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Measurement of the radioactivity in the environment – Air: tritium – Test method using bubbler sampling

Mesurage de la radioactivité dans l'environnement — Air: tritium — Méthode d'essai avec échantillonnage par barbotage

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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*, SC 2, *Radiological protection*.

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 include cosmic rays and naturally occurring radioactive substances which exist on Earth such as flora, fauna or 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 effluents 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 [42])

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

- a) improve the understanding of global levels and temporal trends of public and worker exposure,
- b) evaluate the components of exposure so as to provide a measure of their relative impact, and
- c) 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, liquid and/or gaseous effluents and/or environmental samples.

Radioactivity from several naturally-occurring and anthropogenic sources is present throughout the environment. Thus, atmosphere can contain radionuclides of natural, human-made, or both origins.

- Natural 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 transuranic elements (americium, plutonium, neptunium, and curium), ^3H , ^{14}C , ^{90}Sr and gamma-ray emitting radionuclides can also be found gaseous effluent discharges, 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.

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 sampling, 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

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comparability of the test results over time and between different testing laboratories. Laboratories apply them to demonstrate their technical competencies and to complete proficiency tests successfully during interlaboratory comparisons, two prerequisites for obtaining national accreditation.

Today, over a hundred International standards, prepared by Technical Committees of the International Organization for Standardization, including those produced by ISO/TC 85, and the International Electrotechnical Commission (IEC), are available to testing laboratories for measuring radionuclides in different matrices.

Tritium (^3H) is a radioactive isotope of hydrogen. It is a pure beta emitting radionuclide, with a maximum energy equal to 18,591 (1) keV and a radiological half-life equal to 12,312 (0,025) years (see Reference [23]). It is naturally occurring and continuously produced in the upper atmosphere by interaction of cosmic rays with nitrogen and oxygen nuclei (see Reference [34]).

Two main chemical species of both natural and anthropogenic tritium are present in the environment. The most abundant chemical form is tritiated water (HTO) (see Reference [45]). Tritium can also be present in the form of tritiated gas (HT or T_2) usually present in the vicinity of tritium-emitting facilities (see Reference [56]), tritiated methane (CH_3T), or in other various organic forms of tritium commonly observed in terrestrial, aquatic continental, and marine ecosystems (see References [6], [7], [8] and [9]).

Anthropogenic tritium compounds come from radioactive releases of nuclear facilities i.e., nuclear power plants, irradiated fuel reprocessing and recycling plants, military defence, medical research applications, and past atmospheric testing of nuclear devices (see Annex A).

This document describes the method to assess the activity concentration of atmospheric tritium via air sampling by bubbler devices which trap tritiated water vapour and tritiated gas in a water solution. The method can be used for any type of environmental study or monitoring.

The test method is used in the context of a quality assurance management system (ISO/IEC 17025). It can be adapted so that the characteristic limits, decision threshold, detection limit and uncertainties ensure that the test results of the atmospheric tritium activity concentrations can be verified to be below guidance levels required by a national authority for either planned or existing situations or for an emergency situation.

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Measurement of the radioactivity in the environment – Air: tritium – Test method using bubbler sampling

1 Scope

This document describes a test method to determine the activity concentration of atmospheric tritium by trapping tritium in air by bubbling through a water solution. Atmospheric tritium activity concentration levels are expressed in becquerel per cubic metre (Bq·m⁻³).

The formulae are given for a sampling system with four bubblers. They can also be applied to trapping systems with only one trapping module consisting of two bubblers if only HTO is in the atmosphere to be sampled.

This document does not cover laboratory test sample results, in becquerel per litre of trapping solution, according to ISO 9698 or ISO 13168.

The test method detection limit result is between 0,2 Bq·m⁻³ and 0,5 Bq·m⁻³ when the sampling duration is about one week.

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.

~~<std>ISO 4788, Laboratory glassware — Graduated measuring cylinders</std>~~
~~<std>ISO 4788, Laboratory glassware — Graduated measuring cylinders</std>~~
~~ISO 9698, Water quality — Tritium — Test method using liquid scintillation counting</std>~~
~~<std>ISO 11929-1, Determination of the characteristic limits (decision threshold, detection limit and limits of the coverage interval) for measurements of ionizing radiation — Fundamentals and application — Part 1: Elementary applications</std>~~
~~<std>ISO 13168, Water quality — Simultaneous determination of tritium and carbon 14 activities — Test method using liquid scintillation counting</std>~~
~~<std>ISO/IEC Guide 98-3, Uncertainty of measurement — Part 3: Guide to the expression of uncertainty in measurement (GUM:1995)</std>~~
~~<std>ISO/IEC Guide 99, International vocabulary of metrology — Basic and general concepts and associated terms (VIM)</std>~~
~~<std>ISO/IEC 17025, General requirements for the competence of testing and calibration laboratories</std>~~
~~<std>ISO 80000-10, Quantities and units — Part 10: Atomic and nuclear physics</std>~~

