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Measurement of radioactivity in the environment — Soil —

Part 3: Measurement of gamma-emitting radionuclides

Mesurage de la radioactivité dans l'environnement — Sol —

Partie 3: Mesurages des radionucléides émetteurs gamma

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ISO copyright office
Case postale 56 • CH-1211 Geneva 20
Tel. + 41 22 749 01 11
Fax + 41 22 749 09 47
E-mail copyright@iso.org
Web www.iso.org

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Foreword

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International Standards are drafted in accordance with the rules given in the ISO/IEC Directives, Part 2.

The main task of technical committees is to prepare International Standards. Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

Attention is drawn to the possibility that some of the elements of this document may be the subject of patent rights. ISO shall not be held responsible for identifying any or all such patent rights.

ISO 18589-3 was prepared by Technical Committee ISO/TC 85, *Nuclear energy*, Subcommittee SC 2, *Radiation protection*.

ISO 18589 consists of the following parts, under the general title *Measurement of radioactivity in the environment — Soil*:

— *Part 1: General guidelines and definitions*

— *Part 2: Guidance for the selection of the sampling strategy, sampling and pre-treatment of samples*

— *Part 3: Measurement of gamma-emitting radionuclides*

— *Part 4: Measurement of plutonium isotopes (plutonium 238 and plutonium 239 + 240) by alpha spectrometry*

— *Part 5: Measurement of strontium 90*

— *Part 6: Measurement of gross alpha and gross beta activities*

Introduction

This International Standard is published in several parts to be used jointly or separately according to needs. Parts 1 to 6, concerning the measurements of radioactivity in the soil, have been prepared simultaneously. These parts are complementary and are addressed to those responsible for determining the radioactivity present in soils. The first two parts are general in nature. Parts 3 to 5 deal with radionuclide-specific measurements and Part 6 with non-specific measurements of gross alpha or gross beta activities.

Additional parts may be added to ISO 18589 in the future if the standardization of the measurement of other radionuclides becomes necessary.

Measurement of radioactivity in the environment — Soil —

Part 3:
Measurement of gamma-emitting radionuclides

1 Scope

This part of ISO 18589 specifies the identification and the measurement of the activity in soils of a large number of gamma-emitting radionuclides using gamma spectrometry. This non-destructive method, applicable to large-volume samples (up to about 3 000 cm^3), covers the determination in a single measurement of all the γ-emitters present for which the photon energy is between 5 keV and 3 MeV.

This part of ISO 18589 can be applied by test laboratories performing routine radioactivity measurements as a majority of radionuclides is characterized by gamma-ray emission between 40 keV and 2 MeV.

This part of ISO 18589 is suitable for the surveillance of the environment and the inspection of a site and allows, in case of accidents, a quick evaluation of gamma activity.

2 Normative references

The following referenced documents are indispensable for the application of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 31-9, *Quantities and units — Part 9: Atomic and nuclear physics*

ISO 10703, *Water quality — Determination of the activity concentration of radionuclides — Method by high resolution gamma-ray spectrometry*

ISO 11074, *Soil quality — Vocabulary*

ISO/IEC 17025, *General requirements for the competence of testing and calibration laboratories*

ISO 18589-1, *Measurement of radioactivity in the environment — Soil — Part 1: General guidelines and definitions*

ISO 18589-2, *Measurement of radioactivity in the environment — Soil — Part 2: Guidance for the selection of the sampling strategy, sampling and pre-treatment of samples*

Guide to the expression of uncertainty in measurement (GUM), BIPM/IEC/IFCC/ISO/IUPAC/IUPAP/OIML

3 Terms, definitions and symbols

For the purposes of this document, the terms, definitions and symbols given in ISO 18589-1, ISO 11074, ISO 31-9 and ISO 10703 and the following symbols apply.

m	Mass of the test portion, in kilograms
A	Activity of each radionuclide in the calibration source, at the calibration time, in becquerel
a, a_c	Activity, in becquerel per kilogram, per unit of mass of each radionuclide, without and with corrections
t_g	Sample spectrum counting time, in seconds
t_0	Ambient background spectrum counting time, in seconds
t_s	Calibration spectrum counting time, in seconds
$n_{N,E}$, $n_{N0,E}$, $n_{Ns,E}$	Number of counts in the net area of the peak, at energy, E, in the sample spectrum, in the background spectrum and in the calibration spectrum, respectively
$n_{g,E}$, $n_{g0,E}$, $n_{gs,E}$	Number of counts in the gross area of the peak, at energy, E, in the sample spectrum, in the background spectrum and in the calibration spectrum, respectively
$n_{b,E}$, $n_{b0,E}$, $n_{bs,E}$	Number of counts in the background of the peak, at energy, E, in the sample spectrum, in the background spectrum and in the calibration spectrum, respectively
ε_E	Efficiency of the detector at energy, E, with the actual measurement geometry
P_E	Probability of the emission of gamma radiation with energy, E, for each radionuclide, per decay
$\mu_1(E)$, $\mu_2(E)$	Linear attenuation coefficient at photon energy, E, of the sample and calibration source, respectively, per centimetre
$\mu_{m,i}(E)$	Mass attenuation coefficient, in square centimetres per gram, at photon energy, E, of element i
h	Height of the sample in the container, in centimetres
w_i	Mass fraction of element i (no unit)
ρ	Bulk density, in grams per cubic centimetre, of the sample
λ	Decay constant of each radionuclide, per second
$u(a)$, $u(a_c)$	Standard uncertainty, in becquerel per kilogram, associated with the measurement result, with and without corrections, respectively
U	Expanded uncertainty, in becquerel per kilogram, calculated by $U = k \cdot u(a)$ with $k = 1, 2, \ldots$
a^*, a_c^*	Decision threshold, in becquerel per kilogram, for each radionuclide, without and with corrections, respectively
$a^\#$, $a_c^\#$	Detection limit, in becquerel per kilogram, for each radionuclide, without and with corrections, respectively
$a^\triangleleft$, $a^\triangleright$	Lower and upper limits of the confidence interval, for each radionuclide, in becquerel per kilogram

4 Principle

The activity of gamma-emitting radionuclides present in the soil samples is determined using gamma spectrometry techniques based on the analysis of the energies and the peak areas of the full-energy peaks of the gamma lines. These techniques allow the identification and the quantification of the radionuclides [1], [2].

