IEC TR 61577-5

Edition 1.0 2019-07

TECHNICAL REPORT

colour inside

Radiation protection instrumentation – Radon and radon decay product measuring instruments –
Part 5: General properties of radon and radon decay products and their measurement methods

IEC TR 61577-5:2019-07(en)

IEC Central Office
3, rue de Varembé
CH-1211 Geneva 20
Switzerland

Tel.: +41 22 919 02 11
info@iec.ch
www.iec.ch

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INTERNATIONAL
ELECTROTECHNICAL
COMMISSION

ICS 13.280

ISBN 978-2-8322-7123-0

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INTERNATIONAL ELECTROTECHNICAL COMMISSION

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RADIATION PROTECTION INSTRUMENTATION – RADON AND RADON DECAY PRODUCT MEASURING INSTRUMENTS –

Part 5: General properties of radon and radon decay products and their measurement methods

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IEC TR 61577-5, which is a Technical Report, has been prepared by subcommittee 45B: Radiation protection instrumentation, of IEC technical committee 45: Nuclear instrumentation.

The text of this Technical Report is based on the following documents:

Enquiry draft	Report on voting
45B/912/DTR	45B/926/RVDTR

Full information on the voting for the approval of this technical report can be found in the report on voting indicated in the above table.

This document has been drafted in accordance with the ISO/IEC Directives, Part 2.

A list of all parts in the IEC 61577 series, published under the general title *Radiation protection instrumentation – Radon and radon decay product measuring instruments*, can be found on the IEC website.

The committee has decided that the contents of this document will remain unchanged until the stability date indicated on the IEC website under "http://webstore.iec.ch" in the data related to the specific document. At this date, the document will be

- reconfirmed,
- withdrawn,
- replaced by a revised edition, or
- amended.

A bilingual version of this publication may be issued at a later date.

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RADIATION PROTECTION INSTRUMENTATION – RADON AND RADON DECAY PRODUCT MEASURING INSTRUMENTS –

Part 5: General properties of radon and radon decay products and their measurement methods

1 Scope

This part of IEC 61577 provides basic data and technical information in order to support the design of instruments and their practical application for the measurement. The document covers ^{222}Rn as well as ^{220}Rn and the short-lived decay products of both. It is an accompanying document for the application of the technical standards series IEC 61577, and provides physical and technical fundamentals of the measurements methods. For more information, reference is made to the Bibliography.

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.

IEC 61577-1, *Radiation protection instrumentation – Radon and radon decay product measuring instruments – Part 1: General principles*

IEC 61577-2, *Radiation protection instrumentation – Radon and radon decay product measuring instruments – Part 2: Specific requirements for ^{222}Rn and ^{220}Rn measuring instruments*

IEC 61577-3, *Radiation protection instrumentation – Radon and radon decay product measuring instruments – Part 3: Specific requirements for radon decay product measuring instruments*

IEC 61577-4, *Radiation protection instrumentation – Radon and radon decay product measuring instruments – Part 4: Equipment for the production of reference atmospheres containing radon isotopes and their decay products (STAR)*

IEC TR 62461:2015, *Radiation protection instruments – Determination of uncertainty in measurement*

3 Symbols, quantities and units

3.1 Symbols

L	Ostwald coefficient
$C_{Rn,liquid}$, C_{Rn}	Activity concentration of radon in a liquid, activity concentration of radon in air in Becquerels per cubic meter (Bq·m^{-3})
T_{H_2O}	Temperature of water in degrees Celsius (°C)
A, A_i	Activity, activity of radionuclide i in Becquerels (Bq)
λ_i	Radioactive decay constant of radionuclide i in per second (s^{-1})
q_i	Supply rate, production rate of radionuclide i in per second (s^{-1})
t, t_s	Time, sampling time in seconds (s)
V	Volume in cubic metres (m^3)
EEC, C_{eq}	Equilibrium equivalent concentration in Becquerels per cubic metres (Bq·m^{-3})
k_i	Weighting coefficient of radionuclide i
C_p	Potential alpha energy concentration in Joules per cubic metre (J·m^{-3})
F	Equilibrium factor
P_{Rn}	Exposure to radon in Becquerel hours per cubic metre (Bq·h·m^{-3})
ε_p	Potential alpha energy in Joules (J)
N_i	Number of atoms of radionuclide i
P_p	Potential alpha energy exposure in Joule hours per cubic metre (J·h·m^{-3})
$u(x)$	Standard uncertainty of quantity x
$U(x)$	Expanded uncertainty $U(x)=k\cdot u(x)$ with the coverage factor $k=2$
v	Volume flow rate of air in litres per minute (m^3 s^{-1})
I_i	Number of counts of radionuclide i
ε , ε_s , ε_{si} , ε_c ε_{ci}	Efficiency, sampling efficiency, sampling efficiency of radionuclide i, counting efficiency of radionuclide i
α_k , α	Elapsed time after cessation of sampling until the beginning of time interval k, time at which the counting interval begins in seconds (s)
β_k , β	Elapsed time after cessation of sampling until the end of time interval k, time at which the counting interval ends in seconds (s)
D_k	Coefficients of vector **D**: Number of alpha disintegrations observed during the kth time interval (counting period α_k to β_k)
$a_{k,i}(\alpha_k,\beta_k)$	Coefficients of matrix **A**: ith coefficient of count determination k within the limits α_k and β_k
$N_i(t_s)$	Coefficients of vector **N**: the five unknown ^{222}Rn and ^{220}Rn short-lived decay products sampled and deposited onto the filter at time t_s
C_i	Coefficients of vector **C**: mean activity concentrations of the five short-lived decay products in the sampled air

$\mathbf{A}, \mathbf{A}^{\mathrm{T}}, \mathbf{C}, \mathbf{D}, \mathbf{N}, \mathbf{M}$	Matrices and vectors ($\mathbf{A}^{\mathrm{T}}$ is the transposed matrix of $\mathbf{A}$)
κ_k , $\eta_{k,l}$	Substitutions defined in the text
M	Output quantity of the linear model function, h
$h(X_1, \ldots, X_T)$	Linear model function with the input quantities $X_1, \ldots, X_T$
$X_1, \ldots, X_T$	Input quantities of the linear model function, h
$\hat{x}_1, \ldots, \hat{x}_T$	Best estimate of the input quantities
$u_{rel}(x)$	Relative standard uncertainty of a quantity x
$c_1, \ldots, c_T$	Sensitivity coefficient

The indices i refer to the following radionuclides:

index $i = 1$ refers to ^{218}Po

index $i = 2$ refers to ^{214}Pb

index $i = 3$ refers to ^{214}Bi/^{214}Po

index $i = 4$ refers to ^{212}Pb

index $i = 5$ refers to ^{212}Bi

index $i = 5'$ refers to ^{212}Po

3.2 Quantities and units

In this document, units of the International System (SI) are used[1]. The definitions of radiation quantities are given in IEC 60050-393 and IEC 60050-395. The corresponding old units (non SI) are indicated in brackets.

Multiples and submultiples of SI units will be used, when practicable, according to the SI system.

4 Radon in the environment

4.1 Origin, genesis and decay

The heavy metals uranium and thorium are natural components of the lithosphere. Both elements can be detected in different quantities in minerals, in soils and in water. The average concentration in the lithosphere for uranium is between 2,5 – 4 mg/kg and for thorium about 13 mg/kg [1][2]. Naturally occurring uranium is a mixture of three isotopes: 99,27 % ^{238}U, 0,72 % ^{235}U and 0,01 % ^{234}U. The primordial radionuclides ^{238}U and ^{232}Th are the mother nuclides of the decay chains by which ^{222}Rn and ^{220}Rn are formed, respectively.

