{rel}}^2(R_{\mathrm{n,r}})} \qquad (12)$$

5.1.2 Equations for calculating the specific activity from multiple gamma lines

The specific activity a_{r} relative to fresh mass (FM) and the time of sampling of a number of j intensities (j greater than 1) at gamma-ray energies of radionuclide r sufficiently detected are calculated according to equation (13):

$$a_{\mathrm{r}} = \varphi_m \cdot A_{\mathrm{r}} = \frac{\mathrm{e}^{\lambda_{\mathrm{r}} \cdot t_{\mathrm{A}}} \cdot f_3}{m_{\mathrm{A}} \cdot q_{\mathrm{F}}} \cdot A_{\mathrm{r}} \qquad (13)$$

Herein, A_{r} represents the activity at the time of measurement, which is calculated as a weighted mean from the intensity of a number of single gamma-ray energies of nuclides,

$$A_{\mathrm{r}} = u^2(A_{\mathrm{r}}) \cdot \sum_j \frac{A_{\mathrm{r},j}}{u^2(A_{\mathrm{r},j})} \qquad (14)$$

wherein the variance of the uncertainty $u^2(A_{\mathrm{r}})$ is given by equation (15):

$$u^2(A_{\mathrm{r}}) = \frac{1}{\sum_j \frac{1}{u^2(A_{\mathrm{r},j})}} \qquad (15)$$

The index j in these equations identifies single gamma-ray energies. The activities $A_{\mathrm{r},j}$ of the individual gamma-ray energies are calculated according to equation (5):

$$A_{\mathrm{r},j} = R_{\mathrm{n,r},j} \cdot \frac{f_{1,\mathrm{r},j}}{\varepsilon_{A,\mathrm{r},j} \cdot p_{\gamma,\mathrm{r},j}} = R_{\mathrm{n,r},j} \cdot \varphi_{\mathrm{r},j} \qquad (16)$$

Herein are:

φ_m line-independent, mass-referred calibration factor, in kg^{-1};

$\varphi_{r,j}$ line-referred calibration factor, in Bq·s;

$R_{n,r,j}$ net count rate of the gamma-ray energy j of the radionuclide r, in s^{-1};

$\varepsilon_{A,r,j}$ detection efficiency of the gamma-ray energy j in ash, in $Bq^{-1}\cdot s^{-1}$ (see section 4.2.3);

$p_{\gamma,r,j}$ emission intensity of the gamma-ray energy j;

$f_{1,r,j}$ coincidence summing correction factor for the gamma-ray energy j.

The variances of $A_{r,j}$ are calculated according to the following equation:

$$u^2(A_{r,j}) = \varphi_{r,j}^2 \cdot u^2(R_{n,r,j}) + R_{n,r,j}^2 \cdot u^2(\varphi_{r,j}) \quad (17)$$

When the net count rates are calculated using the trapezoidal background procedure, their standard uncertainties are calculated according to equation (10). If peak fitting is used instead, that factor $(1 + b/(2\,L))$, representing the trapezoidal background procedure, changes into a factor f_B, which depends on the method used for peak fitting:

$$u^2(R_{n,r,j}) = \frac{R_{n,r,j}}{t_m} + \mu_{2,r,j} \quad (18)$$

$$\mu_{2,r,j} = \frac{R_{T,r,j}}{t_m} \cdot f_B + R_{0,r} \cdot \left(\frac{1}{t_m} + \frac{1}{t_0}\right) + \frac{R_{T,0,r}}{t_0} \cdot \left(1 + \frac{b_0}{2\,L_0}\right) \quad (19)$$

The factor f_B generally depends on the averaged background continuum per channel underneath the gamma-ray energy and on the ratio

$$\frac{R_{n,r,j} \cdot t_m}{\sqrt{R_{T,r,j} \cdot t_m}}$$

For net count rates close to the detection limit or the decision threshold, f_B may be approximated by a fixed value; see the calculation example in section 5.2.2.

Finally, the standard uncertainty of the specific activity is calculated as follows:

$$u(a_r) = a_r \cdot \sqrt{u_{rel}^2(\varphi_m) + u_{rel}^2(A_r)} \quad (20)$$

with

$$u(a_r) = a_r \cdot \sqrt{u_{rel}^2(\varphi_m) + u_{rel}^2(A_r)} \quad (21)$$

Note
The calculations presented in this section are laborious if carried out manually; therefore, the software UncertRadio has been used to calculate the parameters of the example (see section 5.2.2). Such software is required for the calculation of the decision threshold and the detection limit by linear unfolding, which uses iteration, and therewith differs from the example shown in section 6.1.

