Procedure for the continuous monitoring of the artificial gross gamma activity concentration in seawater

D-γ-GESAMT-MWASS-01

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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 the continuous monitoring of the artificial gross gamma activity concentration in seawater

1 Scope

The procedure outlined in the following is used as a screening method for almost real-time, continuous monitoring of even low activity concentrations of artificial, gamma-emitting radioactive substances in seawater.

In order to ensure a comprehensive monitoring, a measuring network of probes was set up in the environmental area "seawater" that meets the requirements of the general administrative regulation for the "Integrated Measurement and Information System for Monitoring Environmental Radioactivity" (AVV-IMIS) according to the Radiation Protection Act (StrlSchG) [1, 2, 3, 4].

In the open sea, the probes are stationary on oceanographic measuring buoys, whereas onshore they are fixed on permanent facilities such as piers or gauge houses where they are hanging freely in the seawater [5]. In addition, vessels of the Federal Maritime and Hydrographic Agency (BSH) are equipped as movable stations with a corresponding measuring device where the probe is mounted in a recess in the vessel's bottom (strom box).

With this procedure, an artificial gross gamma activity concentration of about 800 $Bq \cdot m^{-3}$ can be detected.

2 Sampling

For this procedure, sampling is not required.

3 Analysis

3.1 Principle of the procedure

With this integral procedure, the count rates resulting from the interaction of gamma radiation in seawater with thallium-doped sodium iodide (NaI(Tl)) detectors are determined directly in a quasi-continuous way, viz. cyclically with a measurement duration of one hour.

The radiation arising from artificial radioactive substances is determined from the ratio of the count rates in two defined energy ranges of the pulse height spectrum below and above 900 keV (see Figure 3). The current background radiation is recorded separately in both energy ranges. In routine operation, the proportions of radiation in the two energy ranges have a fixed ratio to one another.

When artificial radioactive substances are introduced into the environment, the count rate ratio changes in favor of the lower energy range. The determined count rate ratio is used to calculate the artificial gross gamma activity concentration.

The threshold value for the net count rate due to artificial gamma radiation in the energy range A, $R_{\mathrm{n,K,A}}$ is 3 s^{-1}, which corresponds to a gross gamma activity concentration of 1000 $\mathrm{Bq \cdot m^{-3}}$. If the threshold value is exceeded, the federal coordinating office is automatically informed electronically.

The procedure also allows the recording and evaluation of pulse height spectra in the form of hourly spectra, which can be summed up to form daily or weekly spectra if required. Although the low energy resolution of the NaI(Tl) detectors only permits a nuclide-specific evaluation to a limited extent, individual radionuclides such as caesium-137 and radionuclide mixtures typical for accidents can be recognized well and differentiated from natural radioactive substances due to the different gamma radiation energies. The hourly and daily spectra are used also to guarantee the proper operation of the measuring systems and to ensure the data quality.

The nuclide-specific evaluation of the spectra and an estimation of the activity concentrations of existing artificial radionuclides are described in the procedure D-γ-GESAMT-MWASS-02.

3.2 Sample preparation

A sample preparation is not required.

3.3 Radiochemical separation

A radiochemical separation is not required.

4 Measuring the activity

4.1 General

The activity concentrations of artificial, gamma-emitting radioactive substances in seawater are estimated using the procedural measurand artificial gross gamma activity concentration $c_{\mathrm{G\gamma}}$. Due to the complexity of the evaluation, this artificial gross gamma activity concentration $c_{\mathrm{G\gamma}}$ differs somewhat from the definition in the glossary of this Procedures Manual. In the case of a free-hanging measuring device, it does not relate to a reference radionuclide used in the calibration, but approximately to a fictitious radionuclide that emits one gamma quantum with each radioactive decay. The radioactive decay can therefore not be taken into account in this screening method.

A NaI(Tl) detector is used to detect the gamma radiation. The measuring device (Figure 1) was adapted to the special requirements of the measuring network operation for the marine environment. The technical data for such a measuring device are listed in Section 8.2.

For adequate shielding of the measuring device against secondary cosmic and sediment radiation through the water column, the measuring device must be installed in a position that has a minimum water depth of 3 m with a water coverage of the probe of at least 1,5 m.

At stations close to the coast, the water coverage is recorded by a pressure sensor. The operation of these stations is interrupted when the water coverage of the probe falls below 1 m. Corresponding correction factors are used if the water coverage is between 1 m and 1,5 m.

Note:
With this special measuring arrangement of a probe with NaI(Tl) detector hanging in seawater, the following applies: Through interaction with water (instead of air), the energy of the primary gamma radiation from the water is significantly reduced over distances of sometimes a hundred centimeters before the release of the energy in the detector can take place. As a result, the photo peak in relation to the Compton continuum is much smaller than would be expected, for example, from a measurement with a point source in air. This proves to be an advantage for the gross gamma measurement in water, because even for gamma emitters such as zinc-65 or cobalt-60, with gamma energies from 1116 keV to 1333 keV, the main part of the measured count rate falls in the energy range below 900 keV. This means that the energy range below 900 keV in the pulse height spectrum is sufficiently sensitive for the detection of artificial gamma radiation.

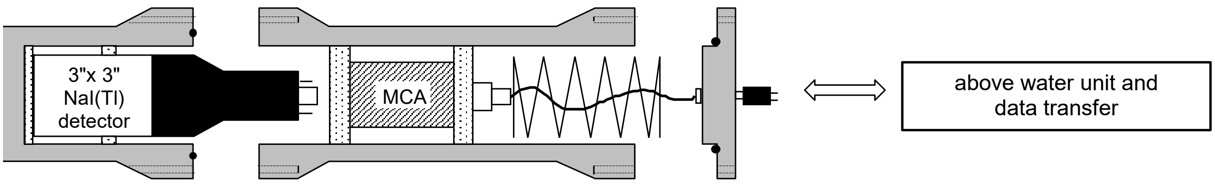


Fig. 1: Measuring device for determining the artificial gross gamma activity concentration in seawater.

4.2 Calibration

In case of contamination of seawater, it is to be expected that the origin and type of the radionuclides or radionuclide mixtures involved are unknown. Therefore, a calibration with several radionuclides is carried out. In order to be able to universally detect gamma-emitting radionuclides with the calibration, the energy-dependent gamma-ray efficiency of the detector of the measuring device is calibrated instead of a nuclide-specific calibration, depending on whether the probe hangs freely in the water or is in the vessel's strom box.

The validity of the calibration must be ensured by checking the characteristics of the probes regularly.

4.2.1 Calibration of the measuring arrangement “probe hanging freely in the water”

In the measurement arrangement "probe hanging freely in the water", the gamma quanta arise by radioactive decay in the surrounding large volume of water. These are absorbed or scattered by interactions in water, mainly caused by the photo or Compton effect. Therefore, they rarely reach the device directly.

