
**Measurement of radioactivity in the
environment — Soil —**

Part 4:
**Plutonium 238 and plutonium 239
+ 240 — Test method using alpha
spectrometry**

Mesurage de la radioactivité dans l'environnement — Sol —

*Partie 4: Plutonium 238 et plutonium 239 + 240 — Méthode d'essai
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.

The procedures used to develop this document and those intended for its further maintenance are described in the ISO/IEC Directives, Part 1. In particular the different approval criteria needed for the different types of ISO documents should be noted. This document was drafted in accordance with the editorial rules of the ISO/IEC Directives, Part 2 (see www.iso.org/directives).

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. Details of any patent rights identified during the development of the document will be in the Introduction and/or on the ISO list of patent declarations received (see www.iso.org/patents).

Any trade name used in this document is information given for the convenience of users and does not constitute an endorsement.

For an explanation on the voluntary nature of standards, the meaning of ISO specific terms and expressions related to conformity assessment, as well as information about ISO's adherence to the World Trade Organization (WTO) principles in the Technical Barriers to Trade (TBT) see the following URL: www.iso.org/iso/foreword.html.

This document was prepared by Technical Committee ISO/TC 85, *Nuclear energy, nuclear technologies and radiological protection*, Subcommittee SC 2, *Radiological protection*.

This second edition cancels and replaces the first edition (ISO 18589-4:2009), which has been technically revised.

The main change compared to the previous edition are as follows:

- The introduction has been reviewed accordingly to the generic introduction adopted for the standards published on the radioactivity measurement in the environment.
- Reference in the text to ISO 18589-2 has been made mandatory.

A list of all parts in the ISO 18589 series can be found on the ISO website.

Any feedback or questions on this document should be directed to the user's national standards body. A complete listing of these bodies can be found at www.iso.org/members.html.

Introduction

Everyone is exposed to natural radiation. The natural sources of radiation are cosmic rays and naturally occurring radioactive substances which exist in the earth and flora and fauna, including the human body. Human activities involving the use of radiation and radioactive substances add to the radiation exposure from this natural exposure. Some of those activities, such as the mining and use of ores containing naturally-occurring radioactive materials (NORM) and the production of energy by burning coal that contains such substances, simply enhance the exposure from natural radiation sources. Nuclear power plants and other nuclear installations use radioactive materials and produce radioactive effluent and waste during operation and decommissioning. The use of radioactive materials in industry, agriculture and research is expanding around the globe.

All these human activities give rise to radiation exposures that are only a small fraction of the global average level of natural exposure. The medical use of radiation is the largest and a growing man-made source of radiation exposure in developed countries. It includes diagnostic radiology, radiotherapy, nuclear medicine and interventional radiology.

Radiation exposure also occurs as a result of occupational activities. It is incurred by workers in industry, medicine and research using radiation or radioactive substances, as well as by passengers and crew during air travel. The average level of occupational exposures is generally below the global average level of natural radiation exposure (see Reference [1]).

As uses of radiation increase, so do the potential health risk and the public's concerns. Thus, all these exposures are regularly assessed in order to:

- improve the understanding of global levels and temporal trends of public and worker exposure;
- evaluate the components of exposure so as to provide a measure of their relative importance;
- identify emerging issues that may warrant more attention and study. While doses to workers are mostly directly measured, doses to the public are usually assessed by indirect methods using the results of radioactivity measurements of waste, effluent and/or environmental samples.

To ensure that the data obtained from radioactivity monitoring programs support their intended use, it is essential that the stakeholders (for example nuclear site operators, regulatory and local authorities) agree on appropriate methods and procedures for obtaining representative samples and for handling, storing, preparing and measuring the test samples. An assessment of the overall measurement uncertainty also needs to be carried out systematically. As reliable, comparable and 'fit for purpose' data are an essential requirement for any public health decision based on radioactivity measurements, international standards of tested and validated radionuclide test methods are an important tool for the production of such measurement results. The application of standards serves also to guarantee comparability of the test results over time and between different testing laboratories. Laboratories apply them to demonstrate their technical competences and to complete proficiency tests successfully during interlaboratory comparisons, two prerequisites for obtaining national accreditation.

Today, over a hundred International Standards are available to testing laboratories for measuring radionuclides in different matrices.

Generic standards help testing laboratories to manage the measurement process by setting out the general requirements and methods to calibrate equipment and validate techniques. These standards underpin specific standards which describe the test methods to be performed by staff, for example, for different types of sample. The specific standards cover test methods for:

- naturally-occurring radionuclides (including ^{40}K , ^3H , ^{14}C and those originating from the thorium and uranium decay series, in particular ^{226}Ra , ^{228}Ra , ^{234}U , ^{238}U and ^{210}Pb) which can be found in materials from natural sources or can be released from technological processes involving naturally occurring radioactive materials (e.g. the mining and processing of mineral sands or phosphate fertilizer production and use);

- human-made radionuclides, such as transuranium elements (americium, plutonium, neptunium, and curium), ^3H , ^{14}C , ^{90}Sr and gamma-ray emitting radionuclides found in waste, liquid and gaseous effluent, in environmental matrices (water, air, soil and biota), in food and in animal feed as a result of authorized releases into the environment, fallout from the explosion in the atmosphere of nuclear devices and fallout from accidents, such as those that occurred in Chernobyl and Fukushima.

The fraction of the background dose rate to man from environmental radiation, mainly gamma radiation, is very variable and depends on factors such as the radioactivity of the local rock and soil, the nature of building materials and the construction of buildings in which people live and work.