5.2 Worked example

5.2.1 Determining the specific Cs-137 activity

The following values are used for calculating the specific Cs-137 activity in 1,1 kg fish flesh (FM):

$R_{\mathrm{n,Cs-137}}$	= 0,00632 s^{-1};	$u_{\mathrm{rel}}(R_{\mathrm{n,Cs-137}})$	= 3,809·10^{-4} s^{-1};
$R_{\mathrm{T,Cs-137}}$	= 0,00271 s^{-1};	$b/(2 \cdot L)$	= 0,523;
m_{A}	= 0,01317 kg;	$u_{\mathrm{rel}}(m_{\mathrm{A}})$	= 0,004;
q_{F}	= 83,54;	$u_{\mathrm{rel}}(q_{\mathrm{F}})$	= 0,02;
p_{γ}	= 0,8499;	$u_{\mathrm{rel}}(p_{\gamma})$	= 0,00235;
ε_{W}	= 0,03056 $Bq^{-1}{\cdot}s^{-1}$;	$u_{\mathrm{rel}}(\varepsilon_{\mathrm{W}})$	= 0,029;
f_1	= 1,000;	$u_{\mathrm{rel}}(f_1)$	= 0;
f_5	= 0,976;	$u_{\mathrm{rel}}(f_5)$	= 0,008.

The standard uncertainties of the following input quantities are negligible:

t_{A}	= 1,9788·10^{7} s;	$\lambda_{\mathrm{Cs-137}}$	= 7,30937·10^{-10} s^{-1};
t_{m}	= 72000 s.		

This leads to the correction factor f_3 for the decay during the measurement duration:

$$f_3 = \frac{7{,}30937 \cdot 10^{-10}\ \mathrm{s}^{-1} \cdot 72000\ \mathrm{s}}{1 - \mathrm{e}^{-7{,}30937 \cdot 10^{-10} \mathrm{s}^{-1} \cdot 72000\ \mathrm{s}}} = 1{,}00003$$

The detection efficiency in ash ε_{A} is calculated according to equation (4):

$$\varepsilon_{\mathrm{A}} = \frac{0{,}03056}{0{,}976}\ \mathrm{Bq}^{-1} \cdot \mathrm{s}^{-1} = 0{,}03131\ \mathrm{Bq}^{-1} \cdot \mathrm{s}^{-1}$$

The procedural calibration factor defined in equation (5) is given by:

$$\varphi = \frac{1{,}0 \cdot 1{,}00003}{0{,}03131 \cdot 0{,}8499 \cdot 0{,}01317 \cdot 83{,}54} \cdot \mathrm{e}^{7{,}30937 \cdot 10^{-10} \cdot 1{,}9788 \cdot 10^{7}}\ \mathrm{Bq} \cdot \mathrm{s} \cdot \mathrm{kg}^{-1} =$$

$$= 34{,}655\ \mathrm{Bq} \cdot \mathrm{s} \cdot \mathrm{kg}^{-1}$$

The specific activity $a_{\mathrm{Cs-137}}$in the fish flesh relative to the fresh mass (FM) results from equation (5):

$$a_{\mathrm{Cs-137}} = 34{,}655 \cdot 0{,}00632\ \mathrm{Bq \cdot kg^{-1}} = 0{,}219\ \mathrm{Bq \cdot kg^{-1}}\ \mathrm{(FM)}$$

The relative standard uncertainty of the procedural calibration factor is calculated by equation (11):

$$u_{\mathrm{rel}}\,(\varphi) = \sqrt{0^2 + 0{,}008^2 + 0{,}029^2 + 0{,}00235^2 + 0{,}004^2 + 0{,}02^2} = 0{,}03642$$

The standard uncertainty of the net count rate is obtained by equation (10) with neglecting the last two terms:

$$u\big(R_{\mathrm{n,Cs-137}}\big) = \sqrt{\frac{0{,}00632\ \mathrm{s^{-1}}}{72000\ \mathrm{s}} + \frac{0{,}00271\ \mathrm{s^{-1}}}{72000\ \mathrm{s}} \cdot (1 + 0{,}523)} = 3{,}809 \cdot 10^{-4}\ \mathrm{s^{-1}}$$

The relative standard uncertainty of the specific activity $u(a_{\mathrm{Cs-137}})$ is calculated according to equation (12):

$$u(a_{\mathrm{Cs-137}}) = 0{,}219\ \sqrt{0{,}03642^2 + \left(\frac{3{,}809 \cdot 10^{-4}}{0{,}00632}\right)^2}\ \mathrm{Bq \cdot kg^{-1}} = 0{,}015\ \mathrm{Bq \cdot kg^{-1}}\ \mathrm{(FM)}$$

Finally, the specific Cs-137 activity is obtained as:

$$a_{\mathrm{Cs-137}} = (0{,}219 \pm 0{,}015)\ \mathrm{Bq \cdot kg^{-1}}\ \mathrm{(FM)}$$

5.2.2 Determining the specific Cs-134 activity

For calculating the specific Cs-134 activity, the two main gamma energies at 604,7 keV and 795,9 keV were evaluated in a sample 3,7 kg (FM) of flesh from North Atlantic cod off Greenland, whose ash was measured over a period of about 15 days on a Germanium detector with relative efficiency of 35 %. The numerical values used for the calculations are the following:

m_{A}	= 0,05121 kg;	$u_{\mathrm{rel}}(m_{\mathrm{A}})$	= 0,004;
q_{F}	= 72,4;	$u_{\mathrm{rel}}(q_{\mathrm{F}})$	= 0,02.