The nature and geometry of the detectors as well as the samples call for appropriate energy and efficiency calibrations [3], [4], [5]. If well-type detectors are used to measure small-mass samples, it is necessary to take special care to consider coincidence and summation effects (see 8.1.4).

NOTE Sodium iodide detectors can be used for the measurement of radioactivity in soil only in certain cases. Therefore, this part of ISO 18589 deals exclusively with gamma spectrometry using semiconductor detectors.

5 Gamma-spectrometry equipment

Gamma-spectrometry equipment generally consists of

— a semiconductor detector with a cooling system (liquid nitrogen, cryogenic assembly, etc.),

— a shield, consisting of lead and/or other materials, against ambient radiation,

— appropriate electronics (high-voltage power supply; signal-amplification system; an analogue-to-digital converter),

— a multi-channel amplitude analyser,

— a personal computer to display the measurement spectra and to process the data.

The semiconductor detectors generally used are made of high-purity germanium crystals (HP Ge). The type and geometry of these detectors determine their field of application. For example, when detecting photons with an energy below 400 keV, the use of detectors with a thin crystal is recommended in order to limit interference from high-energy photons. However, it is better to use a large-volume, P-type coaxial detector to measure high-energy photons (above 200 keV) or an N-type coaxial detector to detect both low- and high-energy radiation.

At the level of natural radioactivity, it is advantageous for the measurement to use an ultra-low-level measuring instrument, i.e. a set-up arranged with a choice of materials for the detector and shielding that guarantees a very low background level. This includes very low-noise electronic preamplifiers and amplifiers. The shielding case should be large enough to allow sufficient distance from all walls and the detector set up in the centre of the case, when 1-l samples are inserted. This allows the use of a room with a very low specific activity of building materials and a very low radon concentration in the room air to be chosen. It is optimal to erect the measuring instruments in the middle of the room with the maximum distance available to the room walls. Forced ventilation of the measuring room can possibly contribute to stabilizing the background level. On the other hand, forced ventilation can then cause problems when the outside air drawn in contains excess radon as a result of a warming-up of the soil (in particular, when the soil thaws in spring). It is always good practice to fill the inner part of the shielding with nitrogen. For this, the gaseous nitrogen escaping from the Dewar vessel of the detector arrangement can be passed permanently into the shielding.

The main characteristics that allow the estimation of a detector performance are as follows:

a) energy resolution (total width at half maximum of the full-energy peak), which enables the detector to separate two neighbouring gamma peaks;

b) absolute efficiency, which specifies the percentage of photons detected in the full-energy peak relative to the number of photons emitted;

c) peak-to-Compton ratio.

Depending on the required accuracy and the desired detection limit, it is generally necessary to use high-quality detectors whose energy resolution is less than 2,2 keV (for the ^{60}Co peak at 1,332 keV) and with a peak/Compton ratio between 50 and 80 for ^{137}Cs.

Some natural radionuclides, e.g. ^{210}Pb and ^{238}U via ^{234}Th, can be measured only via gamma lines in the energy range of 100 keV. In this case, the use of an N-type detector is recommended. Low-energy, low-level detectors offered by manufacturers have been optimized for this purpose and can additionally be used in other areas of environmental monitoring, e. g. for measurements of ^{129}I and ^{241}Am in samples from the vicinity of nuclear facilities.

The computer, in combination with the available hardware and software, shall be carefully selected [6], [7]. It is recommended that the results of the computer analysis of the spectrum be visually checked regularly.

Comparison with a certified reference material is recommended to check the performance of the apparatus. Participation in proficiency and inter-laboratory tests and inter-comparison exercises can also help to verify the performance of the apparatus and the status of the analysis [10], [11].

6 Sample container

Measuring gamma radioactivity in soils requires sample containers that are suited to gamma spectrometry. These containers should have the following characteristics:

— be made of materials with low absorption of gamma radiation;

— have volumes adapted to the shape of the detector for maximum efficiency;

— be watertight and not react with the sample constituents;

— have a wide-necked, airtight opening to facilitate filling;

— be unbreakable.

In order to verify easily that the content of the container conforms to the standard counting geometry, a transparent container with a mark to check the filling can be selected.

7 Procedure

7.1 Packaging of samples for measuring purposes

The soil samples packaged for gamma spectrometry measurements are usually dried, crushed, and homogenized in accordance with ISO 18589-2.

The procedure shall be carried out as follows.

a) Choose the container that is best suited to the volume of the sample so as to measure as much material as possible. To decrease self-absorption effects, the height of the contents should be minimized.

b) Fill the container to the level of the volume mark. It is recommended to use a mechanical filling device (for example, a vibrating table) to pack the sample to avoid any future losses in volume.

c) Note the sample mass. This information is useful when using the measurements to express the result as specific activity and when carrying out self-absorption corrections.

d) Visually check the upper level of the sample and make sure that it is horizontal before measuring. Where applicable, add more material to the sample until the mark has been reached and adjust the noted sample mass accordingly.

e) Hermetically seal the container if volatile or natural radionuclides are being measured.

f) Clean the outside of the container to remove potential contamination due to the filling process.

If measurements are required quickly, the processing method described in ISO 18589-2 can be ignored. This shall be mentioned in the test report and the results cannot be expressed in becquerels per kilogram of dry soil.

When measuring Rn-222 via decay products of Ra-226, the sealed container shall be stored long enough to allow radioactive equilibrium to be reached.

7.2 Laboratory background level

As some radionuclides found in the soil (see Annex B) are the same as in building materials, the detector and sample shall be adequately shielded against natural background radiation. Frequently, it is sufficient to shield the detector in a 10 cm thick, low-background lead case wall. Reduction of radon inside the shield is desirable. Further information is given in references [1], [2].