The procedural gross efficiency, which depends on the energy of the gamma radiation, is defined as the ratio of all impulses registered in the observed energy range per time and per water volume to the number of photons emitted per time.

$$\varepsilon_{\mathrm{t}}\left(E_{\gamma}\right)=\frac{R_{\mathrm{t,n}}}{c\cdot p_{\gamma}\left(E_{\gamma}\right)} \tag{1}$$

Herein are:

$\varepsilon_{\mathrm{t}}(E_{\gamma})$	procedural gross efficiency, dependent on the energy of the gamma radiation E_{γ} of the reference nuclide, in $m^3 \cdot Bq^{-1} \cdot s^{-1}$;
$R_{\mathrm{t,n}}$	ross net count rate in the considered energy range, in s^{-1};
c	activity concentration, in $Bq \cdot m^{-3}$;
$p_{\gamma}\ (E_{\gamma})$	emission intensity of the gamma line.

By means of a single energy-dependent calibration with the radionuclides K-40, Mn-54, Ru-106/Rh-106 and Ce-144, it was possible to show that the procedural gross efficiency $\varepsilon_{\mathrm{t}}(E_{\gamma})$ is almost constant with an average value of $3 \cdot 10^{-3}\ m^3 \cdot Bq^{-1} \cdot s^{-1}$ in the considered energy range between 0,3 MeV and 1,5 MeV, in which the gamma radiation energies of relevant radionuclides are located (see Table 1 and Figure 2).

Note:
The implementation of this single calibration is explained in detail in Annex A. This calibration can also be carried out using Monte Carlo simulation.

Figure 2 shows the efficiencies calculated for the individual gamma radiation energies and the calibration curve interpolated from them.

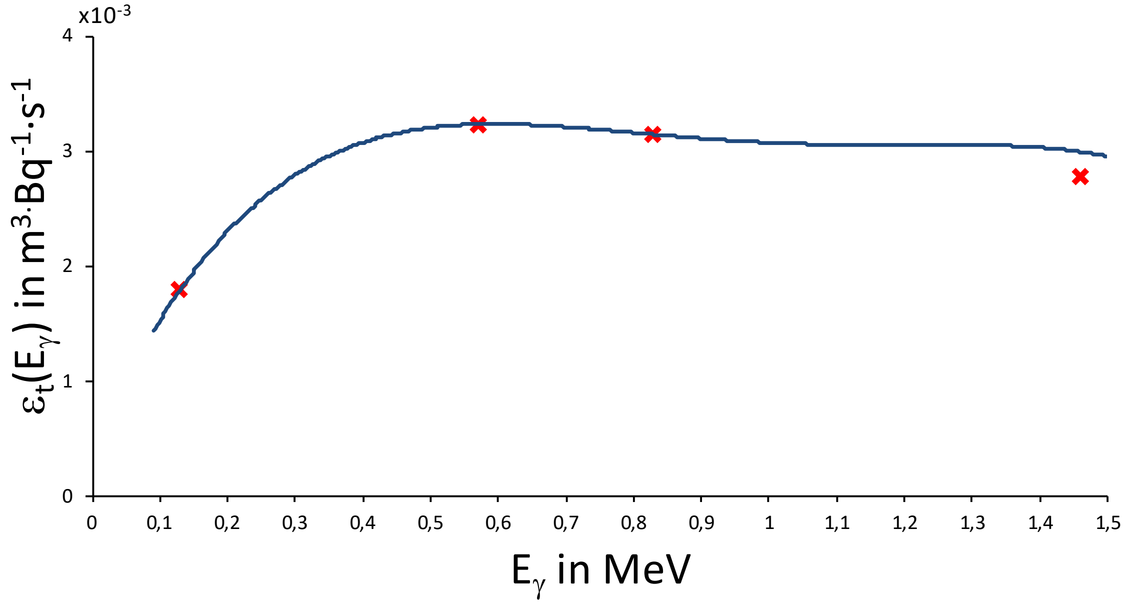


Fig. 2: Representation of the energy-dependent procedural gross efficiency $\varepsilon_t(E_\gamma)$ for the measuring arrangement "probe hanging freely in the water".

This means, on the one hand, that a calibration with potassium-40 (K-40) alone is generally sufficiently representative for the measurement arrangement "probe hanging freely in the water", and on the other hand that the energy-dependent procedural gross efficiency $\varepsilon_t(E_\gamma)$ is almost independent of energy.

Note:

In simple terms, $\varepsilon_t(E_\gamma)$ consists of two factors:

a) the energy-dependent attenuation of a gamma quantum on the way through the seawater to the detector surface;

b) the energy-dependent efficiency that a gamma quantum that has reached the detector is also detected in it.

Factor a) decreases with increasing gamma radiation energy, so that a larger measurement volume is recorded. On the other hand, factor b) increases with decreasing gamma radiation energy, so that the incoming gamma radiation from the decreasing measurement volume is measured increasingly more effectively. These two factors are opposing; their product therefore remains almost constant in the described measuring arrangement in the energy range of 0,3 MeV to 1,5 MeV.

In Table 1, the procedural gross efficiencies $\varepsilon_t(E_\gamma)$ from Figure 2 according to Equation (1) for the gamma radiation energies of relevant radionuclides are listed.

Tab. 1: Selection of relevant radionuclides with associated calibration data for the calibration of the measuring arrangement "probe hanging freely in the water"

Radionuclide	Radionuclide data [6]		Procedural gross efficiency
	E_γ	p_γ	$\varepsilon_t(E_\gamma)$
	in MeV		in $m^3 \cdot Bq^{-1} \cdot s^{-1}$
K-40	1,46	0,11	$2{,}8 \cdot 10^{-3}$
Mn-54	0,83	1,0	$3{,}2 \cdot 10^{-3}$
Zn-65	1,12	0,50	$3{,}1 \cdot 10^{-3}$
Ru-106	0,51	0,21	$3{,}2 \cdot 10^{-3}$
	0,62	0,10	$3{,}2 \cdot 10^{-3}$
	1,06	0,015	$3{,}1 \cdot 10^{-3}$
I-131	0,36	0,81	$3{,}0 \cdot 10^{-3}$
	0,64	0,07	$3{,}2 \cdot 10^{-3}$
Cs-134	0,60	0,98	$3{,}2 \cdot 10^{-3}$
	0,80	0,85	$3{,}2 \cdot 10^{-3}$
	0,57	0,15	$3{,}2 \cdot 10^{-3}$
Cs-137	0,66	0,85	$3{,}2 \cdot 10^{-3}$
Ce-144	0,13	0,11	$1{,}9 \cdot 10^{-3}$

From column 4 in Table 1 it is evident that any radionuclide can be used as a radiation source which has at least a gamma radiation energy between 0,3 MeV and 1,5 MeV. K-40 is usually used as the reference nuclide for calibration because, in addition to its property as a single-line nuclide, it also fulfills the following criteria:

— high solubility in water;

— unproblematic procurement in larger quantities;

— manageability outside radiation protection regulations.

For the calculation of the artificial gross gamma activity concentration, the reciprocal value of the procedural gross efficiency $\varepsilon_t(E_\gamma)$ is used as procedural calibration factor φ:

$$\varphi = \frac{1}{3{,}0 \cdot 10^{-3}\ \mathrm{m}^3 \cdot \mathrm{Bq}^{-1} \cdot \mathrm{s}^{-1}} = 333{,}33\ \mathrm{Bq} \cdot \mathrm{s} \cdot \mathrm{m}^{-3}$$

4.2.2 Calibration of the measurement arrangement "probe in the vessel's strom box"

A calibration of probes mounted in the vessel's strom box cannot be carried out in the laboratory because of the measurement geometry. For this reason, the vessel-specific calibration is carried out at sea and is applied for all identically constructed strom boxes.