The values for calculating the activities of the two Cs-134 gamma lines are:

Gamma line 1 at 604,7 keV:

$R_{\mathrm{n,Cs-134,1}}$ =	4,7955·10^{-4} s^{-1};	$R_{\mathrm{T,Cs-134,1}}$	= 4,9727·10^{-3} s^{-1};
$p_{\gamma,\mathrm{Cs-134,1}}$ =	0,9763;	$u_{\mathrm{rel}}\big(p_{\gamma,\mathrm{Cs-134,1}}\big)$ =	0,00082;
$\varepsilon_{\mathrm{W,Cs-134,1}}$=	0,02071 Bq^{-1}·s^{-1};	$u_{\mathrm{rel}}\big(\varepsilon_{\mathrm{W,Cs-134,1}}\big)$=	0,01950;
$f_{1,\mathrm{Cs-134,1}}$ =	1,1332;	$u_{\mathrm{rel}}\big(f_{1,\mathrm{Cs-134,1}}\big)$ =	0,01209;
$f_{5,\mathrm{Cs-134,1}}$ =	0,9243;	$u_{\mathrm{rel}}\big(f_{5,\mathrm{Cs-134,1}}\big)$ =	0,01.

Gamma line 2 at 795,9 keV:

$R_{\mathrm{n,Cs-134,2}}$ = 3,835·10^{-4} s^{-1}; $R_{\mathrm{T,Cs-134,2}}$ = 4,9608·10^{-3} s^{-1};

$p_{\gamma,\mathrm{Cs-134,2}}$ = 0,854; $u_{\mathrm{rel}}(p_{\gamma,\mathrm{Cs-134,2}})$ = 0,00105;

$\varepsilon_{\mathrm{W,Cs-134,2}}$ = 0,01638 Bq^{-1}·s^{-1}; $u_{\mathrm{rel}}(\varepsilon_{\mathrm{W,Cs-134,2}})$ = 0,01632;

$f_{1,\mathrm{Cs-134,2}}$ = 1,1368; $u_{\mathrm{rel}}(f_{1,\mathrm{Cs-134,2}})$ = 0,01415;

$f_{5,\mathrm{Cs-134,2}}$ = 0,932; $u_{\mathrm{rel}}(f_{5,\mathrm{Cs-134,2}})$ = 0,01.

The standard uncertainties of the following input quantities are negligible:

t_{A} = 1,365·10^{7} s; $\lambda_{\mathrm{Cs-134}}$ = 1,640·10^{-8} s^{-1};

t_{m} = 1,314·10^{6} s.

In this example, a value of 1,08 is attributed to the factor f_{B} .

The factor φ_m and its relative standard uncertainty $u_{\mathrm{rel}}(\varphi_m)$ are obtained according the equations (13) and (21).

$$\varphi_m = \frac{\mathrm{e}^{1{,}64\cdot10^{-8}\ \mathrm{s}^{-1}\cdot 1{,}365\cdot10^{7}\ \mathrm{s}}}{0{,}05121\ \mathrm{kg}\cdot 72{,}4}\cdot\frac{1{,}64\cdot10^{-8}\ \mathrm{s}^{-1}\cdot 1{,}314\cdot10^{6}\mathrm{s}}{1-\mathrm{e}^{-1{,}64\cdot10^{-8}\mathrm{s}^{-1}\cdot 1{,}314\cdot10^{6}\ \mathrm{s}}} = 0{,}31403\ \mathrm{kg}^{-1}$$

$$u_{\mathrm{rel}}(\varphi_m) = \sqrt{0{,}004^2 + 0{,}02^2} = 0{,}020396$$

With the line-referred calibration factors $\varphi_{\mathrm{r},j}$, the activities in each of both Cs-134-gamma lines are calculated according equation (16):

$$A_{\mathrm{Cs-134,1}} = 4{,}7955\cdot10^{-4}\ \mathrm{s}^{-1}\cdot\frac{1{,}1332\cdot 0{,}9243}{0{,}02071\ \mathrm{Bq}^{-1}\cdot\mathrm{s}^{-1}\cdot 0{,}9763} =$$

$$= 4{,}7955\cdot10^{-4}\cdot 51{,}8031\ \mathrm{Bq} = 0{,}02484\ \mathrm{Bq}$$

$$A_{\mathrm{Cs-134,2}} = 3{,}835\cdot10^{-4}\ \mathrm{s}^{-1}\cdot\frac{1{,}1368\cdot 0{,}932}{0{,}01638\ \mathrm{Bq}^{-1}\cdot\mathrm{s}^{-1}\cdot 0{,}854} =$$

$$= 3{,}835\cdot10^{-4}\cdot 75{,}4051\ \mathrm{Bq} = 0{,}02905\ \mathrm{Bq}$$

The standard uncertainties of both net count rates are calculated by equations (18) and (19) neglecting the last two terms in equation (19):