A reliable determination of the activity concentration of gamma-ray emitting radionuclides in various matrices is necessary to assess the potential human exposure, to verify compliance with radiation protection and environmental protection regulations or to provide guidance on reducing health risks. Gamma-ray emitting radionuclides are also used as tracers in biology, medicine, physics, chemistry, and engineering. Accurate measurement of the activities of the radionuclides is also needed for homeland security and in connection with the Non-Proliferation Treaty (NPT).

This document describes the generic requirements to quantify the activity of ^{238}Pu and $^{239} + ^{240}$ isotopes of plutonium in soil samples after proper sampling, sample handling and test sample preparation in a testing laboratory or in situ.

This document is to be used in the context of a quality assurance management system (ISO/IEC 17025).

ISO 18589 is published in several parts for use jointly or separately according to needs. These parts are complementary and are addressed to those responsible for determining the radioactivity present in soil, bedrocks and ore (NORM or TENORM). The first two parts are general in nature describe the setting up of programmes and sampling techniques, methods of general processing of samples in the laboratory (ISO 18589-1), the sampling strategy and the soil sampling technique, soil sample handling and preparation (ISO 18589-2). ISO 18589-3 to ISO 18589-5 deal with nuclide-specific test methods to quantify the activity concentration of gamma emitters radionuclides (ISO 18589-3 and ISO 20042), plutonium isotopes (ISO 18589-4) and ^{90}Sr (ISO 18589-5) of soil samples. ISO 18589-6 deals with non-specific measurements to quantify rapidly gross alpha or gross beta activities and ISO 18589-7 describes in situ measurement of gamma-emitting radionuclides.

The test methods described in ISO 18589-3 to ISO 18589-6 can also be used to measure the radionuclides in sludge, sediment, construction material and products following proper sampling procedure.

This document is one of a set of International Standards on measurement of radioactivity in the environment.

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:

Plutonium 238 and plutonium 239 + 240 — Test method using alpha spectrometry

1 Scope

This document describes a method for measuring ^{238}Pu 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 test methods described in this document can also be used to measure the radionuclides in sludge, sediment, construction material and products following proper sampling procedure^{[2][3][4][5][7][8]}.

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 documents are referred to in the text in such a way that some or all of their content constitutes requirements 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.

<https://standards.iteh.ai/>
<https://www.iso.org/iso/11074.html>, *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*

ISO 80000-10, *Quantities and units — Part 10: Atomic and nuclear physics*

ISO/IEC Guide 98-3, *Uncertainty of measurement — Part 3: Guide to the expression of uncertainty in measurement (GUM:1995)*

3 Terms, definitions and symbols

For the purposes of this document, the terms and definitions given in ISO 11074, ISO 18589-1 and ISO 80000-10 apply.

ISO and IEC maintain terminological databases for use in standardization at the following addresses:

- ISO Online browsing platform: available at <https://www.iso.org/obp>
- IEC Electropedia: available at <http://www.electropedia.org/>

4 Symbols

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 $k = 1, 2, \dots$;
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.

5 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^{[9][10][11][12]}. 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 hydrolysable 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.

6 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.

7 Procedure

7.1 Plutonium desorption

It is necessary to desorb the plutonium from the soil test portion and into solution. When the plutonium is adsorbed 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 550 °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^[12].

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.

7.2 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^[14], separation by anionic resin^[14] or by extraction chromatographic resin^{[16][17]}.

7.3 Preparation of the source to be measured

7.3.1 General

The source can be prepared by deposition, either by electro-deposition on a planchette (a stainless steel disk) ([7.3.2](#)), or by co-precipitation ([7.3.3](#)).

7.3.2 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^{[17][18]}. 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.

7.3.3 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](#).

7.4 Background determination

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

7.5 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 measurements of the sample, the background and the reference measurements standards.

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.

8 Expression of results

8.1 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 $^{239} + ^{240}\text{Pu}$, 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, $r_{0\text{t}}$ and r_0 , for the tracer and the plutonium isotopes, respectively.

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 [Formula \(1\)](#):

$$a = (r_{\text{gt}} - r_0) / (m \cdot R) = (r_{\text{g}} - r_0) \cdot w \quad \text{ISO 18589-4:2019} \quad (1)$$

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

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 [Formula \(2\)](#):

$$R = (r_{\text{gt}} - r_{0\text{t}}) / A \quad (2)$$

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

8.2 Standard uncertainty

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

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

where the uncertainty of the counting time is neglected.

The relative standard uncertainty of w is calculated by [Formula \(4\)](#):

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

The relative standard uncertainty of R is calculated by [Formula \(5\)](#):

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

$u_{\text{rel}}(A)$ 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 [in accordance with ISO 11929 (all parts)] 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 [Formula \(6\)](#):

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

8.3 Decision threshold

The decision threshold, a^* , is obtained from [Formula \(6\)](#) for $\tilde{a}=0$ [in accordance with ISO 11929 (all parts)]. This yields [Formula \(7\)](#):

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

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

8.4 Detection limit

The detection limit, $a^\#$, is calculated by [Formula \(8\)](#) [in accordance with ISO 11929 (all parts)]:

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

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

The detection limit can be calculated by solving [Formula \(8\)](#) 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 [Formula \(8\)](#) is given by [Formula \(9\)](#):

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

8.5 Confidence limits

The lower limit, a^\triangleleft , and the upper limit, a^\triangleright , of the confidence interval are calculated using [Formulae \(10\)](#) and [\(11\)](#) [in accordance with ISO 11929 (all parts)]:

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