Note:

The implementation of this calibration is described in Annex A. This calibration can also be carried out using Monte Carlo simulation.

The procedural, gross efficiency $\varepsilon_{\mathrm{t}}(E_{\gamma})$ of the gamma radiation energy of K-40 is also calculated according to Equation (1); however, it is based on a different measurement geometry. The following value applies to the vessel's strom box used in the federal coordinating office:

$$\varepsilon_{\mathrm{t}}\left(E_{\gamma}\right) = 1{,}185 \cdot 10^{-3}\ \mathrm{m}^3 \cdot \mathrm{Bq}^{-1}\ \cdot \mathrm{s}^{-1}$$

When calculating the procedural, gross efficiency for the measurement arrangement "probe in the vessel's strom box" according to Equation (1), the count rates measured in seawater serve as the gross count rate. The count rates obtained in freshwater are used as the background count rate. The activity concentration of K-40 is given by the salt content in the seawater, see Annex A.3 and literature [4].

To calculate the artificial gross gamma activity concentration, the reciprocal value of $\varepsilon_{\mathrm{t}}(E_{\gamma})$ is used for the procedural calibration factor φ:

$$\varphi = \frac{1}{1{,}185 \cdot 10^{-3}\ \mathrm{m}^3 \cdot \mathrm{Bq}^{-1} \cdot \mathrm{s}^{-1}} = 843{,}88\ \mathrm{Bq} \cdot \mathrm{s} \cdot \mathrm{m}^{-3}$$

4.3 Measurement

The measurement is carried out continuously in seawater. The difference between the gamma radiation energies of natural and artificial radionuclide mixtures described in Section 3.1 is used for the evaluation. For this purpose, the count rate ratio of two count rates $R_{\mathrm{g,A}}$ and $R_{\mathrm{g,B}}$ of the energy ranges A and B is determined, whereby in the energy range A only gamma radiation with energies below 900 keV is registered (Figure 3); the gamma radiation energies of artificial radioactive substances are predominantly in this energy range. If natural radioactive substances from the uranium-radium decay series are introduced, the count rates of the two energy ranges increase in a ratio characteristic for the spectrum of the Rn-222 decay products.

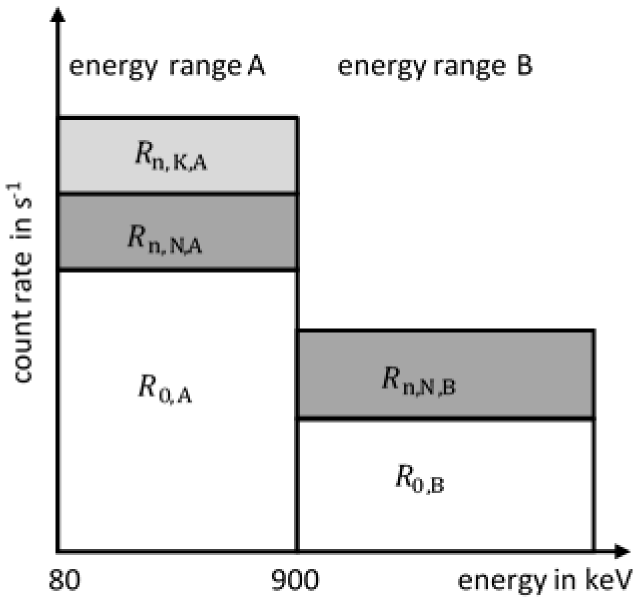


Fig. 3: Schematic presentation of the separation of the total gross count rate R_g in the gamma spectrum into two gamma radiation energy ranges A and B.

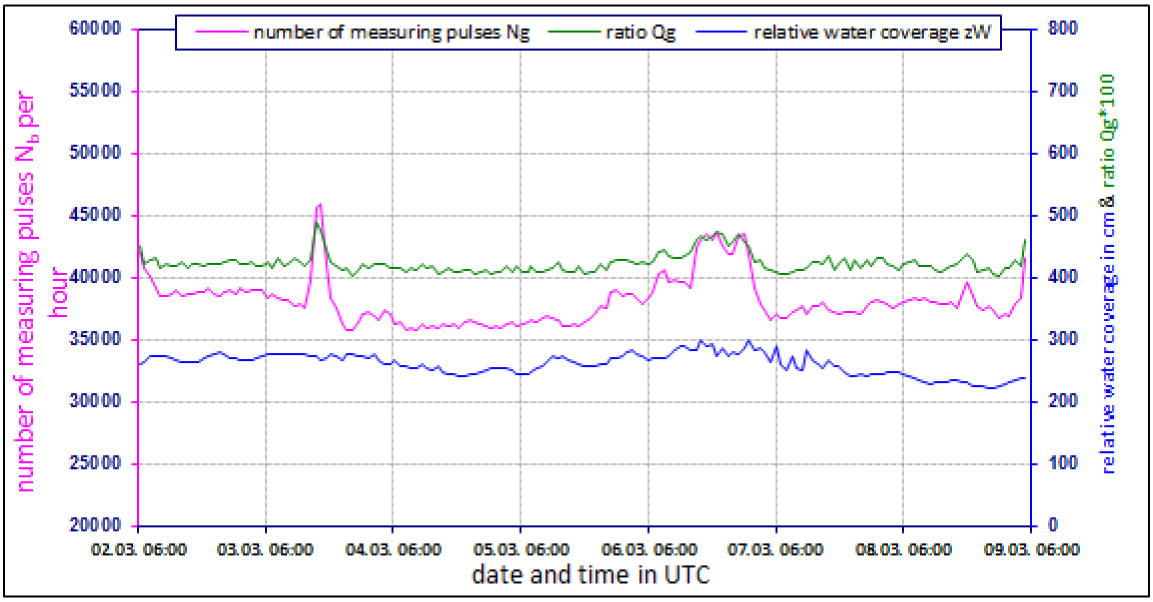
The calculated values of the total gross count rate $R_g = R_{g,A} + R_{g,B}$ and the count rate ratio, $q_g = R_{g,A}/R_{g,B}$, the pulse height spectrum and in case of coastal stations also the water coverage of the probe are calculated by the device every hour and automatically transmitted from the measuring station, e. g. via mobile communications, to the central computer of the federal coordinating office and are pre-evaluated there.

