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Measurement of radioactivity in the environment — Soil —

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

Mesurage de la radioactivité dans l'environnement — Sol —

Partie 4: Mesurage des isotopes du plutonium (plutonium 238 et plutonium 239 + 240) par spectrométrie alpha

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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

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 18589-4 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: Measurements 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

ISO 18589 is published in several parts for use 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 soil. The first two parts are general in nature. Parts 3 to 5 deal with nuclide-specific measurements and Part 6 deals with non-specific measurements of gross alpha or gross beta activities.

Additional parts can 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 4:
Measurement of plutonium isotopes (plutonium 238 and plutonium 239 + 240) by alpha spectrometry

1 Scope

This part of ISO 18589 describes a method for measuring plutonium 238 and 239 + 240 isotopes in soil by alpha spectrometry samples using chemical separation techniques. The method can be used for any type of environmental study or monitoring. These techniques can also be used for measurements of very low levels of activity, one or two orders of magnitude less than the level of natural alpha-emitting radionuclides.

The mass of the test portion required depends on the assumed activity of the sample and the desired detection limit. In practice, it can range from 0,1 g to 100 g of the test sample.

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 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*

3 Terms, definitions and symbols

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

m	Mass of the test portion, expressed in kilograms
a	Activity per unit of mass, expressed in becquerel per kilogram
A	Activity of the tracer added, expressed in becquerel, at the time of measurement
t_g	Sample counting time, expressed in seconds

t_0	Background counting time, expressed in seconds
r_g, r_{gt}	Gross count rate per second from measured plutonium and tracer, respectively
r_0, r_{0t}	Background count rate per second from measured plutonium and tracer, respectively
R	Total measurement yield
$u(a)$	Standard uncertainty associated with the measurement result, expressed in becquerel per kilogram
U	Expanded uncertainty, expressed in becquerel per kilogram, calculated by $U = k \cdot u(a)$ with $k = 1, 2, \ldots$
a^*	Decision threshold, expressed in becquerel per kilogram
$a^\#$	Detection limit, expressed in becquerel per kilogram
$a^{\triangleleft}$, $a^{\triangleright}$	Lower and upper limits of the confidence interval, expressed in becquerel per kilogram

4 Principle

The plutonium is deposited as a thin source for measurement by alpha spectrometry using a grid chamber or semi-conductor detector-type apparatus. The sources are usually prepared by electro-deposition or co-precipitation after chemical separation and purification of the plutonium isotopes present in the test portion [1], [2], [3], [4]. Direct deposition on the planchette, such as electro-spraying, can also be used.

Specific chemical separation and purification procedures are required in order to avoid interference from the presence of natural or artificial α-emitters and stable nuclides in the sample, in quantities that are often greater than those of the plutonium isotopes during their measurement.

These procedures allow the removal of the main sources of interference, including

- the salt content of the leaching solutions, especially hydrolyzable elements, in order to prepare the thinnest deposited source,
- other α-emitting radionuclides, such as ^{241}Am and the thorium isotopes, whose emissions can interfere with those of plutonium isotopes.

The total yield for each analysis (chemical separation yield, thin-layer deposit and measurement) is determined by adding a standard solution of ^{236}Pu or ^{242}Pu considered as tracer. As a result, the procedure shall include a valence cycle, adjusting the tracer and the plutonium isotopes being measured to the same oxidation state, in order to achieve identical chemical behaviour for all of them.

5 Chemical reagents and equipment

The chemical reagents and equipment are described in Annex A for plutonium desorption, in Annexes B, C and D for chemical treatment and in Annexes E and F for the preparation of the deposited source.

All the chemical reagents required to carry out this procedure shall be of analytical grade.

6 Procedure

6.1 Plutonium desorption

It is necessary to desorb the plutonium from the soil test portion and into solution. When the plutonium is absorbed onto soil particles from an aqueous solution or onto global fallout particles directly deposited on the soil, it is readily desorbed by direct acid treatment. Plutonium metabolized by animals or plants forms an organic complex that can be found in soil samples. It is released by the destruction of organic matter present in the soil by calcination of the test sample at 500 °C to 600 °C. In some cases, when heated, plutonium can form refractory compounds that require hydrofluoric acid treatment or fusion to make them soluble [4].