$$u(R_{\mathrm{n,Cs-134,1}}) = \sqrt{\frac{4{,}7955\cdot10^{-4}\ \mathrm{s}^{-1}}{1{,}314\cdot10^{6}\mathrm{s}} + \frac{4{,}9727\cdot10^{-3}\ \mathrm{s}^{-1}}{1{,}314\cdot10^{6}\mathrm{s}}\cdot 1{,}08} = 6{,}6724\cdot10^{-5}\ \mathrm{s}^{-1}$$

$$u(R_{\mathrm{n,Cs-134,2}}) = \sqrt{\frac{3{,}835\cdot10^{-4}\ \mathrm{s}^{-1}}{1{,}314\cdot10^{6}\mathrm{s}} + \frac{4{,}9608\cdot10^{-3}\ \mathrm{s}^{-1}}{1{,}314\cdot10^{6}\mathrm{s}}\cdot 1{,}08} = 6{,}6100\cdot10^{-5}\ \mathrm{s}^{-1}$$

The relative standard uncertainties of both line-referred calibration factors $u_{\mathrm{rel}}(\varphi_{\mathrm{r},j})$ result in:

$$u_{\mathrm{rel}}(\varphi_{\mathrm{Cs-134,1}}) = \sqrt{0{,}01209^2 + 0{,}01^2 + 0{,}01950^2 + 0{,}00082^2} = 0{,}02504$$

$$u_{\mathrm{rel}}(\varphi_{\mathrm{Cs-134,2}}) = \sqrt{0{,}01415^2 + 0{,}01^2 + 0{,}01632^2 + 0{,}00105^2} = 0{,}02383$$

With these values, the variances of the activities of the two lines are calculated according to equation (17):

$$u^2(A_{\mathrm{Cs-134,1}}) = [51{,}8031^2 \cdot (6{,}6724 \cdot 10^{-5})^2 + (4{,}7955 \cdot 10^{-4})^2 \cdot 0{,}02504^2]\ \mathrm{Bq}^2 =$$

$$= 1{,}19476 \cdot 10^{-5}\ \mathrm{Bq}^2$$

$$u^2(A_{\mathrm{Cs-134,2}}) = [75{,}4051^2 \cdot (6{,}6077 \cdot 10^{-5})^2 + (3{,}835 \cdot 10^{-4})^2 \cdot 0{,}02383^2]\ \mathrm{Bq}^2 =$$

$$= 2{,}48258 \cdot 10^{-5}\ \mathrm{Bq}^2$$

Now, the variance of the Cs-134 activity is calculated according to equation (15) followed by the calculation of the standard uncertainty:

$$u^2(A_{\mathrm{Cs-134}}) = \frac{1}{\frac{1}{1{,}19476 \cdot 10^{-5}\ \mathrm{Bq}^2} + \frac{1}{2{,}48258 \cdot 10^{-5}\ \mathrm{Bq}^2}} = 8{,}06585 \cdot 10^{-6}\ \mathrm{Bq}^2$$

$$u(A_{\mathrm{Cs-134}}) = 2{,}840 \cdot 10^{-3}\ \mathrm{Bq}$$

Afterwards, the activity of Cs-134 $A_{\mathrm{Cs-134}}$ is obtained using equation (14):

$$A_{\mathrm{Cs-134}} = 8{,}06585 \cdot 10^{-6}\ \mathrm{Bq}^2 \cdot \left(\frac{0{,}02484\ \mathrm{Bq}}{1{,}19476 \cdot 10^{-5}\ \mathrm{Bq}^2} + \frac{0{,}02905\ \mathrm{Bq}}{2{,}48258 \cdot 10^{-5}\ \mathrm{Bq}^2}\right) =$$

$$= 8{,}06585 \cdot 10^{-6} \cdot\ (2079{,}08 + 1170{,}15)\ \mathrm{Bq} = 0{,}0262\ \mathrm{Bq}$$

Finally, the value and the associated standard uncertainty of the specific Cs-134 activity are determined according to equations (13) and (20):

$$a_{\mathrm{Cs-134}} = 0{,}31403\ \mathrm{kg}^{-1} \cdot 0{,}0262\ \mathrm{Bq} = 8{,}2276 \cdot 10^{-3}\ \mathrm{Bq} \cdot \mathrm{kg}^{-1}\ (\mathrm{FM})$$

$$u(a_{\mathrm{Cs-134}}) = 8{,}2276 \cdot 10^{-3} \cdot \sqrt{0{,}020396^2 + \left(\frac{2{,}840 \cdot 10^{-3}}{0{,}0262}\right)^2}\ \mathrm{Bq} \cdot \mathrm{kg}^{-1} =$$

$$= 9{,}075 \cdot 10^{-4}\ \mathrm{Bq} \cdot \mathrm{kg}^{-1}\ (\mathrm{FM})$$

Thus, the specific Cs-134-activity results in:

$$a_{\mathrm{Cs-134}} = (8{,}2276 \pm 0{,}9075) \cdot 10^{-3}\ \mathrm{Bq \cdot kg^{-1}}\ (\mathrm{FM})$$

5.3 Consideration of uncertainties

The combined standard uncertainty of the specific activity of Cs-137 amounts to be less than 10 % in the IMIS routine measurement program, where the largest contribution originates from the net count rate. If the self-attenuation correction for ash from fish flesh is neglected, another systematic error of up to 5 % will be the result.