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3 Terms, definitions and symbols

For the purposes of this document, the definitions, symbols and abbreviations given in, ~~ISO/IEC Guide 98-3, ISO/IEC Guide 99, ISO 80000-10~~ and the following 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~~<https://www.iso.org/obp>
- IEC Electropedia: available at ~~https://www.electropedia.org/~~<https://www.electropedia.org/>

3.1 Terms and definitions

3.1.1

aerosol

dispersion of very fine solid particles or liquid droplets in air or gases

3.1.2

air sample

representative part of the atmosphere sampled routinely, intermittently or continuously to examine its various characteristics

3.1.3

bubbler

glass container that holds the *trapping water solution* (3.1.11)

3.1.4

bubbler sample

bubbler (3.1.3) which an *air sample* (3.1.2) bubbled through

3.1.5

oxidizing efficiency

ratio of atmospheric tritium gas compounds converted into *tritiated water vapour* (3.1.13), oxidized with a catalytic converter furnace, to tritium gas compounds in the atmosphere during the sampling period

3.1.6

sampling module

composed of two *bubblers* (3.1.3) connected in series to trap tritium species *HTO* (3.1.13) or *no-HTO* (3.1.12)

3.1.7

sampling system

device for sampling atmospheric tritium by bubbling through a water solution that consists of a sampling head which is the air inlet, a transport line, collector, and flow conditioning system

Note 1 to entry:— Recorded samples are analysed off-line in a testing laboratory.

3.1.8

standard conditions

temperature of 273,13 K (0 °C) and a pressure of 101 325 Pa

Note 1 to entry:— Used to convert air densities into a common basis. Other temperature and pressure conditions may be used and should be applied consistently.

3.1.9

test sample

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representative volume taken from the *bubbler sample* (3.1.4) to analyse the tritium activity concentration by a testing laboratory

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3.1.10 trapping efficiency

ratio of *tritiated water vapour* (3.1.13) activity concentration collected, during the sampling period, to atmospheric *tritiated water vapour* (3.1.13) activity concentration

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3.1.11 trapping water solution

Any types of colourless water with no apparent biological activities to trap atmospheric tritium by molecular and/or isotopic exchange between the tritium atoms in water vapour of the air and the hydrogen atoms of the water molecules in solution

3.1.12 tritiated gas

no-HTO tritium gas compounds where HT and CH₃T molecules are predominant chemical gas species in atmosphere

3.1.13 tritiated water vapour

HTO water vapour where one hydrogen atom of a water molecule is substituted by one tritium atom

3.2 Symbols, definitions and units

Table 1 — Symbols, definitions and units

Symbol	Definition and unit
A_i	tritium activity of the bubbler sample A_i , in becquerel (Bq) where $i = 1, 2, 3$ or 4
A_{ref}	reference tritium activity of tritiated water vapour (HTO) in the atmosphere in becquerel (Bq)
c_i	tritium activity concentration of the test sample c_i , in becquerel per litre (Bq·l ⁻¹)
c_i^*	decision threshold of the tritium activity concentration of the test sample c_i , in becquerel per litre (Bq·l ⁻¹)
$c_i^{\#}$	detection limit of the tritium activity concentration of the test sample c_i , in becquerel per litre (Bq·l ⁻¹)
c_{ref}	reference tritium activity concentration of tritiated water vapour (HTO) in the atmosphere in becquerel per cubic metre (Bq·m ⁻³) at standard conditions
c_w	tritium activity concentration of tritiated water vapour (HTO) in the atmosphere in becquerel per cubic metre (Bq·m ⁻³) at standard conditions
c_g	tritium activity concentration of tritiated gas compounds (no-HTO) in the atmosphere in becquerel per cubic metre (Bq·m ⁻³) at standard conditions
c_w^* and c_g^*	decision threshold of the tritium activity concentration of HTO and no-HTO respectively in the atmosphere in becquerel per cubic metre (Bq·m ⁻³) at standard conditions