The direct mother nuclide of ^{222}Rn is ^{226}Ra and of ^{220}Rn, it is ^{224}Ra. ^{226}Ra has formerly gained technical importance as luminescent paint for dials of watches and instruments. The alpha particles emitted by disintegration of radium excite a phosphor radiating luminescence light. ^{226}Ra was also applied as radiation source in medicine.

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1 International Bureau of Weights and Measures: The International System of Units, 8th edition, 2006.

2 Numbers in square brackets refer to the Bibliography.

In contradiction to all other radionuclides of the ^{238}U- and ^{232}Th-decay chains, radon isotopes are gaseous. Radon is soluble in water. Particularly, ^{222}Rn can be enriched in groundwater, if the aquifer layers contain elevated values of natural radioactivity (e.g. granite stone). A technical application of the radon isotopes is not known. In medicine, radon is used for the treatment of chronic diseases of the musculoskeletal system. As typical indication for a radon therapy, rheumatic diseases of the joints are often cited [2].

Elevated concentrations of radon have been mostly found at underground workplaces (mining), in radon spas or in water works. Elevated concentrations of ^{222}Rn in houses and other buildings can particularly occur, when a high concentration of ^{222}Rn in soil exists, and radon penetrates the house via entry paths in the below-ground structural elements. Elevated exposures to ^{220}Rn are mostly credited to thorium-containing building materials (e.g. limestone) [3].

The main source for the radiation effect is not attributed to the inhalation of radon itself but to the simultaneous inhalation of its short-lived decay products mostly attached to aerosol particles. The short-lived ^{222}Rn decay products are ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po and those of ^{220}Rn are ^{216}Po, ^{212}Pb, ^{212}Bi and ^{212}Po. The short-lived decay products are deposited in the respiratory tract and decay there. Alpha particles emitted by ^{218}Po and ^{214}Po, or ^{212}Bi and ^{212}Po respectively, transfer their energy along the penetration way to the radiation sensitive cells, which cause possible health effects. ^{216}Po is an exemption. Because of its short half-life, it disintegrates during inhalation, and does therefore not markedly contribute to the exposure in the lung.

Whilst the ^{220}Rn decay chain ends up with the stable lead isotope ^{208}Pb, the ^{222}Rn decay chain has further stages following the short-lived decay products: Headed by ^{210}Pb, ^{210}Bi and ^{210}Po follows until it ultimately ends up with the stable lead isotope ^{206}Pb. Because of the long half-life of ^{210}Pb of more than 20 years, the remaining radionuclides are moved out of the respiratory tract by lung clearance, being excreted or deposited mainly in the mineral component of the bones [4]. In view of radiation protection, the radiation effects of the long-lived radionuclides are not relevant.

4.2 Radon in the rocks and soils and its transport towards the atmosphere

In rocks and soils, the permanent generation of radon is performed by alpha decay of radium. Radon atoms are subject to various processes on their path from the generation up to the atmosphere.

The emanation is the discharge of radon from the solid, mostly crystalline, phase of rocks and soils into the free pore volume, micro cracks, and fissures of the subsoil. The quantity, which defines the ratio between the number of radon atoms escaped the solid phase and the total number of radon atoms created in the solid phase, is the radon emanation coefficient. The process of discharge is initiated by the recoil due to alpha decay. The efficiency of this process depends on the distribution of radium in the mineral grain. The main part of radon escapes from radium located on the surface of the mineral grain or in the vicinity of the surface with depths lower than the recoil distance. A discharge of radon inside the mineral grain is only possible if sufficient pathways inside the grain are available. Very important for the emanation is therefore the grain size distribution.

The presence of water can increase the radon discharge. Due to adsorption of kinetic energy, radon atoms continue to stay in the pore water, from which it can attain the air-filled pore volume by diffusion. Soils and rocks reach the maximum of the radon emanation at various moistures. With the pore volume increasingly filling up with water, the part in the gas phase is getting lower, and the connectivity of the pore volumes by menisci is disturbed. Between dry and moisturized, the emanation can vary by a factor of 5 [5].

The movement of radon through rocks and soils is named migration. It is subject to geo-mechanical and hydrological conditions in the subsoil (permeability, fissions, flow of ground water and soil air). The main transport process is the diffusion, which can be supplemented by convection as an additional process. The diffusion is the mass transport of radon through inter-granular volumes, capillaries and fine pores caused by gradients of concentration. The coefficient of diffusion is a measure for the displacement forced by the gradient of concentration. In Table A.6, the effective diffusion coefficients for different materials are given. The coefficients take into account the prolongation of diffusion pathway due to ramifications of pores conducting the gas around the solid particles (tortuosity).

By convection, radon is transported together with carrier media, like ground waters or soil gases. The convective radon transport can result in radon anomalies. Within the bedrock, fissions and faults channelize the movement of radon-containing gases or ground waters, leading to an inhomogeneous distribution of the radon concentration far away from the origin of radon atoms.

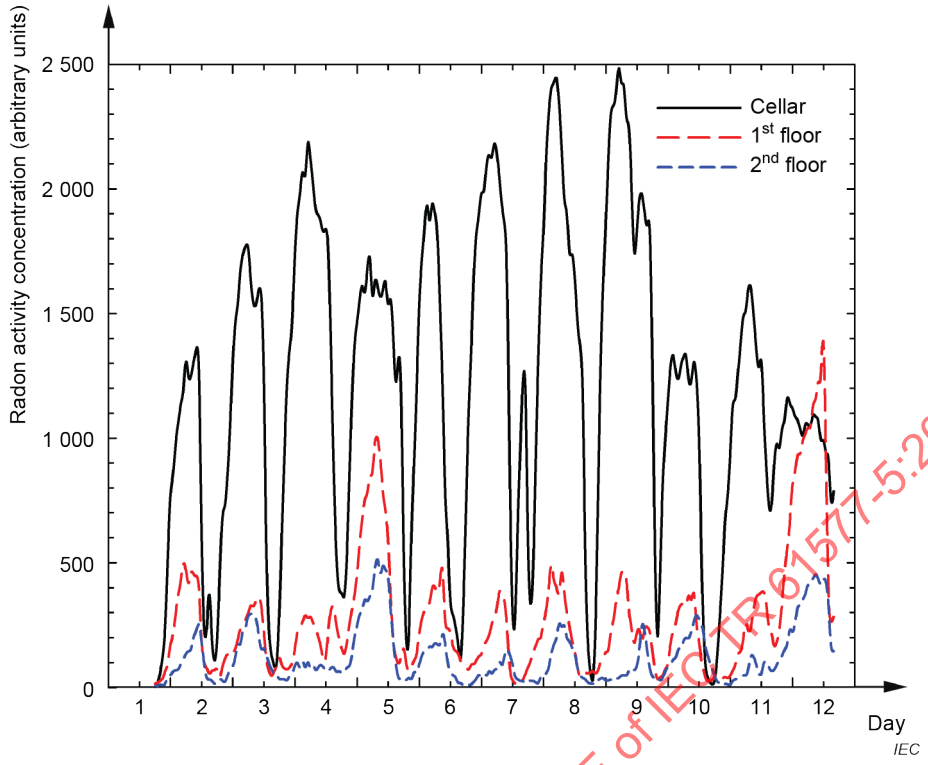
Besides the basic availability of radon by emanation and transport from the bedrock, the concentration of radon in the upper soil layers depends on the permeability, which in turn, depends on the pore size distribution and the degree of saturation with moisture, and can therefore locally and seasonally vary over several magnitudes. The discharge of radon from soil surface into air is denoted as exhalation.