The natural radionuclides and their decay products occur widely and with large concentration ranges in floors, walls, ceilings, the air of the measuring rooms and in the materials of which detectors and shielding are made.

There are isotopes of the decay chain of the rare gas radon, whose emanation from the materials surrounding the measuring instruments depends on various physical parameters. Thus, large fluctuations in the concentration of radon and of the decay products can occur in room air and in the air of the detector shielding. This is a particular problem in basements of old buildings with defective floors.

The background of the measuring instruments shall be kept as low as possible and, in particular, as stable as possible by appropriate measures. This includes vacuuming the shielding and removing the dust by filtration. Frequent measurements of the background level permit the verification of its stability. This is necessary because the peaks of the background spectrum shall be subtracted from those of a sample spectrum.

7.3 Calibration

7.3.1 Energy calibration

Energy calibration is carried out using sources of a radionuclide with different emission lines (for example ^{152}Eu) or sources containing a mixture of several radionuclides. This calibration allows the establishment of the relationship between the channel numbers of the analyser and the known energy of the photons [12], [13], [14]. Generally, this task is carried out with appropriate software, which uses the standard spectra to automatically convert the channel scale of the multi-channel analyzer into a photon energy scale and to record the useful information necessary for future analyses. By using the energy calibration spectra, the full-width at half the maximum of the full-energy peaks can be determined as a function of the gamma energy. This information is usually required by the spectrometry analysis software.

Further information is given in ISO 10703 and References [8] and [9].

7.3.2 Efficiency calibration

Efficiency calibration is carried out either via *ab initio* calculations of the detector efficiency using transport theory and Monte Carlo techniques (not covered in this part of ISO 18589) or by using a radionuclide source having different emission lines or a mixed-radionuclide source. This calibration allows the establishment of the detection efficiency of the detector as a function of the energy of the radiation.

When using a radionuclide source with different emission lines for calibration, summation effects or coincidence losses should be taken into account.

The sample measurement shall be performed with the same measuring conditions as used for calibrating the gamma spectrometry system. In particular, the settings of the electronics (gain and high voltage), the measurement geometry, the position of the source in relation to the detector and the sample and standard matrices shall be identical.

For this purpose, a calibration source should have the same physical and chemical properties as the sample. It may, for instance, be produced by spiking an appropriate sample of soil.

With these conditions, the efficiency at energy E shall be calculated as given in Equation (1):

$$\varepsilon_E = \frac{n_{\mathrm{Ns},E} / t_\mathrm{s}}{A \cdot P_E} \quad (1)$$

For an undisturbed peak at an energy E, the count, $n_{\mathrm{Ns},E}$, in the net-peak area of a γ-spectrum is calculated as given in Equation (2):

$$n_{\mathrm{Ns},E} = n_{\mathrm{gs},E} - n_{\mathrm{bs},E} \quad (2)$$

When the physical and chemical nature of the sample (chemical composition, bulk density) is different from the conditions of the efficiency calibration, a correction for the self-absorption of gamma radiation should be applied.

Further information is given in ISO 10703 and References [8] and [9].

7.4 Measurements of and corrections for natural radionuclides

If activities of natural radionuclides in the soil are being measured, the areas of full-energy peaks used for evaluating their activities shall be corrected for the background contribution of those same radionuclides inside the detector shielding, taking into account potential differences of the duration of the sample and background measurements.

Special advice to take into account during the measurement of natural radionuclides in soil and information on spectroscopic interferences is given in Annex B.

The gamma ray of the radionuclides in the background and/or of natural radionuclides inside the sample can also interfere with measurements of artificial radionuclides and can require appropriate corrections.

8 Expression of results

8.1 Calculation of the activity per unit of mass

8.1.1 General

The activity per unit of mass, a, of each radionuclide present in the sample is obtained from the net count, $n_{\mathrm{N},E}$, from the peak of an individual γ-line without interference using Equation (3):

$$a = \frac{n_{\mathrm{N},E} / t_\mathrm{g}}{P_E \cdot \varepsilon_E \cdot m \cdot f_E} \quad (3)$$

where

f_E is the correction factor considering all necessary corrections according to Equation (4):

$$f_E = f_\mathrm{d} \cdot f_{\mathrm{att},E} \cdot f_{\mathrm{cl},E} \cdot f_{\mathrm{s},E} \quad (4)$$

where

f_d is the factor to correct for decay for a reference date;

$f_{att,E}$ is the factor to correct for self-absorption;

$f_{cl,E}$ is the factor to correct for coincidence losses;

$f_{s,E}$ is the factor to correct summing-up effects by coincidences.

For an undisturbed peak with energy, E, the count, $n_{N,E}$, in the net-peak area of a γ-spectrum is calculated by Equation (5):

$$n_{N,E} = n_{g,E} - n_{b,E} \tag{5}$$

Thus, Equation (3) can be expressed as given in Equation (6):

$$a = \frac{n_{N,E} / t_g}{P_E \cdot \varepsilon_E \cdot m \cdot f_E} = \frac{n_{g,E} - n_{b,E}}{P_E \cdot \varepsilon_E \cdot m \cdot f_E \cdot t_g} = (n_{g,E} - n_{b,E}) \cdot w / t_g \text{ with } w = \frac{1}{P_E \cdot \varepsilon_E \cdot m \cdot f_E} \tag{6}$$

8.1.2 Decay corrections

Depending on the half-life of the radionuclide being measured, the activity per unit of mass shall be corrected by f_d. To take into account the radioactive decay during the counting time and during the time between the reference instant ($t = 0$) and the measuring instant ($t = t_i$), f_d shall be calculated by Equation (7):

$$f_d^{-1} = e^{\lambda \cdot t_i} \cdot \left[\frac{\lambda \cdot t_g}{1 - e^{-\lambda \cdot t_g}} \right] \tag{7}$$

8.1.3 Self-absorption correction

Measurement of radioactivity in soils by gamma spectrometry can involve a calibrated source whose matrix is different from that of the sample being measured. In this case, a correction factor should be applied to the result obtained. The lower the radiation energy, the larger the correction factor.