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Symbol	Definition and unit
$c_w^{\#}$ and $c_g^{\#}$	detection limit of the tritium activity concentration of HTO and no-HTO respectively in the atmosphere in becquerel per cubic metre $(\text{Bq}\cdot\text{m}^{-3})(\text{Bq}\cdot\text{m}^{-3})$ at standard conditions
$c_w^{\triangleleft}, c_w^{\triangleleft}$ and $c_g^{\triangleleft}, c_g^{\triangleleft}$	lower and upper limits of the probabilistically symmetric coverage interval of HTO and no-HTO respectively in the atmosphere in becquerel per cubic metre $(\text{Bq}\cdot\text{m}^{-3})(\text{Bq}\cdot\text{m}^{-3})$ at standard conditions
$c_w^{\triangleleft}, c_w^{\triangleleft}$ and $c_g^{\triangleleft}, c_g^{\triangleleft}$	lower and upper limits of the shortest coverage interval of HTO and no-HTO respectively in the atmosphere in becquerel per cubic metre $(\text{Bq}\cdot\text{m}^{-3})(\text{Bq}\cdot\text{m}^{-3})$ at standard conditions
ε_{Bi}	trapping efficiency of each bubbler sample i
ε_F	oxidizing efficiency of the catalytic converter furnace
k	coverage factor with $k=1, 2, 3, \dots, k=1, 2, 3, \dots$
q_p	air flow rate of sampling system in cubic metre per hour $(\text{m}^3\cdot\text{h}^{-1})(\text{m}^3\cdot\text{h}^{-1})$ at standard conditions
t_i	counting duration of the test sample i in seconds (s)
t_p	sampling duration in hour (h)
$u(c_i)$	standard uncertainty of the tritium activity concentration of the test sample i in becquerel per litre $(\text{Bq}\cdot\text{l}^{-1})(\text{Bq}\cdot\text{l}^{-1})$
$u(y)$	standard uncertainty associated with parameter y , result $(k=1)(k=1)$
$U(y)$	expanded uncertainty calculated by $U(y) = k \cdot u(y)$ with $k > 1, k > 1$
$u_{\text{rel}}(y)$	relative standard uncertainty associated with parameter y , result calculated by $u_{\text{rel}}(y) = u(y) \cdot y^{-1}$
$U_{\text{rel}}(y)$	relative expanded uncertainty calculated by $U_{\text{rel}}(y) = k \cdot u_{\text{rel}}(y)$ with $k > 1$
V	sampled air volume in cubic metre $(\text{m}^3)(\text{m}^3)$ at standard conditions where $V = q_p \cdot t_p$
V_{Bi}	water volume of bubbler sample, B_i , at the end of sampling duration in litre (l)
V_{Bref}	initial same volume of water in each bubbler, B_i , in litre (l)
w_i	correction factor for the tritium activity concentration of the test sample i in per litre $(\text{l}^{-1})(\text{l}^{-1})$

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4 Principle

The bubbler sampling method consists of trapping airborne tritium compounds in water solution. The sampled air is continuously pumped through a series of bubblers containing trapping water and transformed as micro-bubbles in the water. The micro bubbles allow for the efficient capture of airborne tritium water vapour in the trapping solution by molecular and isotopic exchanges.

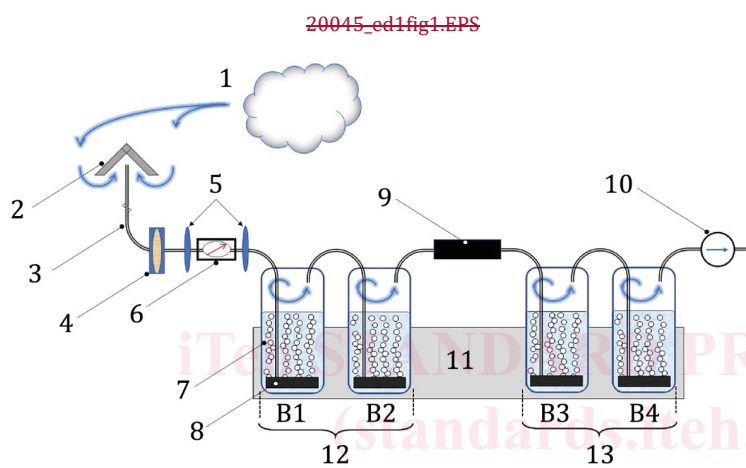
After filtering of solid aerosol particles by the dust filter, the sampled air passes through a first sampling module of two bubblers. This unit collects tritiated water vapour from the air. A second module, specifically for no-HTO compounds, can also be connected in series. In this case, the sampled air shall

pass through a catalytic converter furnace which converts no-HTO compounds into HTO. This second module collects residual HTO not trapped by the first module and no-HTO compounds that have been converted into HTO.

The flow of air through the sampling system is controlled by a mass flow metre.

The Figure 1 shows a diagram of an example of a sample system. Other air flow control and injection configurations can be used.

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Key

- 1 atmospheric air to monitor at temperature, T , and relative humidity RH in %
- 2 sampling head
- 3 connection pipe
- 4 anti-dust filter
- 5 hydrophobic filter
- 6 mass flow meter
- 7 bubbler with trapping water solution
- 8 micro-bubbles generator
- 9 catalytic converter furnace
- 10 pump
- 11 cooling module
- 12 first module for HTO trapping (bubblers B1 and B2)
- 13 second module for no-HTO and residual HTO trapping (bubblers B3 and B4)

Figure 1 — Example of an atmospheric air sampling system diagram with two sampling modules

At the end of the sampling period, trapping solutions shall be collected separately and transported as soon as possible to the testing laboratory.

Tritium activity concentration of water from each bubbler sample, in becquerel per litre of bubbler sample, shall be estimated by liquid scintillation in accordance with ISO 9698 or ISO 13168.

Activity concentrations of atmospheric tritium shall be calculated taking into account:

- air volume sampled;

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