4.3 Radon concentration in the outdoor air

Weather conditions with variations in air temperature from day to night cause variations in the outdoor radon concentration [6]. During the day the soil surface and the lower layers of the atmosphere heat up more intensively then the upper air layers by solar radiation, thus making the thermal stratification instable. The rising warm air results in a vertical intermixing of the atmosphere with low radon concentrations at the day. During the night and the early hours of the morning, the soil surface and the lower layers of the atmosphere cool down resulting in a stable stratification of the atmosphere (inversion at night). This process reduces the vertical intermixing of the atmosphere, which causes high radon concentration in the lower outdoor air layers. The variation of the radon concentration between day and night is greater, the greater the contrast in air temperature is. Measurements have shown that the day-to-night variations in the radon concentrations are mainly provoked by fluctuations of the vertical stratification in the atmosphere [7]. By contrast, the daily variations of the radon exhalation from the ground do not have substantial impact on the day-to-night variations in the outdoor radon concentrations [7].

4.4 Radon concentration in houses and at workplaces

Radon can enter a house through its substructure due to depressurization caused by the wind load or the temperature difference between indoor and outdoor. Indoor to outdoor temperature differences cause convective air flows, by which outdoor air flows into the building at the base



NOTE The maxima were measured in the early morning hours and the minima in the afternoon. The variations have a period of 24 h. The figure indicates the time lag of the variations and the reduced concentrations in the floors above the cellar [8].

Figure 1 – Diurnal variations of the radon activity concentration in the cellar, 1st and 2nd floor of a detached house measured over 12 days

and out of the building at the upper floors or the ceiling. The architecture of the house affects the distribution of radon throughout the building. In basements, radon-laden soil gas flows through cracks in the floor slab and walls, block wall cavities, plumbing connections, and sump wells. The radon concentration in the soil gas entering the house depends basically on the geology, the content of ^{226}Ra in the soil and its humidity. The transport of radon is enhanced when buildings are under significant negative pressure, particularly at floor level [6][7][9]. The indoor radon concentration undergoes diurnal and seasonal periods [10][11][12][13][14]. The long-term trends resulting from the use of the building or the relevant room(s) as well as from meteorological conditions are superimposed by stochastic fluctuations. Figure 1 represents an example of the diurnal variations of the radon activity concentration in the cellar, 1st and 2nd floor of a detached house measured over 12 days.

5 Radon decay products in the atmosphere

5.1 Physical processes of decay products in gaseous media

After the formation of radon decay products, they are subject to various physical and chemical processes. The processes are described here on the example of ^{218}Po, which has been investigated extensively [15][16][17]. Measurements have shown that after formation more than 10 % to – 60 % of ^{218}Po is positively charged. Different concurrent processes act on the fresh generated decay products: cluster formation, neutralization as well as attachment to aerosols and on surfaces. Within a short time (<1 s) after formation, the radionuclides attach to vapors (predominately H_2O) and to other trace gas molecules in the atmosphere. The thus generated radioactive clusters are denoted as the unattached fraction of the radon decay products, and have diameters between 0,5 nm and 3 nm [18].

Because of their small particle size and their high diffusion coefficient between 0,005 $cm^2 \cdot s^{-1}$ to 0,100 $cm^2 \cdot s^{-1}$ [18], the clusters possess a high mobility. Thus making the clusters almost independent to other transport processes in the air, as turbulence, convection, sedimentation and distraction by electromagnetic fields. A radioactive aerosol is formed by attaching the radioactive clusters to the existing aerosol particles in air. This process takes 1 s to 100 s [19]. The ratio of potential alpha energy of the short-lived radon progeny, which are unattached to aerosol particles, to the potential alpha energy of the attached short-lived radon progeny depends mainly on the aerosol particle concentration, and varies between 0,03 and 0,2 for dwellings [19]. The ratio decreases to below 0,01 in mines due to the increasing particle concentration [20].

5.2 Aerosol characteristics and ventilation

Atmospheric aerosol particle size distributions consist basically of three separate modes [21][22]:

a) the nucleation (or nuclei) mode for particles with diameters smaller than 100 nm and a modal peak in the range between 10 nm and 30 nm range,

b) the accumulation mode for particles with diameters between about 100 nm to about 1 µm and a modal peak at about 300 nm, and

c) the coarse mode corresponding to particles with diameters larger than 1 µm.

The nucleation mode appears if particles are freshly formed or emitted. This mode has a relatively short lifetime. By coagulation with other nuclei and accumulation mode particles, there sizes increase and end up in the next larger mode, the accumulation mode [21]. Several aerosol particle sources, such as cigarette smoke, gas stove, or candles, affect the particle distribution significantly according to the properties of the particles emitted. 10 % to 20 % of the attached activity could be assigned to the nucleation mode between 10 nm and 100 nm [19][23]. Aerosol particles with diameters below 100 nm are also denoted as ultrafine particles.

The accumulation mode results largely from the condensation of water and other vapours, and the attachment of particles by coagulation. This mode is stable with respect to deposition, and has a relative long atmospheric residence time.

The coarse mode particles are usually mechanically formed, or are resuspended particles such as windblown dust. This mode appears mainly in the outdoor environment or at workplaces.

The aerosol particle size distributions at workplaces are influenced by the local ventilation conditions and possibly different aerosol sources. The aerosol particle concentration depends strongly on work activity. During the work activities in mines, the radioactive aerosol particles tend to smaller diameters caused by the large number of particles emitted by diesel engines. At underground workplaces (mines, show caves) the activity size distribution of attached radon progeny can be described by a unimodal lognormal distribution specified by the activity median aerodynamic diameter and the geometric standard deviation [20]. Measurements at aboveground workplaces have identified a tri-modal aerosol size distribution with the focus on the accumulation mode [23]. Depending on the work activities, the particle sources and the ventilation, the nucleation and the coarse mode are more or less distinct [23].

In a radon atmosphere, a mixture of gaseous radon and radon decay products attached or unattached to aerosol particles exists. But not all the decay products are available in the air volume. Because of particle deposition and adhesion, a part of them is deposited to other surfaces, as walls, floors or the possible inventory of the site (e.g. room). This part of radioactivity is not inhaled and does, therefore, not contribute to the radiation effects. The radon equilibrium factor expresses the disturbance of equilibrium between radon and its short-lived decay products (for definition see 7.1). In real atmospheres, the equilibrium factor is below 1. Indoor measurements have shown that the equilibrium factor varies within a 95 % confidence interval from 0,2 to 0,7 around the mean value of 0,4 [24][25].

6 Physical and chemical properties of radon and radon decay products

6.1 Physical and chemical properties

As a noble gas, radon is chemically broadly inert. In the presence of Fluor, radon is not volatile up to 230 °C, what can be caused by the formation of a radon fluoride. Radon is soluble in water and organic solvents [26][27]. Although colourless at standard temperature and pressure, on cooling below its freezing point of −71 °C radon emits a brilliant radio luminescence that turns from yellow to orange-red as the temperature lowers [28][29]. An activity concentration of ^{222}Rn of 100 Bq·m^{-3} is equivalent to a ^{222}Rn concentration of about $5 \cdot 10^7$ atoms per cubic metre. Taking into consideration that a gas contains more than 10^{25} atoms per cubic metre under standard conditions, in all cases where radon is present in the environment, it is a trace gas. Some physical and chemical properties of radon and its decay products are given in Table A.1.

Radon is readily absorbed on charcoal, silica gel and similar adsorbing substances. Radon can, therefore, be effectively removed from a sample air stream by collecting it on activated charcoal cooled to the temperature of solid carbon dioxide (-78,5 °C). Radon is desorbed from charcoal by heating to 350 °C [30].

6.2 Solubility of radon in liquids

Due to diffusion an exchange of gas molecules between the liquid and the gas volume exists at the interface between a gas and a liquid. The transfer into the liquid is proportional to the partial pressure of the gas. The discharge from the liquid is proportional to the concentration of the dissolved gas in the liquid. A dynamic equilibrium has established at saturation concentration. The solubility depends on the temperature of the liquid. When the temperature increases, the solubility decreases [31][32].