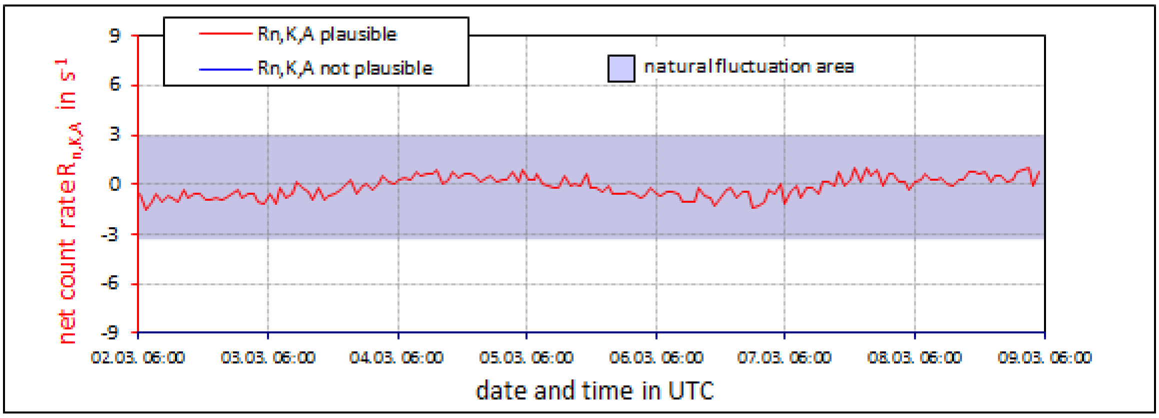
If, as part of the automatic pre-evaluation, the threshold value for the net count rate $R_{n,K,A}$ is exceeded by 3 s^{-1} due to artificial gamma radiation in the energy range A, the federal coordinating office is alerted electronically so that the data sets can be checked as quickly as possible and possible system errors, e. g. in data transmission or hardware, may be excluded.

Within the IMIS routine operation mode, the data sets are generally graphically displayed every working day, within the IMIS intensive operation mode every hour. The data is evaluated manually, checked for plausibility and the results are transmitted to IMIS. An evaluation of the daily spectrum of the previous day is possible for comparison purposes.

Figure 4 shows an example of the graphic presentation of a weekly data set.

Note:
If the graphical presentation gives indications of an entry of artificial radionuclides, a nuclide-specific evaluation of artificial radionuclides according to D-γ-GESAMT-MWASS-02 can be carried out in the corresponding pulse height spectra to estimate their activity concentration.





	Daily mean values according to AVV-IMIS							**week**
	02.03.	**03.03.**	**04.03.**	**05.03.**	**06.03.**	**07.03.**	**08.03.**	**10**
$R_{n,K,A}$ in s^{-1}	- 0,79	- 0,29	0,49	- 0,13	- 0,67	0,13	0,45	- 0,12
$c_{G\gamma}$ in Bq·m^{-3}	- 236,88	- 88,29	148,14	- 38,69	- 202,45	38,09	135,80	- 34,92
$u_{rel}(c_{G\gamma})$ in %	9	24	14	55	11	57	16	23

Fig. 4: Measurement series at the Kühlungsborn station:

a) above: purple - total gross count number N_g per hour,
green – ratio q_g,
blue - relative water coverage z_w;

b) middle: blue-gray - natural fluctuation area,
red - net count rate due to artificial gamma radiation in the energy range A, $R_{n,K,A}$;

c) below: tabular presentation of the daily mean values of the net count rate as a result of artificial gamma radiation in the energy range A, $R_{n,K,A}$, the calculated artificial gross gamma activity concentration $c_{G\gamma}$ and the related relative standard uncertainty $u_{rel}(c_{G\gamma})$.

4.4 Background

4.4.1 Environmental component

The in-situ monitoring of the artificial gross gamma activity concentration is generally more difficult as the intensity of the natural background radiation is variable. This is mainly influenced by two aspects:

— Due to heavy rainfall events, high concentrations of natural radionuclides occur temporarily in seawater, in particular of Rn-222 decay products from the uranium-radium decay series.

  Note:
  Further information on the gamma spectrometric determination of natural radionuclides can be found in the general chapter γ-SPEKT/NATRAD of these procedure manuals.

— A fouling of the probes, for example with barnacles, algae and mussels, increases the background count rate, as the radioactive substances accumulate in the fouling. This effect can be reduced by an antifouling paint or coating to protect against fouling and by removing the fouling every six months.

In individual cases, the secondary cosmic radiation and the turbidity of the water can have a minor impact on the count rates of the background radiation.

4.4.2 Device inherent component

4.4.2.1 Background during calibration

To determine the procedural, gross efficiency $\varepsilon_t(E_\gamma)$ according to Equation (1), different background count rates are used for the measuring arrangements "probe hanging freely in the water" and "probe in the vessel's strom box":

— for the measurement arrangement "probe hanging freely in the water", the background count rate of tap water is used,

— for the measurement arrangement "probe in the vessel's strom box", the background count rate, which is measured when traveling on an inland waterway in fresh water, is used.

This is explained in more detail in Annex A.

4.4.2.2 Background during measurement

In contrast to the definition of the background according to the glossary of this Procedures Manual, the background in the present procedure is the total gross count rate registered by the measuring device as a result of the radiation from the vicinity of the detector, as long as there is no additional contribution from artificial radionuclides due to a current event.

5 Calculation of the results

5.1 Equations

From the pulse height spectrum transmitted every hour, the total gross count number N_{g} and the quotient q_{g} calculated from the count rates of the two energy ranges A and B are used for evaluation.

5.1.1 Output quantity

According to Equation (2), the total gross count rate R_{g} is the sum of all gross count rates in the energy ranges A and B:

$$R_{\mathrm{g}} = R_{\mathrm{g,A}} + R_{\mathrm{g,B}} \tag{2}$$

Herein are:

R_{g} total gross count rate, in s^{-1};

$R_{\mathrm{g,A}}$ gross count rate in the energy range A, in s^{-1};

$R_{\mathrm{g,B}}$ gross count rate in the energy range B, in s^{-1}.

It can also be calculated using the total gross count number according to Equation (3):

$$R_{\mathrm{g}} = \frac{N_{\mathrm{g}}}{t_{\mathrm{m}}} = \frac{N_{\mathrm{g,A}} + N_{\mathrm{g,B}}}{t_{\mathrm{m}}} \tag{3}$$

Herein are:

N_{g} total gross count number;

$N_{\mathrm{g,A}}$ gross count number in the energy range A;

$N_{\mathrm{g,B}}$ gross count number in the energy range B;

t_{m} duration of measurement, in s.

In routine operation mode, the total gross count rate R_{g} usually corresponds to the sum of the individual background count rates and thus to the total background count rate R_0:

$$R_{\mathrm{g}} = R_{0,\mathrm{A}} + R_{0,\mathrm{B}} = R_0 \tag{4}$$

Herein are:

R_0 total background count rate, in s^{-1};

$R_{0,\mathrm{A}}$ background count rate in the energy range A, in s^{-1};

$R_{0,\mathrm{B}}$ background count rate in the energy range B, in s^{-1}.

If natural radioactive substances, e. g. from the uranium-radium decay series, and artificial radioactive substances get into the seawater, the composition of the gross count rates changes in the individual energy ranges. In energy range A, which records gamma radiation energies up to an energy of 900 keV, the gross count rate, $R_{\mathrm{g,A}}$, continuously measured in this energy range, is made up of the following components:

$$R_{\mathrm{g,A}} = R_{\mathrm{0,A}} + R_{\mathrm{n,N,A}} + R_{\mathrm{n,K,A}} \tag{5}$$

Herein are:

$R_{\mathrm{n,N,A}}$ net count rate as a result of natural gamma radiation in the energy range A, in s^{-1};

$R_{\mathrm{n,K,A}}$ net count rate as a result of artificial gamma radiation in the energy range A, in s^{-1}.

In energy range B, the artificial radioactive substances make a negligible contribution to the gross count rate, $R_{\mathrm{g,B}}$, which can be described using Equation (6).

$$R_{\mathrm{g,B}} = R_{\mathrm{0,B}} + R_{\mathrm{n,N,B}} \tag{6}$$

with

$R_{\mathrm{n,N,B}}$ net count rate as a result of natural gamma radiation in energy range B, in s^{-1}.