Two methods of plutonium desorption are described in Annex A.

In order to quantify the recovery yield, a tracer is added at the start of this step of the procedure; time is allowed, usually up to one day, to obtain equilibrium before starting the plutonium desorption.

6.2 Chemical separation

There are three commonly used techniques for the chemical separation of plutonium: liquid-liquid extraction, extraction on an ion-exchange resin or specific-extraction chromatographic resin. One method from each technique is presented in Annexes B to D: organic solvent [5], separation by anionic resin [6] or by extraction chromatographic resin [7], [8].

6.3 Preparation of the source to be measured

The source can be prepared by deposition, either by electro-deposition on a planchette (a stainless steel disk), or by co-precipitation.

6.3.1 Electro-deposition method

Electro-deposition is carried out after the chemical separation of the plutonium from interfering elements. It allows the electrochemical deposition of the radionuclides in an ultra-thin layer onto the planchette [9], [10]. The procedure described in Annex E applies to the three chemical separation methods described in Annexes B, C and D.

NOTE Electro-deposition is not a selective method because the metal cations likely to form insoluble hydroxides can be deposited at the same time as the plutonium.

6.3.2 Co-precipitation method

Co-precipitation, using fluoride compounds, can be carried out after the chemical separation of the plutonium from other interfering elements. It allows the precipitation of the radionuclide(s) in the form of a thin layer on a filter. The procedure described in Annex F can be applied to the three chemical separation methods described in Annexes B to D.

6.4 Background determination

Measure the background using a blank prepared for the method chosen, starting with a clean test portion (or directly distilled water).

6.5 Measurement

The plutonium activity per unit of mass is calculated by counting the sample source for an appropriate time. The same equipment conditions should be used for the sample, the background and the calibration source measurements.

The counting time required depends on the sample and background count rates and also the detection limit and decision threshold required.

The spectra should be inspected to confirm good peak separation and no interfering peaks.

7 Expression of results

7.1 Calculation of the activity per unit of mass

The plutonium activity is calculated by integrating the number of counts in the corresponding peaks of the radionuclide tracer, ^{238}Pu and/or $^{239+240}$Pu, of the alpha emission spectrum of the sample, obtained by alpha spectrometry. The results of these integrations, divided by the counting time, are the gross count rates, r_{gt} and r_{g}, for the tracer and the plutonium isotopes, respectively.

r_{gt} and r_{g} are corrected for the background contribution and, if needed, for the contribution of the tailing of higher-energy peaks, which depends on the detector characteristics.

Background count rates are calculated from the alpha-emission spectrum of a blank sample by integrating the number of counts in the regions of interest (ROI) in which the peaks appear in the sample spectrum. The result of this integration, divided by the counting time, is the background count rate, $r_{0\mathrm{t}}$ and r_0, for the tracer and the plutonium isotopes, respectively.

NOTE The blank sample is obtained and measured by applying the procedure used in the analysis without soil and with or without tracer.

The activity per unit of mass, a, of the plutonium isotope is calculated as given in Equation (1):

$$a = \left(r_{\mathrm{g}} - r_0\right) / (m \cdot R) = \left(r_{\mathrm{g}} - r_0\right) \cdot w \qquad (1)$$

where $w = \dfrac{1}{m \cdot R}$.

The total measurement yield, R, is determined from the activity, A, of the tracer added, and the net count rate in the corresponding peak is calculated as given in Equation (2):

$$R = \left(r_{\mathrm{gt}} - r_{0\mathrm{t}}\right) / A \qquad (2)$$

The detector efficiency allows one to calculate the chemical yield. This value is important for quality control.

7.2 Standard uncertainty

According to ISO/IEC Guide 98-3 [13], the standard uncertainty of a is calculated by Equation (3):

$$\begin{aligned} u(a) &= \sqrt{w^2 \cdot \left[u^2\left(r_{\mathrm{g}}\right) + u^2\left(r_0\right)\right] + a^2 \cdot u_{\mathrm{rel}}^2(w)} \\ &= \sqrt{w^2 \cdot \left(r_{\mathrm{g}}/t_{\mathrm{g}} + r_0/t_0\right) + a^2 \cdot u_{\mathrm{rel}}^2(w)} \end{aligned} \qquad (3)$$

where the uncertainty of the counting time is neglected.