In literature the solubility of radon in a liquid is often described by the Ostwald coefficient. The Ostwald coefficient is defined as the volume of a gas dissolved at a given temperature and pressure divided by the volume of the solvent at the same temperature and pressure [33]. Because radon is a trace gas, whose dissolved fraction does not alter the volume of the solvent, the Ostwald coefficient L at a given temperature can be calculated by the ratio of the radon activity concentrations given by the formula:

$$L = \frac{C_{\mathrm{Rn,liquid}}}{C_{\mathrm{Rn}}}, \tag{1}$$

where the parameter $C_{\mathrm{Rn,liquid}}$ is the activity concentration of dissolved radon in the liquid and C_{Rn} is the activity concentration of radon in air.

If the solvent is water, the dependence of the Ostwald coefficient L with the temperature of water $T_{\mathrm{H_2O}}$ in Celsius (°C) can be expressed by [33]

$$L = 0{,}105 + 0{,}403 \cdot \exp\left(-0{,}0502 \cdot T_{\mathrm{H_2O}}\right), \tag{2}$$

The Ostwald coefficients for various organic solvents are given in Table A.5. The measurement of radon in water using the methods of emanometry, gamma spectrometry and liquid scintillation counting is described in the standard series ISO 13164 [33][34][35][36].

6.3 Radiological properties and radioactive equilibrium

In Table A.2 and Table A.3 the data for the natural decay chains of ^{238}U and ^{232}Th starting from ^{226}Ra and ^{224}Ra, respectively, are given [37]. The tables contain only the data which are relevant for radon measurements.

The decay products, which follow from the disintegration of ^{222}Rn or ^{220}Rn, are themselves radioactive. By sequential disintegration, the decay products of the respective next generation arise until the stable lead isotopes eventually terminate the decay chains. In a radioactive decay chain, the activity A_i of the i^{th} nuclide changes by disintegration and production. The activity $A_i(t)$ at time t is the difference of the production rate expressed by $q_i(t)$ and the disintegration rate expressed by $\lambda_i A_i(t)$,

$$\frac{dA_i(t)}{dt} = q_i(t) - \lambda_i A_i(t), \tag{3}$$

where

λ_i indicates the decay constant of the i^{th} nuclide.

A decay chain with n-nuclides, including mother nuclide, is described by an n-dimensional differential equation system, where each of the equations is of this kind. The single formulas are coupled via the production rate $q_i(t)$.

The general solution of the differential formula (3) is

$$A_i(t) = \left[A_i(0) + \int_0^t q_i(t') e^{\lambda_i t'} dt' \right] e^{-\lambda_i t}, \tag{4}$$

a) The running equilibrium

The activity of the mother nuclide ^{222}Rn is preset by a single supply at time $t = 0$, and disintegrates with the decay constant λ_{Rn-222}. The initial activity of all other nuclides in the decay chain is $A_i(0) = 0$.

In the case of the decay chain of ^{222}Rn, the mother nuclide ^{222}Rn is long-living compared to the short-lived decay products ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po, which is indicated by $\lambda_{Rn-222} << \lambda_{Po-218}, \lambda_{Pb-214}, \lambda_{Bi-214}, \lambda_{Po-214}$. Under this condition, the radioactive decay achieves asymptotically a state, which is called running equilibrium (Figure 2).

In a running equilibrium, the activity of the decay products disintegrates with the half-life of the mother nuclide ^{222}Rn,

$$A_i(t) = A_{Rn-222}(t)\, e^{-\lambda_{Rn-222} t} \prod_{k=1}^{i} \frac{\lambda_{i-k+1}}{\lambda_{i-k+1} - \lambda_{Rn-222}}, \tag{5}$$

where the indices $i-k+1, i=1$ represent ^{218}Po, $i-k+1, i=2$ ^{214}Pb, $i-k+1, i=3$ ^{214}Bi, and $i-k+1, i=4$ ^{214}Po.

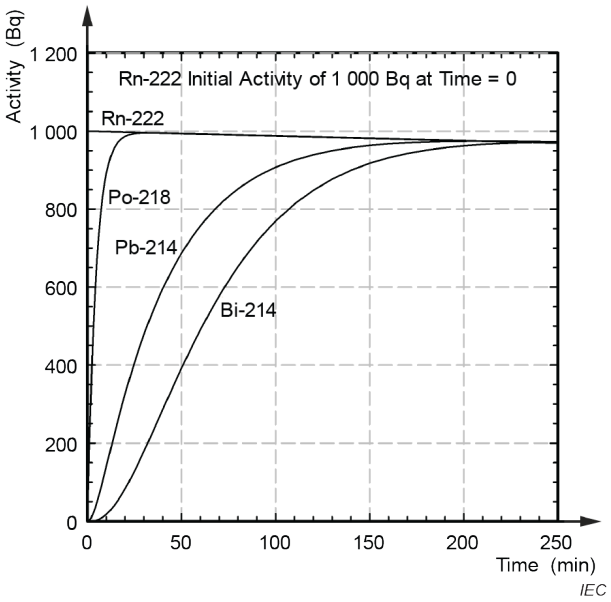


Figure 2 – Decay of ^{222}Rn after injection of 1 000 Bq at the start time and generation of decay products

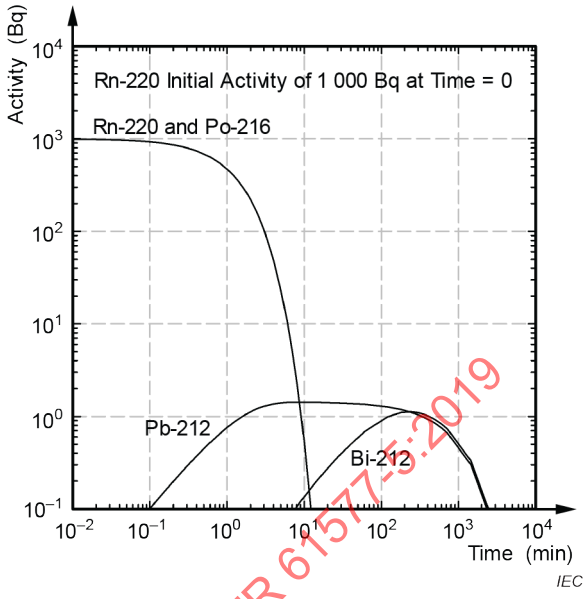


Figure 3 – Decay of ^{220}Rn (Thoron) after injection of 1 000 Bq at the start time and generation of decay products

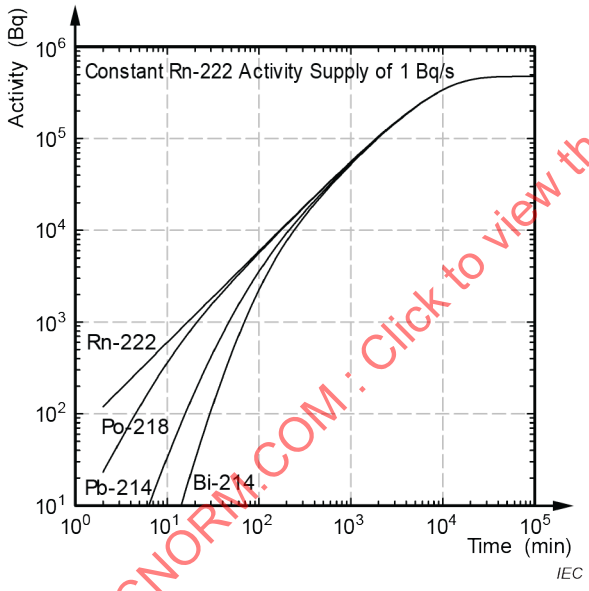


Figure 4 – Activity build-up of ^{222}Rn and its decay products for a continuous supply of ^{222}Rn with a rate of 1 Bq/s (in the absence of initial activities)