Different techniques may be used to determine this correction factor:

— measurement of the attenuation coefficient of gamma radiation in the sample material at a given energy;

— mathematical calculation that takes into account the chemical composition and bulk density of the sample.

For cylindrical sample containers at the level of the detector, the value of the attenuation correction factor, $f_{att,E}$, may be estimated using Equation (8):

$$f_{att,E} = \frac{\mu_2(E) \cdot \left(1 - e^{-\mu_1(E) \cdot X}\right)}{\mu_1(E) \cdot \left(1 - e^{-\mu_2(E) \cdot X}\right)} \tag{8}$$

where X is the average path length, expressed in metres, of the gamma photons in the container.

The linear attenuation coefficient, $\mu(E)$, depends on the photon energy, bulk density, chemical composition of the sample and expresses the exponential decrease of the flux density of gamma rays with distance. It may be calculated using Equation (9):

$$\mu(E) = \left[\sum_i w_i \mu_{\mathrm{m},i}(E) \right] \rho \tag{9}$$

As an approximation and for soils of the same nature, the linear attenuation coefficient, $\mu(E)$, can be obtained directly by multiplying the mass attenuation coefficient by the density.

8.1.4 Summation effects or coincidence losses corrections

For radionuclides with cascade transitions, counting losses due to coincidence summing are to be expected, especially at high counting efficiencies.

These corrections are important for point as well as thin source samples measured very close to the detector surface; they are specific for each radionuclide, detector, measuring geometry and sample-to-detector distance.

Most of the theoretical methods for such calculations are related to the use of transport theory and Monte-Carlo techniques (Geant, EGSnrc, MCNP, Penelope, etc.; see References [18], [19], [20], [21]); given the difficulties associated with modelling detectors, some experimental procedures can be applied for each specific situation.

Some of these experimental procedures use data from specialized literature, but given the wide range of detector possibilities and measuring conditions, direct measurement as given in a) to c) below can be made.

a) Prepare a source containing the multi-line photon-emitting radionuclide whose correction factor at energy, E, shall be calculated along with another radionuclide emitting at a similar energy, E', which is mono-energetic or has negligible summing corrections. The geometry shall be the same as that used for the sample source.

b) Make a measurement with this source at a large distance from the detector. Calculate the relationship between the net peak counts at energies E and E'.

c) Make a measurement with the sample in the normal measuring position. The relationship between the net peak counts at energies E and E' is similar to that calculated above and the theoretical net peak counts, $n_{\mathrm{N},E}^{\mathrm{T}}$, at energy E can be estimated.

The relationship between the theoretical net peak counts, $n_{\mathrm{N},E}^{\mathrm{T}}$, and the measured net peak counts, $n_{\mathrm{N},E}$, is the summing correction factor for energy E of the multi-line photon emitting radionuclide that shall be applied to the analysis of the calibration and source sample spectrum.

Further information is given in References [2] and [9].

8.2 Standard uncertainty

According to *GUM*, the standard uncertainty of a is calculated by Equation (10):

$$u(a) = \sqrt{(w/t_{\mathrm{g}})^2 \cdot \left[u^2(n_{\mathrm{g},E}) + u^2(n_{\mathrm{b},E}) \right] + a^2 \cdot u_{\mathrm{rel}}^2(w)} \tag{10}$$

where the uncertainty of the counting time is neglected.

The relative standard uncertainty of w is calculated by Equation (11):

$$u_{\mathrm{rel}}^2(w) = u_{\mathrm{rel}}^2(P_E) + u_{\mathrm{rel}}^2(m) + u_{\mathrm{rel}}^2(\varepsilon_E) + u_{\mathrm{rel}}^2(f_E) \tag{11}$$

Taking Equation (1) into account, the relative standard uncertainty of ε_E is calculated by Equation (12):

$$u_{\mathrm{rel}}^2(\varepsilon_E) = u_{\mathrm{rel}}^2(n_{\mathrm{Ns},E}) + u_{\mathrm{rel}}^2(A) + u_{\mathrm{rel}}^2(P_E) = u_{\mathrm{rel}}^2(n_{\mathrm{gs},E} - n_{\mathrm{bs},E}) + u_{\mathrm{rel}}^2(A) + u_{\mathrm{rel}}^2(P_E) \tag{12}$$

where $u_{\mathrm{rel}}(A)$ includes all the uncertainties related to the calibration source: standard certificate, preparation of the calibration source.

For the calculation of the characteristic limits (see ISO 11929), it is necessary to know $\tilde{u}(\tilde{a})$, i.e. the standard uncertainty of a as a function of its true value. For a true value $\tilde{a}$, from $n_{\mathrm{g},E} = \tilde{a} \cdot t_{\mathrm{g}} / w + n_{\mathrm{b},E}$ and with $u^2(n_{\mathrm{g}}) = n_{\mathrm{g}}$, one obtains Equation (13):

$$\tilde{u}(\tilde{a}) = \sqrt{(w/t_{\mathrm{g}})^2 \cdot \left[(t_{\mathrm{g}}/w) \cdot \tilde{a} + n_{\mathrm{b},E} + u^2(n_{\mathrm{b},E})\right] + \tilde{a}^2 \cdot u_{\mathrm{rel}}^2(w)} \tag{13}$$

The uncertainties $u(n_{\mathrm{N}})$, $u(n_{\mathrm{g}})$, and $u(n_{\mathrm{b}})$ shall be calculated in accordance with *GUM*, taking into account that the individual counts, n_i, in channel i of a multi-channel spectrum are the result of a Poisson process and hence $u^2(n_i) = n_i$ holds. The values of n_{N}, n_{g}, and n_{b} and their associated standard uncertainties $u(n_{\mathrm{N}})$, $u(n_{\mathrm{g}})$, and $u(n_{\mathrm{b}})$ may be calculated with a computer program. Since there are various methods of subtracting the background below a peak in order to derive the number of counts in the net peak area, no generally applicable equation can be given. An example of the simple case of linear background subtraction is given in Annex A.