For multi-line radionuclides such as Cs-134, the uncertainty contribution of the coincidence summing correction factor (see basic chapters γ–SPEKT/GRUNDL and γ-SPEKT/SUMESC of this procedures manual) has to be taken into account, which may lead to additional few per cents in case of significant corrections.

6 Characteristic limits of the procedure

The calculation of the characteristic limits follows the standard ISO 11929 (9, 10, 11).

For calculation of the detection limit $a_r^{\#}$ it is necessary to calculate the decision threshold of a radionuclide a_r^*, first. To solve this, explicit equations can be used in case of single-line-radionuclides (see section 5.1.1).An Excel spreadsheet (see section 7.3.1) as well as a project file for the software UncertRadio (see section 7.3.2) are available on the website of this procedures manual.

For multi-line emitters (see section 5.1.2) these equations can only be calculated with software support. At present, a corresponding project file for the program UncertRadio (see section 7.3.2) is available on the website of this procedures manual.

Further considerations concerning the characteristic limits are to be found in the general chapters ERK/NACHWEISGR-ISO-01 and ERK/NACHWEISGR-ISO-02 of these procedures manuals.

6.1 Equations

6.1.1 Decision threshold

6.1.1.1 Equations for calculation of the decision threshold of the specific activity of single-line radionuclides

The decision threshold of the single-line radionuclide's specific activity a_{r}^*, e. g. Cs-137, is determined using equation (22):

$$a_{\mathrm{r}}^* = \varphi \cdot k_{1-\alpha} \cdot \sqrt{\mu_2} = \varphi \cdot k_{1-\alpha} \cdot \sqrt{\frac{1}{t_{\mathrm{m}}} \cdot \left(R_{\mathrm{T,r}} + R_{0,\mathrm{r}}\right) + u^2\left(R_{\mathrm{T,r}}\right) + u^2\left(R_{0,\mathrm{r}}\right)} \qquad (22)$$

Herein is:

$k_{1-\alpha}$ quantile of the normal distribution associated with the type I error.

6.1.1.2 Equations of calculation of the decision threshold of the specific activity of multi-line radionuclides

The decision threshold a_r^* for multi-line radionuclides, e. g. Cs-134, may be calculated directly by equation (23):

$$a_r^* = \varphi_m \cdot k_{1-\alpha} \cdot u(A_r = 0) = \varphi_m \cdot k_{1-\alpha} \cdot \sqrt{\frac{1}{\sum_j \frac{t_m}{\varphi_{r,j}^2 \cdot R_{T,r,j} \cdot f_B}}} \tag{23}$$

6.1.2 Detection limit

6.1.2.1 Equations for calculation of the detection limit of the specific activity of single-line radionuclides

The detection limit $a_r^\#$ of single-line radionuclides is calculated with equation (24) using the decision threshold of the radionuclide's specific activity a_r^* obtained by equation (22):

$$a_r^\# = a_r^* \cdot \frac{\psi}{\theta} \cdot \left[1 + \sqrt{1 - \frac{\theta}{\psi^2}\left(1 - \frac{k_{1-\beta}^2}{k_{1-\alpha}^2}\right)}\right] \tag{24}$$

Herein is:

$k_{1-\beta}$ quantile of the normal distribution associated with the type II error.

with auxiliary quantities:

$$\theta = 1 - k_{1-\beta}^2 \cdot u_{rel}^2(\varphi)$$

$$\psi = 1 + \frac{k_{1-\beta}^2}{2 \cdot a_r^*} \cdot \frac{\varphi}{t_m}$$

6.1.2.2 Equations of calculation of the detection limit of the specific activity of multi-line radionuclides

The detection limit of the specific activity of multi-line radionuclides $a_r^\#$, e. g. Cs-134, is estimated according to equation (25).

$$a_r^\# \approx a_r^* + k_{1-\beta} \cdot u(a_r^{\#\prime}) \tag{25}$$

with the standard uncertainty of an iterated specific activity $u(a_r^{\#\prime})$:

$$u(a_r^{\#\prime}) = \sqrt{\left(\frac{a_r^*}{k_{1-\alpha}}\right) + \left[u^2(a_r) - \left(\frac{a_r^*}{k_{1-\alpha}}\right)^2\right] \cdot \frac{a_r^{\#\prime}}{a_r}} \tag{26}$$

6.1.3 Limits of the coverage interval

The calculation of the limits of the coverage interval is not required in this case.