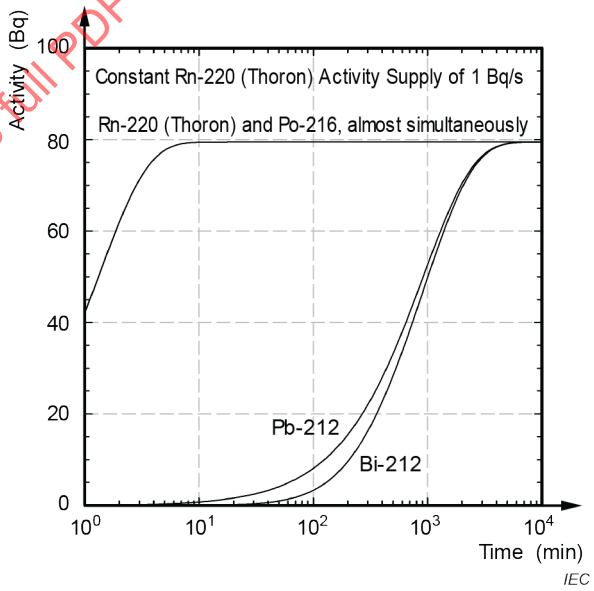


Figure 5 – Activity build-up of ^{220}Rn (Thoron) and its decay products for a continuous supply of ^{220}Rn with a rate of 1 Bq/s (in the absence of initial activities)

In the following, the very common cases shall be considered, from which general characteristics of the radioactive decay chains of ^{222}Rn and ^{220}Rn can be derived.

Since in the equilibrium state the product of coefficients $\lambda_{i-k+1} / (\lambda_{i-k+1} - \lambda_{Rn-222}) > 1$, the activity of the decay product is by this amount larger than the activity of the mother nuclide ^{222}Rn. Figure 2 shows the decay of ^{222}Rn after injection of 1 000 Bq at time $t = 0$, and the generation of the decay products. The running equilibrium has established after approximately 4 h.

Figure 3 shows the situation for ^{220}Rn (Thoron) after injection of the same activity. Since $\lambda_{Rn-220} >> \lambda_{Pb-212}, \lambda_{Bi-212}$, the condition for establishing the running equilibrium is not satisfied. The activity of the decay products achieves its maximum, if the mother nuclide ^{220}Rn has already decayed.

b) The secular equilibrium

The decay product is generated with a constant production rate, $q_i(t) \propto q_{Rn-222} = const$. This is the case, if a very long-lived mother nuclide disintegrates into a short-lived decay product. Asymptotically ($t \to \infty$), a secular equilibrium will establish, where the activities of all radionuclides are equal. For the radon nuclides (^{222}Rn and ^{220}Rn), this can be achieved by a permanent production from a radium or a thorium source. Figure 4 and Figure 5 show the build-up of the secular equilibriums for a constant supply of an activity rate of 1 Bq/s ^{222}Rn or ^{220}Rn, respectively. In the case of ^{222}Rn, the equilibrium between ^{222}Rn and its short-lived decay products is achieved after 1 day (1 440 min). Notwithstanding of that, the activity further increases until the secular equilibrium with the supplied activity rate has established. This is the case after about 28 days (40 000 min). In the case of ^{220}Rn, the secular equilibrium in the entire system has established after about 4 days (6 000 min).

6.4 Interaction of alpha particles with matter and energy deposition

^{222}Rn, ^{220}Rn and their decay products emit alpha and beta particles as well as gamma radiation (see Table A.2 and Table A.3), which interact with matter and deposit energy. The interaction with matter is exploited for radiation measurements. In bio-molecular systems, it can cause modifications in the genetic material that triggers health effects. From the dosimetric point of view, however, alpha particles play the decisive role, since these particles deposit the most part of energy to the radiation sensitive cells of the respiratory tract.

When alpha particles penetrate a medium, they undergo interactions with atoms of the media. The predominant interaction is the inelastic collisions with electrons resulting in excitation and ionization. Only at very low energies (below 150 keV for alpha particles) the elastic Coulomb collisions is important, in which recoil energy is imparted to atoms [38].

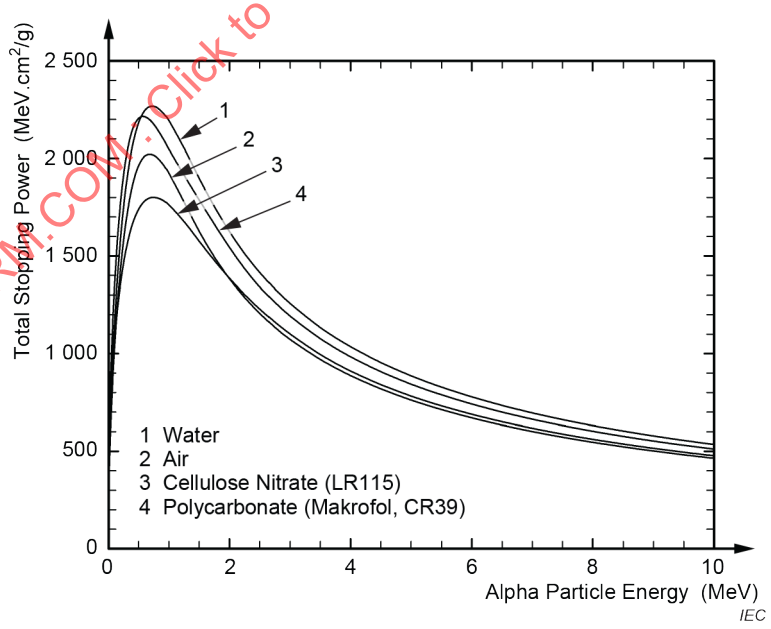


Figure 6 – Total stopping power of alpha particles penetrating different materials, the graphs use data from [38]

The stopping power of material is defined as being the average energy loss per unit path length, to which charged particles are subject when traversing the material. The functions of the total mass stopping power versus alpha energy and penetration depth in different materials are shown in Figure 6.

The ranges of the alpha particles emitted by radon and radon decay products in different materials are given in Table A.4. The ranges are calculated using the continuous-slowing-down-approximation (CSDA), by which energy-loss fluctuations are neglected. It is assumed that alpha particles lose their energy continuously along their pathway through the matter.

7 Measurement of ^{222}Rn and ^{220}Rn and their decay products

7.1 Relevant measurement quantities and units[3]

7.1.1 Activity concentration (C)

Activity A per unit volume V of the respective radon gas.

$$C_{Rn} = \frac{A}{V} \tag{6}$$

NOTE The index has to be used to state either ^{222}Rn or ^{220}Rn. To simplify the formulas here, instead of both isotopes the abbreviation Rn is used in the index.

This quantity is expressed in the SI unit (Bq·m^{-3}).

7.1.2 Equilibrium equivalent activity concentration (EEC, C_{eq})

The activity concentration of radon, C_{eq}, in radioactive equilibrium with its short-lived decay products that has the same potential alpha energy concentration, C_p, as the non-equilibrium mixture to which the C_{eq} refers:

$$C_{eq,Rn-222} = k_{Po-218}\, C_{Po-218} + k_{Pb-214}\, C_{Pb-214} + k_{Bi-214}\, C_{Bi-214} + k_{Po-214}\, C_{Po-214} \tag{7}$$

$$C_{eq,Rn-220} = k_{Po-216}\, C_{Po-216} + k_{Pb-212}\, C_{Pb-212} + k_{Bi-212}\, C_{Bi-212} + k_{Po-212}\, C_{Po-212} \tag{8}$$

The weighting coefficients k are calculated by nuclear data and given in Table 1.