8.3 Decision threshold

The decision threshold, a^*, is obtained from Equation (13) for $\tilde{a} = 0$ (see ISO 11929). This yields Equation (14):

$$a^* = k_{1-\alpha} \cdot \tilde{u}(0) = k_{1-\alpha} \cdot (w/t_{\mathrm{g}}) \sqrt{n_{\mathrm{b},E} + u^2(n_{\mathrm{b},E})} \tag{14}$$

$\alpha = 0{,}05$ and $k_{1-\alpha} = 1{,}65$ are often chosen by default.

8.4 Detection limit

The detection limit, $a^{\#}$, is calculated by Equation (15) (see ISO 11929):

$$\begin{aligned} a^{\#} &= a^* + k_{1-\beta} \cdot \tilde{u}(a^*) \\ &= a^* + k_{1-\beta} \cdot \sqrt{w^2 \left[\left(a^{\#}/w + n_{\mathrm{b},E}/t_{\mathrm{g}}\right)/t_{\mathrm{g}} + u^2(n_{\mathrm{b},E})/t_{\mathrm{g}}^2 \right] + a^{\#2} \cdot u_{\mathrm{rel}}^2(w)} \end{aligned} \tag{15}$$

$\beta = 0{,}05$ and $k_{1-\beta} = 1{,}65$ are often chosen by default.

The detection limit can be calculated by solving Equation (15) for $a^{\#}$ or, more simply, by iteration, starting with the approximation $a^{\#} = 2 \cdot a^*$.

By setting $\alpha = \beta$, then $k_{1-\alpha} = k_{1-\beta} = k$ and the solution to Equation (15) is given by Equation (16):

$$a^{\#} = \frac{2 \cdot a^* + \left(k^2 \cdot w\right)/t_{\mathrm{g}}}{1 - k^2 \cdot u_{\mathrm{rel}}^2(w)} \tag{16}$$

8.5 Confidence limits

The lower, $a^{\triangleleft}$, and upper, $a^{\triangleright}$, limits of the confidence interval are calculated using Equations (17) and (18), respectively (see ISO 11929):

$$a^{\triangleleft} = a - k_p \cdot u(a) \text{ where } p = \omega \cdot (1 - \gamma/2) \quad (17)$$

$$a^{\triangleright} = a + k_q \cdot u(a) \text{ where } q = 1 - \omega \cdot \gamma/2 \quad (18)$$

where $\omega = \Phi\left[y/u(y)\right]$, Φ being the distribution function of the standardized normal distribution.

If $a \geqslant 4 \cdot u(a)$, ω may be set equal to 1 and Equation (19) applies:

$$a^{\triangleleft,\triangleright} = a \pm k_{1-\gamma/2} \cdot u(a) \quad (19)$$

$\gamma = 0{,}05$ and $k_{1-\gamma/2} = 1{,}96$ are often chosen by default.

8.6 Corrections for contributions from other radionuclides and background

8.6.1 General

In gamma spectrometry, it is frequently necessary to correct for two types of contributions.

a) The gamma line of the radionuclide being determined contains contributions from gamma radiation of another radionuclide in the sample. The contributing radionuclide has another gamma line from which the contribution to the line in question can be estimated taking into account the emission probabilities of the gamma lines.

b) The gamma line of the radionuclide being determined occurs also in the background of the spectrometer. By measuring a background spectrum without a sample for a counting time, t_0, this contribution can be corrected, taking into account the different counting times for the two spectra.

For both cases, the activity per unit of mass can be calculated using a model given in Equation (20):

$$a_{\mathrm{c}} = (n_{\mathrm{N},E} / t_{\mathrm{g}} - x \cdot n_{\mathrm{N0},E} / t_0) \cdot w \quad (20)$$

where x is a factor that is a function of the type of correction.

For the both types of contributions, this model gives the necessary correction.

8.6.2 Contribution from other radionuclides

The gamma line being corrected at the energy, E_1, has the net peak area, n_{N,E_1}. The contribution of the radionuclide is calculated using the ratio of the contributing radionuclide for gamma energy, E_1. Equation (20) gives the necessary correction with $x = P_{E_1} \cdot \varepsilon_1 / P_{E_2} \cdot \varepsilon_2$ and $t_0 = t_{\mathrm{g}}$. This yields Equation (21):

$$a_{\mathrm{c}} = (n_{\mathrm{N},E_1} - x \cdot n_{\mathrm{N},E_2}) \cdot w / t_{\mathrm{g}} \quad (21)$$

Neglecting the standard uncertainty of x, the standard uncertainty of a_{c} is calculated from Equation (22):

$$u^2(a_{\mathrm{c}}) = (w/t_{\mathrm{g}})^2 \left\{ n_{\mathrm{g},E_1} + u^2(n_{\mathrm{b},E_1}) + x^2 \left[n_{\mathrm{g},E_2} + u^2(n_{\mathrm{b},E_2}) \right] \right\} + a_{\mathrm{c}}^2 \cdot u_{\mathrm{rel}}^2(w) \quad (22)$$

and with a true value $\tilde{a}_c$ of a_c, Equation (23) can be derived:

$$\tilde{u}^2(\tilde{a}_c) = (w/t_g)^2 \left\{ \tilde{a}_c t_g / w + n_{b,E_1} + u^2(n_{b,E_1}) + x(n_{g,E_2} - n_{b,E_2}) + x^2 \left[n_{g,E_2} + u^2(n_{b,E_2}) \right] \right\} + \tilde{a}_c{}^2 \cdot u_{rel}^2(w) \quad (23)$$