6.2 Worked example

6.2.1 Decision threshold and detection limit of the specific activity of single-line radionuclides with Cs-137 as an example

Using the values of the input quantities from section 5.2.1 the coefficient μ_2 is derived according to equation (9):

$$\mu_2 = \frac{0{,}00271}{72000} \cdot (1 + 0{,}523)\ \mathrm{s}^{-2} = 5{,}732 \cdot 10^{-8}\ \mathrm{s}^{-2}$$

Using this value, the decision threshold of the specific activity $a^*_{\mathrm{Cs-137}}$ is obtained from the procedural calibration factor φ the normal distribution quantile $k_{1-\alpha}$ = 3:

$$a^*_{\mathrm{Cs-137}} = 34{,}655 \cdot 3{,}0 \cdot \sqrt{5{,}732 \cdot 10^{-8}}\ \mathrm{Bq} \cdot \mathrm{kg}^{-1} = 0{,}0249\ \mathrm{Bq} \cdot \mathrm{kg}^{-1}\ (\mathrm{FM})$$

According to equation (14) the detection limit of the specific activity $a^{\#}_{\mathrm{Cs-137}}$ is:

$$a^{\#}_{\mathrm{Cs-137}} = 0{,}0249 \cdot \frac{1{,}0262}{0{,}9964} \cdot \left[1 + \sqrt{1 - \frac{0{,}9964}{1{,}0262^2} \cdot \left(1 - \frac{1{,}645^2}{3{,}0^2}\right)}\right]\ \mathrm{Bq} \cdot \mathrm{kg}^{-1} =$$

$$= 0{,}0406\ \mathrm{Bq} \cdot \mathrm{kg}^{-1}\ (\mathrm{FM})$$

with values of the auxiliary quantities

$$\theta = 1 - 1{,}645^2 \cdot 0{,}03642^2 = 0{,}9964$$

$$\psi = 1 + \frac{1{,}645^2}{2 \cdot 0{,}0249} \cdot \frac{34{,}655}{72000} = 1{,}0262$$

6.2.2 Decision threshold and detection limit of the specific activity of multi-line radionuclides with Cs-134 as an example

First, the following auxiliary quantity from equation (23) is calculated using the values of quantities from section 5.2.2 and the normal distribution quantile $k_{1-\alpha}$ = 3:

$$\left(\frac{1{,}314 \cdot 10^6}{51{,}8031^2 \cdot 4{,}9727 \cdot 10^{-3} \cdot 1{,}08} + \frac{1{,}314 \cdot 10^6}{75{,}4051^2 \cdot 4{,}9608 \cdot 10^{-3} \cdot 1{,}08}\right)\ \mathrm{Bq}^{-2} =$$

$$= (91173{,}337 + 43133{,}873)\ \mathrm{Bq}^{-2} = 1{,}34307 \cdot 10^5\ \mathrm{Bq}^{-2}$$

Therewith, the decision threshold of the specific Cs-134 activity amounts to:

$$a^{*}_{\mathrm{Cs-134}} = 0{,}31403 \cdot 3{,}0 \cdot \sqrt{\frac{1}{1{,}34307 \cdot 10^{5}}}\ \mathrm{Bq \cdot kg^{-1}} = 2{,}5707 \cdot 10^{-3}\ \mathrm{Bq \cdot\ kg^{-1}}\ (\mathrm{FM})$$

This decision threshold value agrees quite well with that value obtained from using UncertRadio (see section 7.3.2). It is smaller compared to the two single-line decision threshold values, which are 3,12 mBq·kg^{-1} and 4,55 mBq·kg^{-1}, respectively.

The detection limit of the specific Cs-134 activity is estimated with a first step of iteration according to equations (26) and (25) as follows:

$$u\left(a_{\mathrm{r}}^{\#\prime}\right) = \sqrt{\frac{2{,}5707 \cdot 10^{-3}}{3{,}0} + \frac{3{,}0 + 1{,}645}{3{,}0} \cdot \frac{2{,}5707 \cdot 10^{-3}}{8{,}2276 \cdot 10^{-3}} \cdot \left[(9{,}075 \cdot 10^{-4})^{2} - \left(\frac{2{,}5707 \cdot 10^{-3}}{3{,}0}\right)^{2}\right]}\ \mathrm{Bq \cdot kg^{-1}} =$$

$$= 8{,}313 \cdot 10^{-4}\ \mathrm{Bq \cdot kg^{-1}}\ (\mathrm{FM})$$

$$a^{\#}_{\mathrm{Cs-134}} \approx\ 2{,}5707 \cdot 10^{-3} + 1{,}645 \cdot 8{,}313 \cdot 10^{-4}\ \mathrm{Bq \cdot kg^{-1}} =$$

$$= 3{,}938 \cdot 10^{-3}\ \mathrm{Bq \cdot kg^{-1}}\ (\mathrm{FM})$$

This iteratively estimated detection limit is in good agreement with the result of the software UncertRadio, 4,031·10^{-3} Bq·kg^{-1} (see section 7.3.2.2).

7 Catalogue of chemicals and equipment

7.1 Chemicals

The chemicals used should be of analytically pure quality.

— Cleaning agent: e. g. RBS-50-Super-Flüssigkonzentrat® 2 %.

7.2 Equipment

Ordinary equipment of a radiochemical laboratory.

7.2.1 Sampling

— ice container / cooling boxes;
— plastic bags;
— deep freezer (ca. -18 °C), if the samples have to be stored.