Since $k_{Po-214} << 1$, $k_{Po-216} << 1$ and $k_{Po-212} << 1$, the corresponding activity concentration can be omitted.

$$C_{eq,Rn-222} = k_{Po-218}\, C_{Po-218} + k_{Pb-214}\, C_{Pb-214} + k_{Bi-214}\, C_{Bi-214} \tag{9}$$

$$C_{eq,Rn-220} = k_{Pb-212}\, C_{Pb-212} + k_{Bi-212}\, C_{Bi-212} \tag{10}$$

[3] The quantities and units relevant for the use of the respective standard are part of this. This subclause contains only the important metrological quantities and units, which are here extensively described. Definitions and values have been taken from ICRU Report 88 [8].

Table 1 – Coefficients for the calculation of the equilibrium equivalent concentration from measured activity concentrations of radon progeny

$k_{Po\text{-}218}$	$u(k_{Po\text{-}218})$	$k_{Pb\text{-}214}$	$u(k_{Pb\text{-}214})$	$k_{Bi\text{-}214}$	$u(k_{Bi\text{-}214})$	$k_{Po\text{-}214}$	$u(k_{Po\text{-}214})$
0,106	0,002	0,513	0,010	0,381	0,009	$5{,}2\cdot10^{-8}$	$1\cdot10^{-9}$
$k_{Po\text{-}216}$	$u(k_{Po\text{-}216})$	$k_{Pb\text{-}212}$	$u(k_{Pb\text{-}212})$	$k_{Bi\text{-}212}$	$u(k_{Bi\text{-}212})$	$k_{Po\text{-}212}$	$u(k_{Po\text{-}212})$
$6{,}684\cdot10^{-6}$	$0{,}223\cdot10^{-6}$	0,9133	0,0001	0,0866	0,0001	$8{,}05\cdot10^{-12}$	$6\cdot10^{-14}$

Calculations according to GUM, CODATA recommended values of fundamental physical constants, nuclear data taken from BIPM-5 Vol. 4, A=133 to 252.

This quantity is expressed in the SI unit $Bq\cdot m^{-3}$.

NOTE 1 For ^{222}Rn the following conversion is valid:

$$C_{eq} = C_p / \left[5{,}57(10)\cdot10^{-9}\,J\cdot Bq^{-1}\right] \tag{11}$$

$$C_{eq} = C_p / \left[3{,}47(7)\cdot10^{10}\,eV\cdot Bq^{-1}\right] \tag{12}$$

NOTE 2 For ^{220}Rn the following conversion is valid:

$$C_{eq} = C_p / \left[7{,}565(8)\cdot10^{-8}\,J\cdot Bq^{-1}\right] \tag{13}$$

$$C_{eq} = C_p / \left[4{,}722(5)\cdot10^{11}\,eV\cdot Bq^{-1}\right] \tag{14}$$

7.1.3 Equilibrium factor (F)

The equilibrium factor is the ratio of the equilibrium equivalent concentration, C_{eq}, and the radon activity concentration, C_{Rn}.

$$F = \frac{C_{eq}}{C_{Rn}} \tag{15}$$

NOTE In the case of ^{220}Rn, the relatively long half-life on ^{212}Pb may lead to cases where ^{220}Rn completely disappears before ^{212}Pb grows in; in this case the quantity is not defined.

7.1.4 Exposure to radon (P_{Rn})

The time-integral over the activity concentration during a defined period of time.

$$P_{Rn}(C, \Delta t) = \int_{\Delta t} C_{Rn}\cdot dt \tag{16}$$

This quantity is expressed in the SI unit $(Bq\cdot h\cdot m^{-3})$.

7.1.5 Potential alpha energy (ε_p)

The potential alpha energy, ε_p, is the total alpha energy emitted during the decay of a progeny atom along the decay chain up to ^{210}Pb or ^{208}Pb, respectively, for the decay chains of the ^{222}Rn and ^{220}Rn.

$$\varepsilon_{p,Rn-222} = \varepsilon_{p,Po-218} \cdot N_{Po-218} + \varepsilon_{p,Po-214} \cdot \left(N_{Pb-214} + N_{Bi-214} + N_{Po-214}\right) \tag{17}$$

$$\varepsilon_{p,Rn-220} = \varepsilon_{p,Po-216} \cdot N_{Po-216} + \varepsilon_{p,Pb-212} \cdot \left(N_{Pb-212} + N_{Bi-212}\right) + \varepsilon_{p,Po-212} \cdot N_{Po-212} \tag{18}$$

N is the number of the respective atoms. The potential alpha energy is a quantity for characterizing radon progeny atmospheres, not radon atmospheres. This quantity is expressed in the SI unit J. The values of the potential alpha energy for the radon and thoron progenies are given in Tables 2 and 3.

7.1.6 Potential alpha energy concentration (C_p)

The concentration of any mixture of short-lived radon decay products in air in terms of the alpha energy released during complete decay through ^{210}Pb for ^{222}Rn progeny or through ^{208}Pb for ^{220}Rn progeny.

$$C_{p,Rn-222} = \frac{C_{Po-218}}{\lambda_{Po-218}}\ \varepsilon_{p,Po-218} + \left(\frac{C_{Pb-214}}{\lambda_{Pb-214}} + \frac{C_{Bi-214}}{\lambda_{Bi-214}} + \frac{C_{Po-214}}{\lambda_{Po-214}}\right)\varepsilon_{p,Po-214} \tag{19}$$

$$C_{p,Rn-220} = \frac{C_{Po-216}}{\lambda_{Po-216}}\ \varepsilon_{p,Po-216} + \left(\frac{C_{Pb-212}}{\lambda_{Pb-212}} + \frac{C_{Bi-212}}{\lambda_{Bi-212}}\right)\varepsilon_{p,Pb-212} + \frac{C_{Po-212}}{\lambda_{Po-212}}\ \varepsilon_{p,Po-212} \tag{20}$$

The parameter C is the activity concentration of the respective progeny. This quantity is expressed in the SI unit J·m^{-3}.

NOTE ^{222}Rn: Due to the short half-lives of ^{214}Po, the isotope is in activity equilibrium with its parent nuclide: $C_{\text{Bi-214}} = C_{\text{Po-214}}$. ^{220}Rn: Due to the short half-lives of ^{216}Po and ^{212}Po, these isotopes are in activity equilibrium with their parent nuclide: $C_{\text{Rn-220}} = C_{\text{Po-216}}$ and $0{,}64\ C_{\text{Bi-212}} = C_{\text{Po-212}}$ with the transition probability of 0,64 for the decay branch $^{212}Bi \rightarrow {}^{212}Po$.

Table 2 – Potential alpha energy per atom for ^{222}Rn progeny including standard uncertainty

Potential alpha-energy	Standard uncertainty (k=1)	Potential alpha-energy	Standard uncertainty (k=1)	Potential alpha-energy	Standard uncertainty (k=1)	Potential alpha-energy	Standard uncertainty (k=1)
ε_p (Po-218)	u(ε_p(Po-218))	ε_p(Pb-214)	u(ε_p(Pb-214))	ε_p(Bi-214)	u(ε_p(Bi-214))	ε_p(Po-214)	u(ε_p(Po-214))
13688,9 keV	0,6 keV	7687,9 keV	0,5 keV	7687,9 keV	0,5 keV	7686,7 keV	0,5 keV
$2{,}19321 \cdot 10^{-12}$ J	$0{,}00009 \cdot 10^{-12}$ J	$1{,}23174 \cdot 10^{-12}$ J	$0{,}00008 \cdot 10^{-12}$ J	$1{,}23174 \cdot 10^{-12}$ J	$0{,}00008 \cdot 10^{-12}$ J	$1{,}23155 \cdot 10^{-12}$ J	$0{,}00008 \cdot 10^{-12}$ J