Then, the decision threshold, a_c*, is given by Equation (24):

$$a_c{}^* = k_{1-\alpha} \cdot (w/t_g) \cdot \sqrt{n_{b,E_1} + u^2(n_{b,E_1}) + x(n_{g,E_2} - n_{b,E_2}) + x^2 \left[n_{g,E_2} + u^2(n_{b,E_2}) \right]} \quad (24)$$

and the detection limit, $a_c^\#$, by Equation (25)

$$a_c^\# = a_c^* + k_{1-\beta} \sqrt{(w/t_g)^2 \left\{ a_c^\# t_g / w + n_{b,E_1} + u^2(n_{b,E_1}) + x(n_{g,E_2} - n_{b,E_2}) + x^2 \left[n_{g,E_2} + u^2(n_{b,E_2}) \right] \right\} + a_c^{\#2} u_{rel}^2(w)} \quad (25)$$

The detection limit can be calculated by solving Equation (25) for $a_c^\#$ or, more simply, by an iteration starting with the approximation $a_c^\# = 2 \cdot a_c^*$.

By setting $\alpha = \beta$, then $k_{1-\alpha} = k_{1-\beta} = k$ and the solution to Equation (25) is given by Equation (26):

$$a_c^\# = \frac{2 \cdot a_c^* + \left(k^2 \cdot w\right)/t_g}{1 - k^2 \cdot u_{rel}^2(w)} \quad (26)$$

8.6.3 Contribution from background

In this case, Equation (20) is used for the correction by setting $x = 1$ and $u(x) = 0$. $n_{N0,E}$ is the net peak area of the gamma line in the background spectrum and t_0 is the counting time of the background spectrum. This yields Equation (27):

$$a_c = (n_{N,E}/t_g - n_{N0,E}/t_0) \cdot w \quad (27)$$

The standard uncertainty of a_c is calculated by Equation (28):

$$u^2(a_c) = w^2(n_{g,E}/t_g^2 + n_{g0,E}/t_0^2 + u^2(n_{b,E})/t_g^2 + u^2(n_{b0,E})/t_0^2) + a_c^2 \cdot u_{rel}^2(w) \quad (28)$$

and with a true value $\tilde{a}_c$ of a_c, Equation (29) is obtained:

$$\tilde{u}^2(\tilde{a}_c) = w^2 \left\{ \tilde{a}_c/t_g w + \left[n_{b,E} + u^2\left(n_{b,E}\right) \right] \Big/ t_g^2 + \left[n_{g0,E} + u^2\left(n_{b0,E}\right) \right] / t_0^2 + \left(n_{g0,E} - n_{b0,E}\right) \Big/ t_0 t_g \right\} + \tilde{a}_c{}^2 u_{rel}^2(w) \quad (29)$$

Then, the decision threshold, a_c*, is given by Equation (30):

$$a_c^* = k_{1-\alpha} \cdot w \cdot \sqrt{\left[n_{b,E} + u^2(n_{b,E}) \right] \Big/ t_g^2 + \left[n_{g0,E} + u^2(n_{b0,E}) \right] \Big/ t_0^2 + (n_{g0,E} - n_{b0,E}) / t_0 t_g} \quad (30)$$

and the detection limit $a_c^\#$ by Equation (31):

$$a_c^\# = a_c^* + k_{1-\beta} \sqrt{w^2 \left\{ a_c^\# / t_g w + \left[n_{b,E} + u^2(n_{b,E}) \right] / t_g^2 + \left[n_{g0,E} + u^2(n_{b0,E}) \right] / t_0^2 + (n_{g0,E} - n_{b0,E}) / t_0 t_g \right\} + a_c^{\#2} u_{rel}^2(w)} \quad (31)$$

The detection limit can be calculated by solving Equation (25) for $a_c^\#$ or, more simply, by iteration with a starting approximation $a_c^\# = 2 \cdot a_c^*$.

When taking $\alpha = \beta$, then $k_{1-\alpha} = k_{1-\beta} = k$ and the solution of Equation (31) is given by Equation (32):

$$a_c^\# = \frac{2 \cdot a_c^* + \left(k^2 \cdot w\right) / t_g}{1 - k^2 \cdot u_{rel}^2 (w)} \tag{32}$$

The limits of the confidence interval are calculated according to Equations (17) and (18).

9 Test report

The test report shall conform to ISO 17025 requirements and shall contain the following information:

a) reference to this part of ISO 18589;

b) identification of the sample;

c) units in which the results are expressed;

d) test result, $a \pm u$ or $a \pm U$, with the associated k value.

Complementary information can be provided such as

— probabilities α, β and (1 – γ);

— decision threshold and the detection limit;

— depending on the customer request, there are different ways to present the result:

  — when the activity per unit of mass, a, is compared with the decision threshold (see ISO 11929), the result of the measurement should be expressed as $\leqslant a^*$ when the result is below the decision threshold.

  — when the activity per unit of mass, a, is compared with the detection limit, the result of the measurement can be expressed as $\leqslant a^\#$ when the result is below the detection limit. If the detection limit exceeds the guideline value, it shall be documented that the method is not suitable for the measurement purpose.

— mention of any relevant information likely to affect the results.

Annex A
(informative)

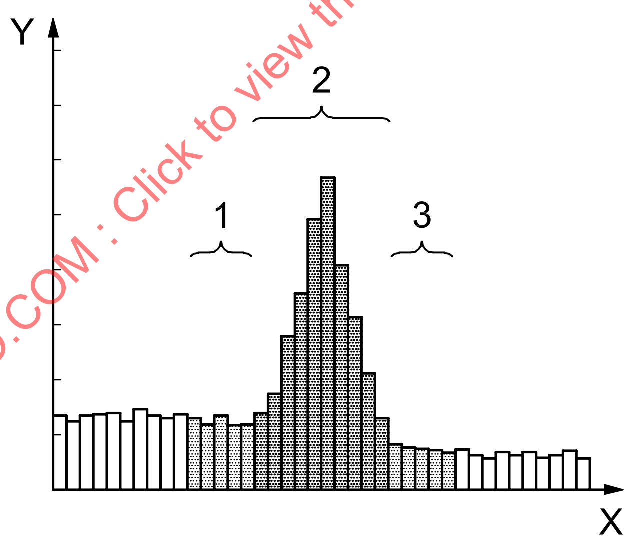
Calculation of the activity per unit mass from a gamma spectrum using a linear background subtraction