7.2.2 Sample preparation

— filleting board made of plastic;
— sharpened filleting knifes;
— cut-resistant gloves.

7.2.3 Drying and ashing

— incineration ovens with catalytical treatment of exhaust gases; organic exhaust gases shall largely be burned to CO_2 und H_2O;
— stainless steel bowls (V4A) or ceramic bowls adapted in size to the internal volume of the oven,
— transparent paper with an areal density of about 90 $g \cdot m^{-2}$ as backing material for the stainless steel containers.

7.2.4 Calibration and measurement

— gamma mitting radionuclides, preferably single-line emitters; if applicable, within a single solution covering the whole energy range to be calibrated;
— cylindrical plastic containers, e. g. made from PVC, with an inner diameter of about 7 cm and a capacity of ca. 220 ml, preferably with plane bottom;
— High-purity Germanium-semiconductor detector with a relative efficiency preferably between 20 % and 60 % relative to a 3‟ x 3‟ NaI(Tl)-crystal and a full-width-at-half-maximum less than 2,0 keV for the 1,33 MeV gamma-line of Co-60;
— measuring electronics;
— lead shielding with ca. 10 cm wall thickness.

7.2.5 Evaluation

— software for accumulating and evaluating gamma-ray spectra;
— software tools for calculating the peak and total efficiencies and correction factors for coincidence summing and self-attenuation.

7.3 Software supported calculation

7.3.1 Example of an Excel spreadsheet

7.3.1.1 Calculating the specific Cs-137 activity

Procedure for determining the specific activities of radionuclides in fish by gamma spectrometry

G-γ-SPEKT-FISCH-01 Version January 2016 / verified March 2020
Procedures manual for monitoring of radioactive substances in the environment and of external radiation (ISSN 1865-8725)

SAMPLE IDENTIFICATION: ***Variant 1: single-line radionuclide Cs-137***

#Number of parameters p	13
k_alpha	3
k_beta	1,645
gamma	0,05

Create Excel variables!

User-Input:	Input of values
	Definition Excel variables
	Input of Excel formulae
Excel-VBA:	#Keywords
	Values from Vbasic

	Data input:		variable names:			Uncertainty budget:		
	#Values of parameters p	Unit	Excel variable	**Input values**	**StdDev**	**partial derivatives**	**uncertainty budget:**	**budget in %**
p 1	**#Number of gross counts Ng**		Ng	650,16	25,4982352	0,00048129	0,0122721	63,31420646
p 2	trapezoidal BG count rate	1/s	RT	2,7100E-03	1,4030E-04	-34,6530414	0,00486195	9,937666022
p 3	b/2L		b2L	5,2300E-01	0,0000E+00	0	0	0
p 4	detection efficiency		eps	3,0560E-02	8,8624E-04	-7,16645951	0,0063512	16,95802214
p 5	emission probability, 661,7 keV		p_gamm	8,4990E-01	1,9970E-03	-0,25768561	0,0005146	0,111326787
p 6	ash mass	kg	mA	1,3170E-02	5,2680E-05	-16,6292333	0,00087603	0,322625867
p 7	ratio fresh mass / ash mass		qF	8,3540E+01	1,6708E+00	-0,00262158	0,00438014	8,06564668
p 8	Time elapsed Sampling-->StartMeasum	s	tA	1,9788E+07	0,0000E+00	1,6008E-10	0	0
p 9	half-live of Cs-137	s	thalf	9,4830E+08	0,0000E+00	-3,3466E-12	0	0
p 10	self-attenuation correction factor		_f2	9,7600E-01	7,8080E-03	0,22439265	0,00175206	1,29050605
p 11	duration of measurement	s	tm	72000	0,0000E+00	-4,346E-06	0	0
p 12	net count rate of the BG peak	1/s	RnNE	0,0000E+00	0,0000E+00		0	0
p 13	counting duration of the BG spectrum	s	tNE	72000	0,0000E+00	0	0	0
	(List can be continued here)							
	Model section		**c = phix * Rn**					
	Auxiliary equations h			(Formulae)				
h 1	**#Gross count rate Rg**	1/s	Rg	0,00903				
h 2	decay correction measuring period		_f3	9,9997E-01				
h 3	decay correction Sampling-->StartMes		_f4	1,0146E+00				
	(List can be continued here)							
	#Net count rate Rn	1/s	Rn	6,3200E-03				
	#Calibration factor, proc.dep.	Bq*s/kg	phix	3,4653E+01				
	#Value output quantity	Bq/kg	Result	2,1901E-01	0,04054204	<-- output value modifiable by VBA		
	#Combined standard uncertainty	Bq/kg	uResult	1,5423E-02				
	#Decision threshold	Bq/kg		0,024890348				
	#Detection limit	Bq/kg		0,040542029				
	further derived values							
	Auxiliary quantity Omega		Omega	1				
	Best estimate	Bq/kg	BestEst	0,219007222				
	Uncertainty best estimate	Bq/kg		0,015422984				
	Lower confidence limit	Bq/kg		0,188778729				
	Upper confidence limit	Bq/kg		0,249235714				

Calculate!