Table 3 – Potential alpha energy per atom for ^{220}Rn progeny including standard uncertainty

Potential alpha-energy	Standard uncertainty (k=1)	Potential alpha-energy	Standard uncertainty (k=1)	Potential alpha-energy	Standard uncertainty (k=1)	Potential alpha-energy	Standard uncertainty (k=1)
ε_p (Po-216)	u(ε_p(Po-216))	ε_p(Pb-212)	u(ε_p(Pb-212))	ε_p(Bi-212)	u(ε_p(Bi-212))	ε_p(Po-212)	u(ε_p(Po-212))
14582,7 keV	5,1 keV	7804,2 keV	5,1 keV	7804,2 keV	5,1 keV	8785,2 keV	4,4 keV
$2{,}33641 \cdot 10^{-12}$ J	$0{,}00081 \cdot 10^{-12}$ J	$1{,}25036 \cdot 10^{-12}$ J	$0{,}00081 \cdot 10^{-12}$ J	$1{,}25036 \cdot 10^{-12}$ J	$0{,}00081 \cdot 10^{-12}$ J	$1{,}40754 \cdot 10^{-12}$ J	$0{,}00071 \cdot 10^{-12}$J

7.1.7 Potential alpha energy exposure (P_p)

The time integral of the potential alpha energy concentration in air, C_p, over a given time period Δt.

$$P_p = \int_{\Delta t} C_p(t)\,dt \tag{21}$$

This quantity is expressed in the SI unit J·h·m^{-3}.

7.1.8 The unattached and attached fraction of potential alpha energy concentration

A fraction of progeny may not become attached to airborne particles and this quantity is often referred to as the free or unattached fraction. The unattached fraction is defined as the fraction of the potential alpha energy concentration of short-lived radon progeny that is not attached to the ambient aerosol.

The attached fraction of the potential alpha energy concentration of short-lived radon progeny is attached to the ambient aerosol.

NOTE 1 The particle size concerned is in the order of magnitude of nanometer. It is proposed to use a particle diameter of 5 nm as an upper limit for the unattached progeny (i.e., cluster carrying progeny).

NOTE 2 The attached progeny may have a tri-modal activity size distribution, which can be approximated by a combination of three lognormal distributions. These consist of the nucleation mode with activity median diameters (AMD) between 10 nm and 100 nm, the accumulation mode with AMD of 100 nm – 450 nm, and a coarse mode with an AMD > 1 µm. Generally the greatest fraction of the potential alpha energy is in the accumulation mode.

NOTE 3 A diameter of 5 nm is proposed as the lower limit for the attached progeny (i.e., aerosol carrying progeny).

NOTE 4 The sum of the attached and the unattached fraction is equal to 1.

7.2 Instruments measuring airborne radon activity concentration

An overview on commonly used types of instrument is outlined in Figure A.1. All the measurements base on the principle that radon enters the sensitive volume of the instrument or is collected by appropriate absorbents. The sensitive volume is generally separated from the surrounding air by a case including filters, which ensures that only radon atoms can get inside. Radon decay products, which always exist in the surrounding air, remain outside. Inside the sensitive volume or the adsorbents, radon atoms decay along the decay chain. The thus emitted radiation is analysed by a radiation detector.

Commonly used instruments differ by means of their sampling and measurement methods. The sampling can be passive or active. The latter actively conveys air into the sensitive volume using a pump. Passive sampling utilizes either adsorption onto activated charcoal or the diffusion of radon through a diffusion barrier (aerosol filter). ^{222}Rn and ^{220}Rn collected inside the sensitive volume disintegrate, and form the decay products consecutively. By applying an appropriate measurement method, the emitted gammas or alphas (in some cases also betas) are measured via direct generation of free charge carriers in the detector, or via secondary effects of the interaction of radiation with the detector (e.g. scintillation light emission, creating latent tracks in solid state polymer detectors).

Passive or active sampling in combination with their respective measurement methods can be used for different measurement tasks [39][40][41]:

a) Instrument with grab or continuous sampling: When a grab sampling is used, an air sample is taken and analysed immediately thereafter. Grab sampling is used to analyse the value of the radon activity concentration in the air at the moment of sampling.

In the case of continuous sampling, the sensitive volume is permanently penetrated by radon. For that purpose, radon containing air enters the sensitive volume either by establishing a continuous air flow using a pump (active sampling), or by diffusion through an aerosol filter (passive sampling). Radon decay products containing in the surrounding air are impeded by an aerosol filter from penetrating the sensitive volume. The continuous sampling enables to acquire the radon activity concentration in the course of time, and to determine its fluctuations and trends. Ionization chambers, scintillation detectors and semiconductor detectors are commonly used for the measurements.

b) Instrument with long-term passive sampling: The sensitive volume (sensor) is deposed from the analysis unit. It contains a detector, which accumulates and stores the radiation effects over a long time. This is called integrating measurement. The integration over a long period of time has the advantage to average out short-term fluctuations. After the exposure, the detector is analysed using separate equipment. The accumulated radiation effects are a measure for the exposure to radon. The average radon activity concentration can be determined by dividing the exposure to radon by the time duration of exposure.

c) Instrument with short-term passive sampling using adsorption: A canister containing an adsorbent (e.g. activated charcoal) is exposed bare to air and collects radon by means of adsorption. The duration of the exposure lasts 2 to 3 days. After the exposure, the activity of the adsorbent is a measure for the average radon activity concentration.

Generally, the sampling and the measurements do not differ between ^{222}Rn and ^{220}Rn. Since both occur in conjunction very often, it can be, however, important to discriminate between both. The very different half-lives of both radionuclides are being exploited hereto. A well-dimensioned diffusion barrier extends the diffusion time into the instrument's sensitive measurement volume. Because of its relative short half-live of below 1 min, the major part of ^{220}Rn disintegrates before it can contribute to the measurement effect. In ideal case ^{222}Rn is measured only. A double chamber system, for example, with and without an extended diffusion barrier makes available the measurement of each of both radon nuclides [42]. Other design solutions exploiting the different half-lives are possible. A discrimination between ^{222}Rn and ^{220}Rn can also be achieved by spectrometric measurements of gammas and alphas emitted by the decay products formed in the sensitive volume.

7.3 Measurement of radon decay products

7.3.1 General overview of instruments

In practice, all types of instrument measuring radon decay products use aerosol filters and a pump, in order to suck the air to be investigated through the filter (Figure A.2). The total activity or the nuclide specific activity of the aerosol particles deposited onto the filter is determined during or after the sampling process using detectors for alpha-, beta- or gamma radiation. The activity concentration of each of the short-lived radon decay products and/or the potential alpha energy concentration is obtained from the activity of the filter and the corresponding air volume having been sucked through the filter (Annex B) [43][44].

This measurement principle can be varied in a wide range by changing the type of filter, the air volume rate, the radiation detector, as well as the time schedule of sampling and measurement. Most instruments distinguish by their sampling principle: continuous or grab air sampling.

Some commercial instruments measure attached and unattached fraction of the short-lived radon decay products. In atmospheres with low concentration of aerosol particles, the unattached fraction of the short-lived radon decay products becomes a non-negligible part of the measurement. Because of the high diffusion velocity, unattached particles increasingly deposit on surfaces, and go lost from the air to be measured. When this is not taken into consideration in designing the instrument, particularly the air inlet, it could affect the measurement results.

7.3.2 Sampling of the unattached radon decay products

The particular importance of the unattached fraction results from the high deposition rate in the respiratory tract [45], which is primarily caused by diffusion. Calculations using the ICRP lung model have shown that dose per exposure for the unattached radon decay products with diameters in the range of about 3 nm is a magnitude higher than for the attached decay products in the accumulation mode [8][45][46].