For the calculation of the artificial gross gamma activity concentration, both energy ranges must be put in relation to one another. For this purpose, three ratios are formed:

— the ratio q_0 from the background effects of the two energy ranges according to Equation (7):

$$q_0 = \frac{R_{\mathrm{0,A}}}{R_{\mathrm{0,B}}} \tag{7}$$

Note:
If required, the quotient q_0 is determined as a station-specific mean value over the period of a day without any particular abnormalities in the measurement data.

— the ratio $q_{\mathrm{n,N}}$ from the net count rates due to natural gamma radiation according to Equation (8):

$$q_{\mathrm{n,N}} = \frac{R_{\mathrm{n,N,A}}}{R_{\mathrm{n,N,B}}} \tag{8}$$

Note:
The ratio $q_{\mathrm{n,N}}$ for the natural radionuclides is calculated once with the help of Equation (9) from the measurement in the event of a strong input of natural radioactive substances, e.g. in a thunderstorm.

— the ratio q_{g} of the gross count rates of the energy ranges A and B according to Equation (9):

$$q_{\mathrm{g}} = \frac{R_{\mathrm{g,A}}}{R_{\mathrm{g,B}}} = \frac{R_{\mathrm{g}}}{R_{\mathrm{g,B}}} - 1 \tag{9}$$

From the Equations (2) to (9), the net count rate $R_{\mathrm{n,K,A}}$ due to artificial gamma radiation can be represented by transformation according to Equation (10):

$$R_{\mathrm{n,K,A}} = R_{\mathrm{g}} \cdot \left(1 - \frac{1 + q_{\mathrm{n,N}}}{1 + q_{\mathrm{g}}}\right) - R_0 \cdot \left(1 - \frac{1 + q_{\mathrm{n,N}}}{1 + q_0}\right) = R_{\mathrm{g}} \cdot f_1 - R_0 \cdot f_3 \tag{10}$$

Equations (9) and (2) can be used to eliminate the related values q_{g} and R_{g} by reducing them to $R_{\mathrm{g,A}}$ and $R_{\mathrm{g,B}}$, where $R_{\mathrm{g,A}}$ takes on the role of the gross count rate.

$$R_{\mathrm{n,K,A}} = R_{\mathrm{g,A}} - R_{\mathrm{g,B}} \cdot q_{\mathrm{n,N}} - R_0 \cdot \left(1 - \frac{1 + q_{\mathrm{n,N}}}{1 + q_0}\right) = R_{\mathrm{g,A}} - R_{\mathrm{g,B}} \cdot q_{\mathrm{n,N}} - R_0 \cdot f_3 \tag{11}$$

The shielding of the secondary cosmic radiation varies with fluctuating water coverage of the probe and affects the registered total background count rate R_0. In order to mathematically eliminate such background changes, the current water coverage z_{w} is registered every hour at the affected coastal stations using a pressure sensor and compared to the mean water coverage $\bar{z}_{\mathrm{w}}$. At these stations a corrected total background count rate R_0' with an empirically determined linear attenuation coefficient μ according to Equation (12) is used instead of the total background count rate R_0.

$$R_0' = R_0 \cdot \mathrm{e}^{\mu \cdot (\bar{z}_{\mathrm{w}} - z_{\mathrm{w}})} = R_0 \cdot f_4 \tag{12}$$

Note:

a) The linear attenuation coefficient μ is empirically determined for each station and is between 0,012 dm^{-1} and 0,004 dm^{-1}.

b) The correction factor f_4 is only taken into account at water coverages between 10 dm and 15 dm; in this case, its value is not equal to 1. If the water coverage is greater than 15 dm, the correction factor f_4 takes the value 1.

c) If the water coverage is less than 10 dm, the measured values are discarded.

Therefore, when the water levels change, the value R_0 from Equation (11) is replaced by the corrected value for R_0' from Equation (12) to calculate $R_{\mathrm{n,K,A}}$.

$$R_{\mathrm{n,K,A}} = R_{\mathrm{g,A}} - R_{\mathrm{g,B}} \cdot q_{\mathrm{n,N}} - R_0' \cdot \left(1 - \frac{1 + q_{\mathrm{n,N}}}{1 + q_0}\right) = R_{\mathrm{g,A}} - R_{\mathrm{g,B}} \cdot q_{\mathrm{n,N}} - R_0 \cdot f_3 \cdot f_4 \tag{13}$$

The artificial gross gamma activity concentration in seawater $c_{\mathrm{G\gamma}}$ is calculated using the net count rate determined by means of Equation (13) as a result of artificial gamma radiation $R_{\mathrm{n,K,A}}$ according to Equation (14):

$$c_{\mathrm{G\gamma}} = \varphi \cdot R_{\mathrm{n,K,A}} = \varphi \cdot \left(R_{\mathrm{g,A}} - R_{\mathrm{g,B}} \cdot q_{\mathrm{n,N}} - R_0 \cdot f_3 \cdot f_4\right) \tag{14}$$

In the Equations (10) to (14) are:

$c_{\mathrm{G\gamma}}$ artificial gross gamma activity concentration, in Bq·m^{-3};

φ procedural calibration factor, in Bq·s·m^{-3},
with
φ = 333,33 Bq · s · m^{-3} for the probe hanging freely in the water;
φ = 843,88 Bq · s · m^{-3} for the probe in the vessel's strom box;

$R_{\mathrm{n,K,A}}$ net count rate due to artificial gamma radiation in energy range A, in s^{-1};

μ linear attenuation coefficient, in dm^{-1};

z_{w} current water coverage, in dm;

$\bar{z}_{\mathrm{w}}$ medium water coverage, in dm;

f_1 procedural weighting factor of the total gross count rate:

$$f_1 = 1 - \frac{1 + q_{\mathrm{n,N}}}{1 + q_{\mathrm{g}}} = \frac{R_{\mathrm{g,A}} - R_{\mathrm{g,B}} \cdot q_{\mathrm{n,N}}}{R_{\mathrm{g}}}$$

f_3 procedural weighting factor of the total background count rate:

$$f_3 = 1 - \frac{1 + q_{\mathrm{n,N}}}{1 + q_0}$$

f_4 procedural weighting factor of the total background count rate with changing water coverage:

$$f_4 = \mathrm{e}^{\mu \cdot (\bar{z}_{\mathrm{w}} - z_{\mathrm{w}})}$$

with f_4 = 1 in case of a water coverage above 15 dm.

The radioactive decay cannot be taken into account in this screening method because the radionuclide composition is unknown.