The relative standard uncertainty of w is calculated by Equation (4):

$$u_{\mathrm{rel}}^2(w) = u_{\mathrm{rel}}^2(R) + u_{\mathrm{rel}}^2(m) \tag{4}$$

The relative standard uncertainty of R is calculated by Equation (5):

$$\begin{aligned} u_{\mathrm{rel}}^2(R) &= u_{\mathrm{rel}}^2\left(r_{\mathrm{gt}} - r_{0\mathrm{t}}\right) + u_{\mathrm{rel}}^2(A) \\ &= \left(r_{\mathrm{gt}}/t_{\mathrm{g}} + r_{0\mathrm{t}}/t_0\right) \Big/ \left(r_{\mathrm{gt}} - r_{0\mathrm{t}}\right)^2 + u_{\mathrm{rel}}^2(A) \end{aligned} \tag{5}$$

$u_{\mathrm{rel}}(A)$ includes all the uncertainties related to the tracer activity, that is, those of the standard solution, of the preparation of the tracer solution and of the addition of the tracer solution to sample.

The calculation of the characteristic limits (see ISO 11929[12]) requires a determination of $\tilde{u}(\tilde{a})$, i.e., the standard uncertainty of a as a function of its true value, as calculated by Equation (6):

$$\tilde{u}(\tilde{a}) = \sqrt{w^2 \cdot \left[\left(\tilde{a}/w + r_0\right)/t_{\mathrm{g}} + r_0/t_0\right] + \tilde{a}^2 \cdot u_{\mathrm{rel}}^2(w)} \tag{6}$$

7.3 Decision threshold

The decision threshold, a^*, is obtained from Equation (6) for $\tilde{a} = 0$ (see ISO 11929). This yields Equation (7):

$$a^* = k_{1-\alpha} \cdot \tilde{u}(0) = k_{1-\alpha} \cdot w \cdot \sqrt{r_0/t_{\mathrm{g}} + r_0/t_0} \tag{7}$$

$a = 0{,}05$ with $k_{1-a} = 1{,}65$ is often chosen by default.

7.4 Detection limit

The detection limit, $a^{\#}$, is calculated by Equation (8) (see ISO 11929):

$$\begin{aligned} a^{\#} &= a^* + k_{1-\beta} \cdot \tilde{u}\left(a^{\#}\right) \\ &= a^* + k_{1-\beta} \cdot \sqrt{w^2 \cdot \left[\left(a^{\#}/w + r_0\right)\Big/t_{\mathrm{g}} + r_0/t_0\right] + a^{\#2} \cdot u_{\mathrm{rel}}^2(w)} \end{aligned} \tag{8}$$

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

The detection limit can be calculated by solving Equation (8) for $a^{\#}$ or, more simply, by iteration with a starting approximation $a^{\#} = 2 \cdot a^*$.

By taking $\alpha = \beta$, then $k_{1-\alpha} = k_{1-\beta} = k$ and the solution of Equation (8) is given by Equation (9):

$$a^{\#} = \frac{2 \cdot a^* + \left(k^2 \cdot w\right)/t_{\mathrm{g}}}{1 - k^2 \cdot u_{\mathrm{rel}}^2(w)} \tag{9}$$

7.5 Confidence limits

The lower limit, $a^{\triangleleft}$, and the upper limit, $a^{\triangleright}$, of the confidence interval are calculated using Equations (10) and (11) (see ISO 11929):

$$a^{\triangleleft} = a - k_p \cdot u(a);\ p = \omega \cdot (1 - \gamma/2) \tag{10}$$

$$a^{\triangleright} = a + k_q \cdot u(a);\ q = 1 - \omega \cdot \gamma/2 \tag{11}$$

where

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

ω may be set equal to 1 if $a \geqslant 4 \cdot u(a)$ and Equation (12) applies:

$$a^{\triangleleft,\triangleright} = a \pm k_{1-\gamma/2} \cdot u(a) \tag{12}$$

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

8 Test report

The test report shall to conform to ISO/IEC 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, $a \pm u(a)$ or $a \pm U$, with the associated k value.

Complementary information can be provided such as the following:

- probabilities α, β and $(1 - \gamma)$;
- 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;
- any relevant information likely to affect the results.