Frequently, the net peak area is calculated by subtraction of a linear background. In this case, three channel areas are defined in the spectrum: an area, P, symmetrical around the peak maximum covering p channels and two areas, $B1$ and $B2$, each covering b channels on either side of P; see Figure A.1. With the full width at half peak height, h, a length of the peak area $p \approx 2{,}5h$ and $b \approx p/2$ are frequently chosen. Then, n_N can be calculated according to Equation (5), as given in Equation (A.1):

$$n_\mathrm{g} = \sum_{i \in P} n_i \ , \ n_{B1} = \sum_{i \in B1} n_i \ , \ n_{B2} = \sum_{i \in B2} n_i \ , \text{ and } n_b = \frac{p}{2b} \cdot (n_{B1} + n_{B2}) \qquad \text{(A.1)}$$

The standard uncertainties are given in Equation (A.2):

$$u(n_\mathrm{g}) = \sqrt{n_\mathrm{g}} \ , \ u(n_b) = \frac{p}{2b} \cdot \sqrt{n_{B1} + n_{B2}} \ , \text{ and } u(n_\mathrm{N}) = \sqrt{n_\mathrm{g} + \left(\frac{p}{2b}\right)^2 \cdot (n_{B1} + n_{B2})} \qquad \text{(A.2)}$$



Key

X channel number, i

Y counts, n_i

1 area $B1$, with length b

2 area P, with length p

3 area $B2$, with length b

Figure A.1 — Scheme of linear background subtraction in gamma spectrometry

The standard uncertainty for the specific activity according to Equation (10) is given by Equation (A.3):

$$u^2(a) = w^2 \cdot \left[n_{\text{g}} + \left(\frac{p}{2b} \right)^2 \cdot (n_{B1} + n_{B2}) \right] \Big/ t_{\text{g}}^2 + a^2 \cdot u_{\text{ref}}^2(w) \tag{A.3}$$

In this case, $\tilde{u}^2(\tilde{a})$ is calculated by Equation (A.4):

$$\tilde{u}^2(\tilde{a}) = (w/t_{\text{g}})^2 \cdot \left\{ \tilde{a} \cdot t_{\text{g}}/w + \left[(p/2b) + (p/2b)^2 \right] \cdot (n_{B1} + n_{B2}) \right\} + \tilde{a}^2 \cdot u_{\text{rel}}^2(w) \tag{A.4}$$

where

$$u_{\text{rel}}(w) = \frac{u(w)}{w} \tag{A.5}$$

The decision threshold is calculated as given in Equation (A.6):

$$a^* = k_{1-\alpha} \cdot \tilde{u}(0) = k_{1-\alpha} \cdot (w/t_{\text{g}}) \cdot \sqrt{\frac{p}{2b} \cdot (n_{B1} + n_{B2}) + \left(\frac{p}{2b} \right)^2 \cdot (n_{B1} + n_{B2})} \tag{A.6}$$

and the detection limit, as given in Equation (A.7):

$$\begin{aligned} a^{\#} &= a^* + k_{1-\beta} \cdot \tilde{u}(a^{\#}) \\ &= a^* + k_{1-\beta} \cdot \sqrt{(w/t_{\text{g}})^2 \cdot \left[a^{\#} \cdot (t_{\text{g}}/w) + (p/2b) \cdot (n_{B1} + n_{B2}) + (p/2b)^2 \cdot (n_{B1} + n_{B2}) \right] + a^{\#2} \cdot u_{\text{rel}}^2(w)} \end{aligned} \tag{A.7}$$

When $\alpha = \beta$, then $k_{1-\alpha} = k_{1-\beta} = k$ and the solution of Equation (A.4) is given by Equation (A.8):

$$a^{\#} = \frac{2 \cdot a^* + \left(k^2 \cdot w \right) / t_g}{1 - k^2 \cdot u_{\text{rel}}^2(w)} \tag{A.8}$$

The limits of the confidence interval are calculated according to Equations (17) and (18).

Annex B
(informative)

Analysis of natural radionuclides in soil samples using gamma spectrometry

B.1 Introduction

Among the natural radionuclides, those besides K-40, belonging to a natural decay chain that can be measured using gamma spectrometry include U-238, Ra-226; Pb-210 of the uranium/radium decay chain; U-235 and Th-227 of the uranium/actinium decay chain; as well as Th-232, Ra-228 and Th-228 of the thorium decay chain; see Figure B.1.

Some radionuclides of the natural decay chains (e.g. U-238, Ra-228, Th-228) cannot be determined directly by gamma spectrometry but only by measuring their daughter radionuclides. In these cases, it is necessary to ensure that there is equilibrium between the parent radionuclide and the daughter radionuclides being measured. Radioactive equilibrium can be disrupted within the media being examined due to the different chemical or biochemical behaviour of the respective elements. For example, the radioactive equilibrium can be shifted strongly due to the different transfer behaviour of parent radionuclide and daughter radionuclides in the soil-vegetation-animal-milk chain.

In these cases it is recommended to keep the samples for a sufficiently long period before measuring them. On the other hand, interference of the radioactive equilibrium in the sample for measurement can result from escaping radon. In the case of measuring the short-lived decay products of Rn-222, the sample material shall additionally be put into a gas-tight glass container such that the dead volume in the glass container between the sample and the lid is as low as possible, and the sample left until radioactive equilibrium has been achieved. Since, as a rule, the severity of the interference with the equilibrium is not known, to be on the safe side, one should assume, when estimating the waiting period that initially there are practically no daughter radionuclides. In the case of a long-lived parent radionuclide and a short-lived decay product, this means that the waiting period should be at least six half-lives of the decay product.