5.1.2 Standard uncertainty of the output quantity

To calculate the standard uncertainty of the output quantity, the standard uncertainties $u(N_{\mathrm{g,A}})$ and $u(N_{\mathrm{g,B}})$ must be calculated first:

$$u\left(N_{\mathrm{g,A}}\right) = \sqrt{N_{\mathrm{g,A}}} \tag{15}$$

$$u\left(N_{\mathrm{g,B}}\right) = \sqrt{N_{\mathrm{g,B}}} \tag{16}$$

The calculation of the standard uncertainty ($c_{G\gamma}$) of the artificial gross gamma activity concentration is not trivial and is shown in Equation (17) using individual terms T_i.

$$u(c_{G\gamma}) = \sqrt{T_1 + T_2 + T_3 \cdot (T_4 + T_5 + T_6 + T_7 + T_8 + T_9)} \tag{17}$$

The individual terms T_i are described in the Equations (18) to (26):

$$T_1 = c_{G\gamma}^2 \cdot u_{\mathrm{rel}}^2(\varphi) \tag{18}$$

$$T_2 = c_{G\gamma} \cdot \frac{\varphi}{t_{\mathrm{m}}} \tag{19}$$

$$T_3 = \varphi^2 \tag{20}$$

$$T_4 = \frac{1}{t_{\mathrm{m}}} \cdot \left(R_{\mathrm{g,B}} \cdot q_{\mathrm{n,N}} + R_0 \cdot f_3 \cdot f_4\right) \tag{21}$$

$$T_5 = q_{\mathrm{n,N}}^2 \cdot \frac{R_{\mathrm{g,B}}}{t_{\mathrm{m}}} \tag{22}$$

$$T_6 = f_3^2 \cdot f_4^2 \cdot \frac{R_0}{t_0} \tag{23}$$

$$T_7 = \left(\frac{R_0 \cdot f_4}{1 + q_0} - R_{\mathrm{g,B}}\right)^2 \cdot u^2(q_{\mathrm{n,N}}) \tag{24}$$

$$T_8 = \frac{R_0^2 \cdot f_4^2 \cdot \left(1 + q_{\mathrm{n,N}}\right)^2}{(1 + q_0)^4} \cdot u^2(q_0) \tag{25}$$

$$T_9 = R_0^2 \cdot f_3^2 \cdot u^2(f_4) \tag{26}$$

where

$u(c_{G\gamma})$ standard uncertainty of the artificial gross gamma activity concentration, in Bq·m^{-3};

$u_{\mathrm{rel}}(\varphi)$ relative standard uncertainty of the procedural calibration factor;

$u_{\mathrm{rel}}(q_{\mathrm{n,N}})$ relative standard uncertainty of the ratio due to natural gamma radiation;

$u(f_4)$ standard uncertainty of the correction factor for the attenuation of the secondary cosmic radiation,

with: $u(f_4) = f_4 \cdot \sqrt{(\bar{z}_{\mathrm{w}} - z_{\mathrm{W}})^2 \cdot u^2(\mu) + \mu^2 \cdot \left(u^2(\bar{z}_{\mathrm{w}}) + u^2(z_{\mathrm{W}})\right)}$

t_{m} duration of measurement, in s;

t_0 duration of the background measurement, in s.

The equation for calculating the standard uncertainty in the case of variable water coverage is omitted here due to the complexity of the calculation.

5.2 Worked example

In this worked example, the interim results and the result are given with four significant digits. Deviations from the calculated values are possible when using another number of significant digits.

The following numerical values are used for the worked example with the measurement arrangement "probe hanging freely in the water":

N_0	=	32 004;	$N_{\mathrm{g,A}}$	=	52 346;
t_0	=	3 600 s;	$N_{\mathrm{g,B}}$	=	7 992;
R_0	=	8,89 s^{-1};	t_{m}	=	3 600 s;
φ	=	333,33 $\mathrm{Bq \cdot s \cdot m^{-3}}$;	$u_{\mathrm{rel}}(\varphi)$	=	0,05;
q_0	=	3,39;	$u_{\mathrm{rel}}(q_0)$	=	0,01;
$q_{\mathrm{n,N}}$	=	14;	$u_{\mathrm{rel}}(q_{\mathrm{n,N}})$	=	0,01;
f_3	=	-2,417;			
f_4	=	1,0665;	$u(f_4)$	=	0,001925.

This means for the net count rate due to artificial gamma radiation in energy range A according to Equation (13):

$$R_{\mathrm{n,K,A}} = \frac{52\,346}{3\,600\ \mathrm{s}} - \frac{7\,992}{3\,600\ \mathrm{s}} \cdot 14 - 8{,}89\ \mathrm{s}^{-1} \cdot (-2{,}417) \cdot 1{,}0665 = 6{,}377\ \mathrm{s}^{-1}$$

The gross gamma activity concentration $c_{\mathrm{G\gamma}}$ is obtained from Equation (14):

$$c_{\mathrm{G\gamma}} = 6{,}377\ \mathrm{s}^{-1} \cdot 333{,}33\ \mathrm{Bq \cdot s \cdot m^{-3}} = 2\,126\ \mathrm{Bq \cdot m^{-3}}$$

with the values for the individual terms from the auxiliary Equations (18) to (26)

$$T_1 = (2\,126\ \mathrm{Bq \cdot m^{-3}})^2 \cdot 0{,}05^2 = 11\,300\ \mathrm{Bq^2 \cdot m^{-6}}$$

$$T_2 = 2\,126\ \mathrm{Bq \cdot m^{-3}} \cdot \frac{333{,}33\ \mathrm{Bq \cdot s \cdot m^{-3}}}{3\,600\ \mathrm{s}} = 196{,}8\ \mathrm{Bq^2 \cdot m^{-6}}$$

$$T_3 = (333{,}33\ \mathrm{Bq \cdot s \cdot m^{-3}})^2 = 111\,109\ \mathrm{Bq^2 \cdot s^2 \cdot m^{-6}}$$

$$T_4 = \frac{1}{3\,600\ \mathrm{s}} \cdot \left(\frac{7\,992}{3\,600\ \mathrm{s}} \cdot 14 + 8{,}89\ \mathrm{s}^{-1} \cdot (-2{,}417) \cdot 1{,}0665\right) = 2{,}268 \cdot 10^{-3}\ \mathrm{s}^{-2}$$

$$T_5 = 14^2 \cdot \frac{7\,992}{(3\,600\ \mathrm{s})^2} = 0{,}1209\ \mathrm{s}^{-2}$$

$$T_6 = (-2{,}417)^2 \cdot 1{,}0665^2 \cdot \frac{8{,}89\ \mathrm{s}^{-1}}{3\ 600\ \mathrm{s}} = 0{,}01641\ \mathrm{s}^{-2}$$

$$T_7 = \left(\frac{8{,}89\ \mathrm{s}^{-1} \cdot 1{,}0665}{1 + 3{,}39} - \frac{7\ 992}{3\ 600\ \mathrm{s}}\right)^2 \cdot (0{,}01 \cdot 14)^2 = 71{,}21 \cdot 10^{-6}\ \mathrm{s}^{-2}$$

$$T_8 = \frac{(8{,}89\ \mathrm{s}^{-1})^2 \cdot 1{,}0665^2 \cdot (1 + 14)^2}{(1 + 3{,}39)^4} \cdot (0{,}01 \cdot 3{,}39)^2 = 0{,}06258\ \mathrm{s}^{-2}$$

$$T_9 = (8{,}89\ \mathrm{s}^{-1})^2 \cdot (-2{,}417)^2 \cdot 0{,}001925^2 = 1{,}711 \cdot 10^{-3}\ \mathrm{s}^{-2}$$

the standard uncertainty $u(c_{\mathrm{G\gamma}})$ of the gross gamma activity concentration is calculated according to Equation (17):

$$u\left(c_{\mathrm{G\gamma}}\right) = \sqrt{11\ 300 + 196{,}8 + \ 111\ 109 \cdot 0{,}2039}\ \ \mathrm{Bq} \cdot \mathrm{m}^{-3} = 184{,}8\ \ \mathrm{Bq} \cdot \mathrm{m}^{-3}$$

Thus, the gross gamma activity concentration is:

$$c_{\mathrm{G\gamma}} = (2\ 126 \pm 185)\ \mathrm{Bq} \cdot \mathrm{m}^{-3}$$

The worked example applies in an analogous manner to the calculation of the artificial gross gamma activity concentration with the measuring arrangement "probe in the vessel's strom box". For this, the corresponding procedural, vessel-specific calibration factor must be used.