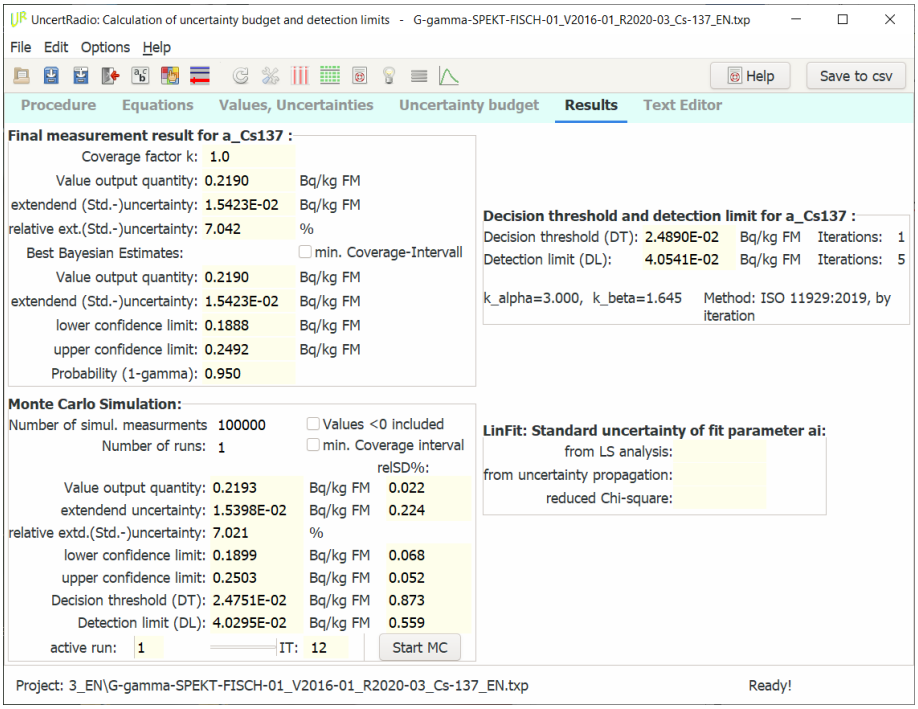
The corresponding Excel spreadsheet is available on the website of this procedures manual.

7.3.1.2 Calculating the specific Cs-134 activity

Because of the complexity of the calculations an Excel file is not available.

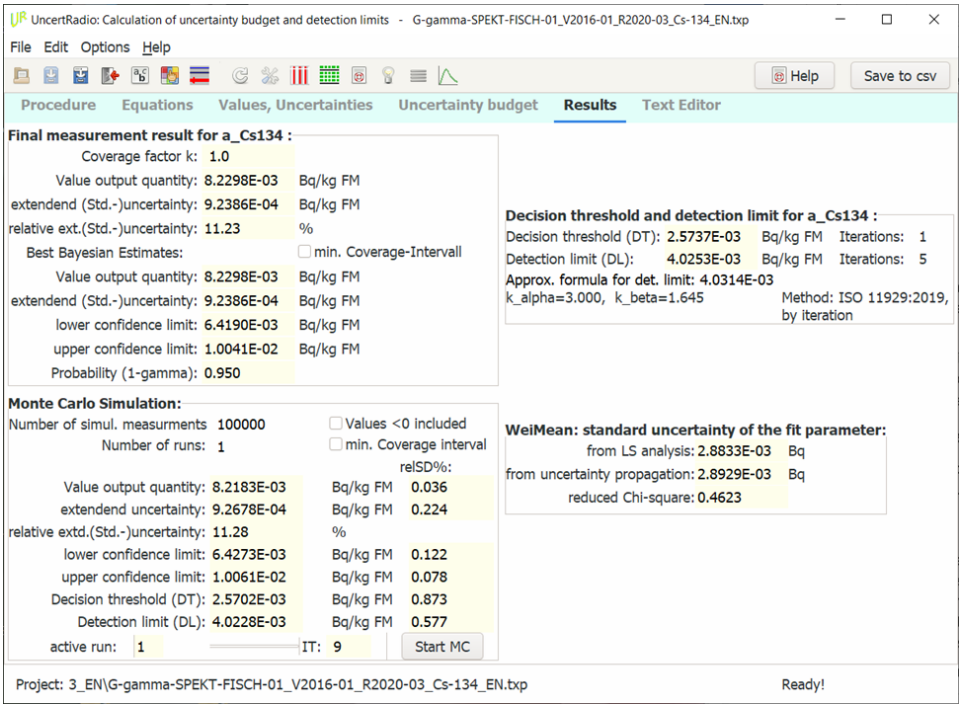
7.3.2 Example of an UncertRadio project

7.3.2.1 Calculating the specific Cs-137 activity



The corresponding UncertRadio project file is available on the website of this procedures manual.

7.3.2.2 Calculating the specific Cs-134 activity



The corresponding UncertRadio project file is available on the website of this procedures manual.

References

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