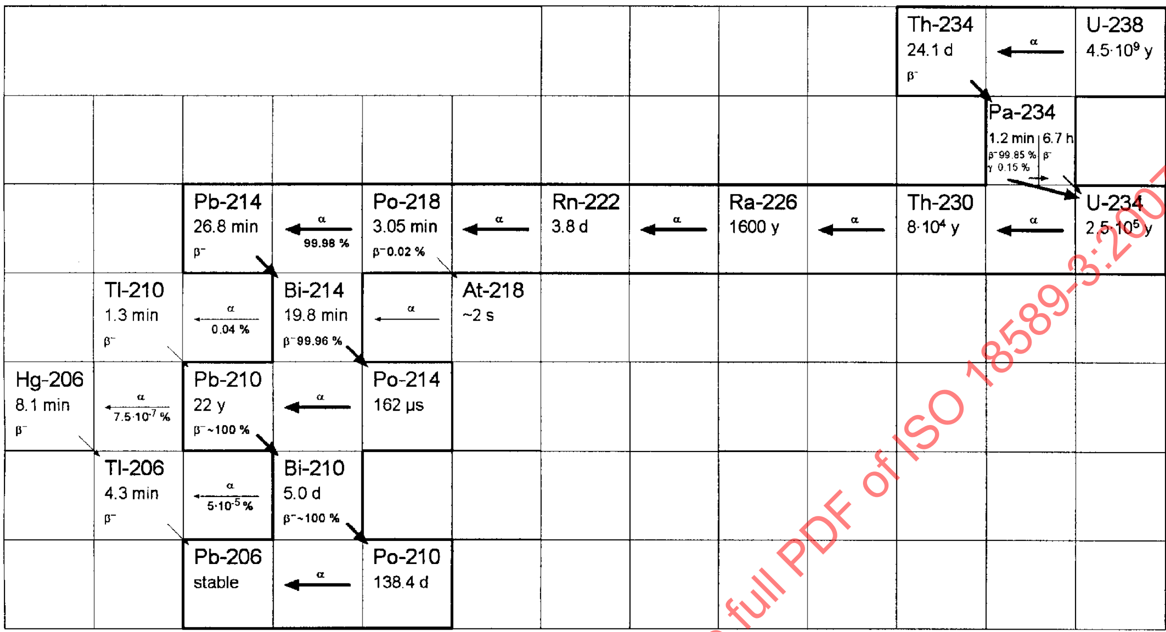
Measured activity values for the radionuclides of the natural decay chains that are not in radioactive equilibrium with the respective longer-lived parent radionuclide shall be counted back to a reference date; in most cases this should be the sampling date. Both the radioactive decay of the respective radionuclide and its decay products from its parent radionuclide shall be taken into account. Examples of such radionuclide pairs are Th-232/Ra-228, Ra-228/Th-228, Ra-226/Pb-210.

Another problem for the gamma spectrometric determination of natural radionuclides is the fact that some radionuclides show gamma lines that are identical or so near each other that it is not possible to resolve them by hardware or software means. In these cases, it is necessary to make corrections using other gamma lines; see 8.6.1 a).

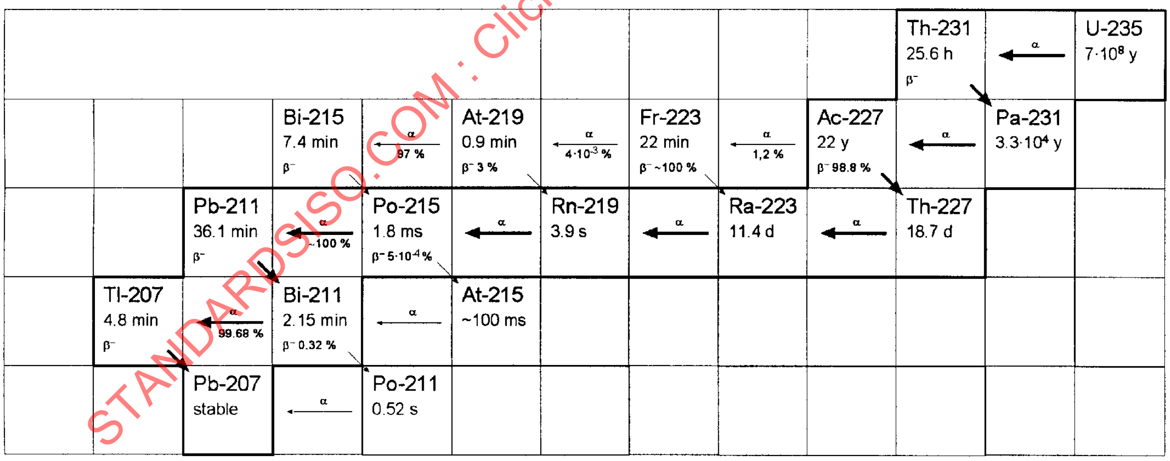
If this procedure cannot be carried out, a correction can be performed only by measuring radionuclides with other methods (e.g. alpha spectrometry or an emanation measurement). It is necessary to use these more sensitive methods when required detection limits cannot be achieved through gamma spectrometry (e.g. due to insufficient sample quantities).

Clauses B.2 to B.10 give additional explanations regarding the gamma spectrometric determination of common natural radionuclides in soil. For some radionuclides, the requirement of correcting self-absorption and summation losses are mentioned. Table B.1 lists the photon energies, E_γ, and the emission probabilities, p_E, of selected radionuclides [15].

When the decay chains are in equilibrium, the listed emission probabilities refer to the decay of the parent nuclide. For example, in the determination of the Th-228 content via Tl-208, it is not necessary to take into account that the branching ratio of the Bi-212 into Tl-208 is only 36,2 %; see Figure B.1 c). This is already considered when stating the emission probability in Table B.1.



a) **Uranium/radium** ($A = 4n + 2$)



b) **Uranium/actinium** ($A = 4n + 3$)

Figure B.1 (*continued*)