5.3 Consideration of the uncertainties

The standard uncertainty of the analysis result includes the standard uncertainties of the statistical counting and the calibration. A possible contribution of artificial gamma radiation in energy range B is not taken into account in the uncertainty analysis in the context of this screening method. The standard uncertainty of the duration of measurement is neglected.

6 Characteristic limits of the procedure

For this screening method, the analytical derivation of the characteristic limits according to the standard ISO 11929 series is omitted [7].

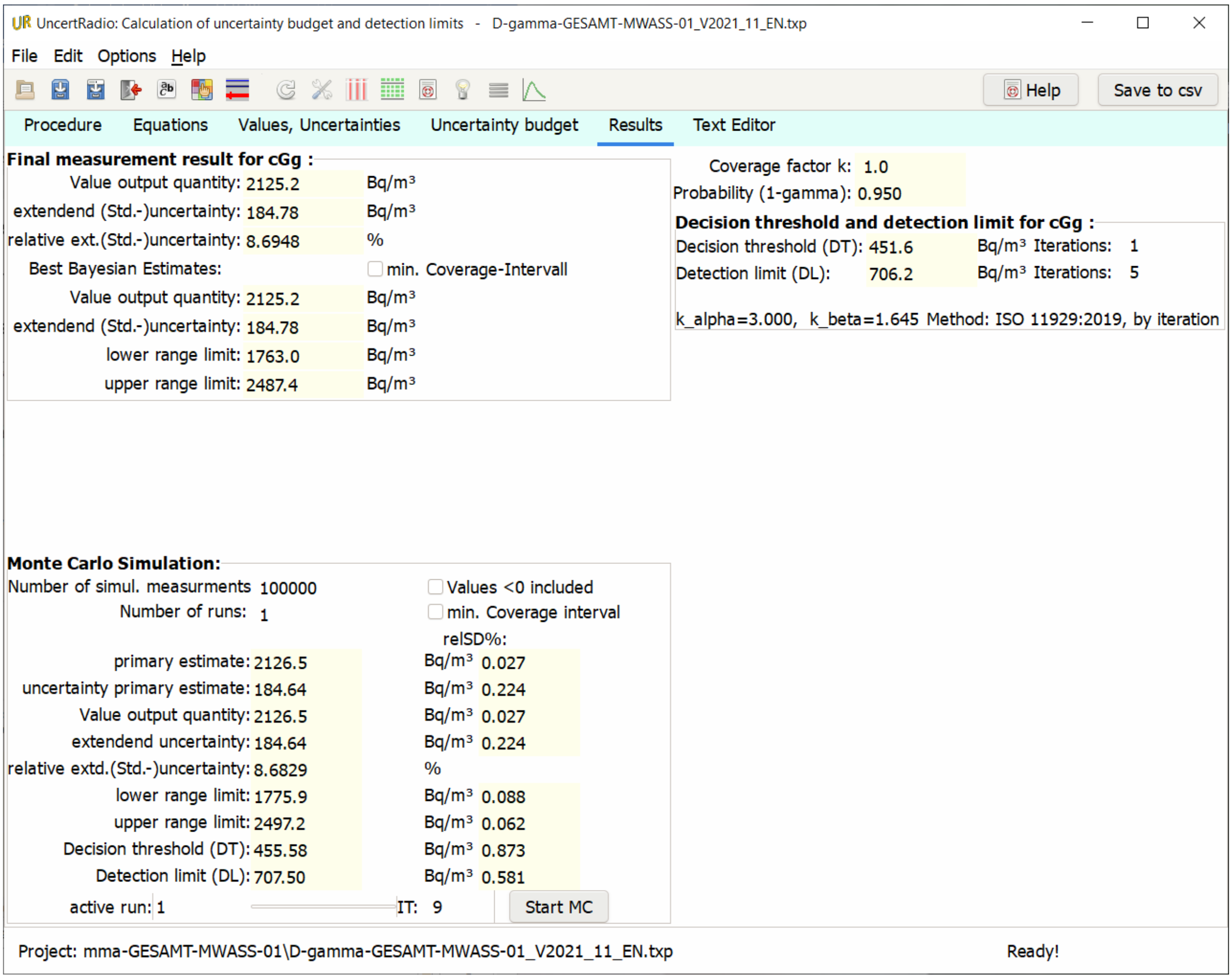
However, if these are of interest, they can be calculated using a project file for the software UncertRadio (see Section 7.2).

7 Software supported calculation

7.1 View of the Excel spreadsheet

Due to the complexity of the calculation, no Excel spreadsheet is available.

7.2 View of the UncertRadio result page



The corresponding UncertRadio project file is available on the website of this Procedures Manual.

8 Catalogue of the chemicals und equipment

8.1 Chemicals

The potassium chloride, KCl, used for this procedure should be of analytically pure quality.

8.2 Equipment

The following equipment is used for the procedure:

— pressure sensors for water level measurement;

— measuring device, for example:

- a 3 "x 3" NaI (Tl) detector with a resolution of 7 % for Cs;
- a multi-channel analyzer with 2 048 channels;

— calibration container measuring 3 m x 3 m x 2,7 m with a volume of around 25 m^3;

— calibration container with a volume of 1 m^3;

References

[1] Wedekind, C., Schilling, G., Grüttmüller, M., Becker, K.: *Neues Messverfahren zur Überwachung der Radioaktivität des Meeres im Bundes-Messnetz des BSH*. In: Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (ed.): 10. Fachgespräch zur Überwachung der Umweltradioaktivität, Hamburg, 28. - 30. April 1998, S. 41-45.

[2] Wedekind, C., Schilling, G., Grüttmüller, M., Becker, K.: *Gamma-radiation monitoring network at sea*. Applied Radiation and Isotopes, 1999, Vol. 50 (4), S. 733-741.

[3] Wedekind, C., Becker, K., Schilling, G., Grüttmüller, M.: *Marine environmental radioactivity monitoring by ''in-situ'' γ-radiation detection*. Kerntechnik, 2000, Vol. 65 (4), S. 190-194.

[4] *Allgemeine Verwaltungsvorschrift zum integrierten Mess- und Informationssystem zur Überwachung der Radioaktivität in der Umwelt (IMIS) nach dem Strahlenschutz-vorsorgegesetz (AVV-IMIS)*. Bundesanzeiger, 2006, Nr. 244a, S. 4-80.

[5] Wedekind, C.: *Gamma-ray spectrometer probe for the measurement of radioactive pollution in the sea*. Health Physics, 1973, Vol. 25 (1), S. 51-58.

[6] Laboratoire National Henri Becquerel (LNHB): *Nucléide - Lara: Library for gamma and alpha emissions*. In: Atomic and Nuclear data. Last Update 21.09.2021. Available at: http://www.lnhb.fr/nuclear-data/module-lara/. [Last access 12.11.2021].

[7] Standard series ISO 11929:2019, *Determination of the characteristic limits (decision threshold, detection limit and limits of the coverage interval) for measurements of ionizing radiation — Fundamentals and application (Parts 1 to 3)*.

Annex A
Procedure for carrying out a calibration of probes in different geometries for the screening method

A.1 General

The initial calibration of the probe is required for two measuring arrangements that occur in practical application. The most common measurement setup is where the probe hangs freely in the seawater. However, there is also the possibility of operating the probe in a tube-like or shaft-like device in the vessel's hull, a so-called "strom box".

The initial calibrations carried out in the federal coordinating office for the “probe hanging freely in the water” and “probe in the vessel's strom box” are described below.

A.2 Measuring arrangement "probe hanging freely in the water"

A.2.1 Calibration with the single-line nuclide potassium-40

The calibration is carried out in a sufficiently large container that contains the calibration solution. The radionuclide of choice used for calibration is potassium-40 (K-40) because, in addition to its property as a single-line nuclide, it also fulfills the following criteria:

— high solubility in water;

— unproblematic procurement in larger quantities;

— manageability outside radiation protection regulations.

A potassium chloride solution is used as the calibration solution, since potassium chloride naturally contains K-40 with a specific activity of 16 566 $Bq \cdot kg^{-1}$. The gamma radiation energy of K-40 is 1,461 keV with a gamma emission intensity of 0,1055. The background measurement is carried out in tap water.

In the case of this calibration with K-40, a distance between the probe and the container wall of at least 1 m must be maintained due to the high gamma radiation energy. In this container with a capacity of e. g. 25 m^3, the probe with a 3" x 3" NaI (Tl) detector is attached centrally and freely hanging in the water.

The procedural gross efficiency of the gamma radiation energy of K-40 is determined analogously to Equation (1) according to Equation (A1):

$$\varepsilon_{\mathrm{t}}(E_\gamma) = \frac{R_{\mathrm{n,t}}}{c_{\mathrm{K-40}} \cdot p_{\gamma,\mathrm{K-40}}(E_\gamma)} \qquad \text{(A1)}$$

Herein are:

$\varepsilon_{\mathrm{t}}(E_\gamma)$ procedural, gross efficiency of the gamma radiation energy of K-40 with a freely hanging probe, in $m^3 \cdot Bq^{-1} \cdot s^{-1}$;

R_{g} gross count rate in the energy range from 0,3 MeV to 1,5 MeV, in s^{-1};

R_0 background count rate in the energy range from 0,3 MeV to 1,5 MeV, in s^{-1};

$c_{\text{K-40}}$ activity concentration of the radionuclide K-40, in $Bq \cdot m^{-3}$;

$p_{\gamma,\text{K-40}}(E_\gamma)$ emission intensity of the gamma line of the radionuclide r.

With a measured value of 59 s^{-1} for the gross count rate of K-40 of a potassium chloride solution, which has a K-40 activity concentration of 115 963 $Bq \cdot m^{-3}$ (corresponds to 7 g KCl), and a background count rate of 25 s^{-1} measured in tap water, the following value is obtained for the procedural gross efficiency of the measurement arrangement "probe hanging freely in the water" according to Equation (A1):

$$\varepsilon_{\mathrm{t}}(E_\gamma) = \frac{59\ \mathrm{s}^{-1} - 25\ \mathrm{s}^{-1}}{115\ 963\ \mathrm{Bq} \cdot \mathrm{m}^{-3} \cdot 0{,}1055} = 2{,}779 \cdot 10^{-3}\ \ \mathrm{m}^3 \cdot \mathrm{Bq}^{-1} \cdot \mathrm{s}^{-1}$$

A.2.2 Calibration with other radionuclides

Investigations with solid preparations of artificial, gamma-emitting radionuclides with gamma energies below 1 MeV have shown that a calibration container with a volume of 1 m^3 is sufficient because the shielding effect of the water means that gamma radiation can no longer be detected above a preparation distance of 0,5 m from the detector.

The gamma-emitting radionuclides Mn-54, Ru-106/Rh-106 and Ce-144 with the values listed in Table 1 are used for the calibration. A fictitious mean gamma energy $\bar{E}_\gamma$ is assumed for the multiline nuclide Ru-106/Rh-106, which is calculated according to Equation (A2):

$$\bar{E}_\gamma = \frac{\sum_1^n E_{\gamma,i} \cdot p_{\gamma,i}}{\sum_1^n p_{\gamma,i}} \tag{A2}$$

Herein are:

$\bar{E}_\gamma$ fictitious mean gamma energy, in eV;

$E_{\gamma,i}$ gamma energy of the gamma line i, in eV;

$p_{\gamma,i}$ emission intensity of the gamma line i.

For Ru-106/Rh-106, it follows from Equation (A2):

$$\bar{E}_{\gamma,\mathrm{Ru-106}} = \frac{0{,}51\ \mathrm{MeV} \cdot 0{,}2052 + 0{,}62\ \mathrm{MeV} \cdot 0{,}0987 + 1{,}05\ \mathrm{MeV} \cdot 0{,}0149}{0{,}2052 + 0{,}0987 + 0{,}0149} = 0{,}57\ \mathrm{MeV}$$

A.3 Measuring arrangement “probe in the vessel’s strom box”

The calibration of probes in the vessel's strom box cannot be carried out in the laboratory. For this reason, the calibration was carried out at different salt contents in the water with the existing measuring device on one of the BSH vessels.

The activity concentration in the sea is determined by the potassium in the sea salt [4]. The potassium content in the seawater of the open North Sea corresponds to a K-40 activity concentration $c_{\text{K-40}}$ of 12 Bq·l^{-1}. The gross count rate R_{g} determined in the North Sea with a salt content of 35 PSU (Practical Salinity Unit, corresponds to 0,74 g·l^{-1}) is 10 s^{-1}.

The background measurement was carried out in the fresh water of the Elbe river because the potassium content there is negligible; the measured background count rate R_0 was 8,5 s^{-1}.

The procedural gross efficiency $\varepsilon_{\text{t}}(E_\gamma)$ for the measuring arrangement "probe in the vessel's strom box", in m^3·Bq^{-1}·s^{-1}, is calculated according to Equation (A3):

$$\varepsilon_{\text{t}}(E_\gamma) = \frac{R_{\text{g}} - R_0}{c_{\text{K}-40} \cdot p_{\gamma,\text{K}-40}(E_\gamma)} \tag{A3}$$

The following value for the procedural gross efficiency is thus obtained for this measuring arrangement:

$$\varepsilon_{\text{t}}(E_\gamma) = \frac{10\ \text{s}^{-1} - 8{,}5\ \text{s}^{-1}}{12\,000\ \text{Bq} \cdot \text{m}^{-3} \cdot 0{,}1055} = 1{,}185 \cdot 10^{-3}\ \ \text{m}^3 \cdot \text{Bq}^{-1}\ \cdot \text{s}^{-1}$$