Wire screens are commonly used to estimate unattached radon decay products. As air flows through a wire screen, aerosol particles precipitate from the air stream and deposit onto the solid parts of the screen, while other particles pass the screen. The fundamentals of the deposition theory of aerosol particles were developed in [47][48][49][50]. A review of the theory can be found in [51][52][53][54]. The basic procedures of particle deposition are diffusion, inertial impaction, interception, gravitational settling and electrostatic depositions. It is assumed that a particle sticks if it contacts the fibre of the screen. That removes permanently the particle from the air stream. Figure 7 shows the contribution of the particular deposition processes to the total efficiency calculated exemplarily for a specific screen. For small particles in the nanometre range, the deposition by diffusion is the dominant process. Larger particles are deposited by interception, and with further increasing sizes, the impaction becomes the dominant deposition process.

The deposition efficiency of aerosol particles depends on the parameters of the wire screen, such as thickness, fibre diameter and solid fraction, as well as on the air flow velocity. The solid fraction, called also packing density or solidity, is defined as the ratio of the fibre volume to the total volume of the screen with values typically in the range between 0,01 and 0,3. The solidity is hydro-dynamically determined via the pressure drop across the screen [55]. For a given wire screen, the deposition efficiency varies with variation of the air flow velocity.

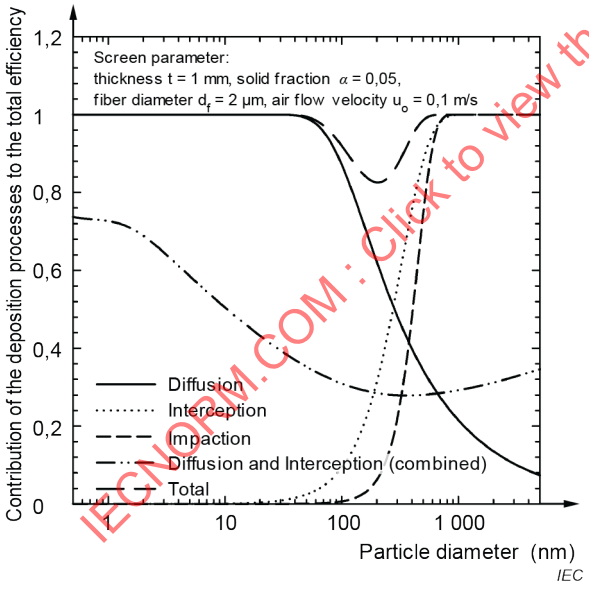


Figure 7 – Contributions of the deposition processes to the total efficiency (calculated exemplarily for a wire screen)

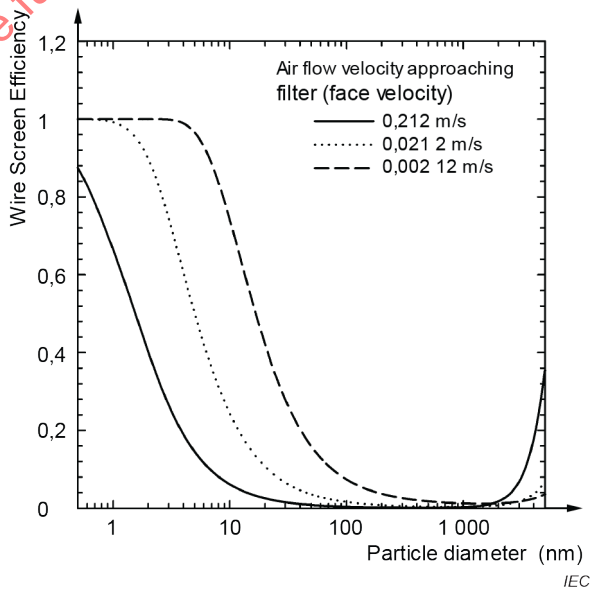


Figure 8 – Variation of deposition efficiency of a wire screen in dependence on air flow velocity (calculated exemplarily for a wire screen)

Figure 8 presents the efficiency curves for different air flow velocities exemplarily calculated for a wire screen. Due to the high deposition by diffusion, the efficiency of the screen is high for small particles. Already for low air flow velocities almost all particles with sizes below 10 nm are deposited. With increasing the air flow velocity, the efficiency curve moves towards lower particle sizes. Hence, only the deposition efficiency of particles with smaller sizes remains high, whilst larger particles that would have been deposited at lower air flows, pass the screen.

According to the definition of unattached fraction as particles in the ultrafine size range of less than 3 nm and considering the function of the deposition efficiency of wire screens for small particles, it is recommend to use wire screens with a deposition efficiency of 0,5 (50 %) for a particle size of 5 nm [18][56]. The appropriate dimensioning of screens including the air flow velocity shall be specified under exploiting the theory of wire screens.

7.3.3 Counting methods for the measurement of the activity concentrations and the potential alpha-energy concentration

7.3.3.1 Method of measurement

The measurement of activity caused by the decay products in air is important for the determination of the exposures to radon, in particular at workplaces, e.g. in mines.

The air of the atmosphere under investigation is carried through an aerosol filter during the sampling in order to deposit the suspended matter being contained in the air. Thereby, the short-lived decay products of ^{222}Rn and ^{220}Rn are simultaneously deposited onto the filter. By measuring the gross alpha counts after cessation of sampling, the quantity potential alpha energy concentration and/or the activity concentrations of each single short-lived decay product can be determined. The complete measurement is undertaken according to a predefined sampling and counting time scheme. The gross counting adds up all events caused by alpha particles above a threshold, without the need of detailed energy resolution. Such counting techniques inevitably discard some of the information carried by the radiation. The corresponding lower requirements on electronic stability in turn enhance the robustness against many environmental influences.

The counting background can generally be neglected for these measurement methods. Correction factors are needed in order to take account for the efficiency of the detector (counting efficiency) or the sampling efficiency. Both are determined by calibration.

Different time schemes were elaborated taking into consideration the decay and the build-up of radionuclides onto the filer during and after the sampling, and the concentrations of the decay products in the investigated air (air exchange rate, aerosol particle concentration, ratio of surface area to room volume). There are many counting methods proposed by various authors. Some of these require three [57][58][59][60][61][62] or two [63] counting periods in order to determine the activity of each of the short-lived decay products. Even methods for the determination of the potential alpha particle energy from a single count [64][65] were proposed.

Besides the systematic measurement errors resulting from various unknown mixtures of short-lived radon decay products presented in air, additional uncertainties occur. This is particularly the case if the air contains other radionuclides, which contribute to the measurement effect but without being considered in analyzing the results. This concerns long-lived radionuclides of the uranium and thorium decay chain, or ^{220}Rn decay products if only ^{222}Rn decay products are being considered or vice versa. Measurement uncertainty caused by self-absorption of alpha particles within the deposited matter can hardly be reasonably estimated under field conditions. These uncertainties can be averted by sufficiently short sampling periods and the usage of membrane filters inhibiting the penetration of particles into the filter.

All the following methods are primarily designed for the measurement of $RnDP_{222}$. A method that calculates the activity concentrations of ^{222}Rn and ^{220}Rn decay products by consecutive measurements of the activity of a filter after exposure and analytical deconvolution of the decay curve is described in addition.

7.3.3.2 Method of THOMAS

The method of THOMAS [57] is an advancement of the method of Tsivoglou [62] and allows the determination of the activity concentrations of ^{218}Po, ^{214}Pb, ^{214}Bi and the calculation of the corresponding potential alpha energy concentration.

The method involves three gross counts of events caused by alpha emission of radon decay products which have been sampled on a filter. After the sampling over a period of time of 300 s and a waiting time of 120 s, the first gross count is to be taken over a period of time of 180 s. The second counting period follows after waiting for further 60 s, and the third counting period starts 1 260 s (21 min) after the cessation of sampling. The complete time scheme is given in Table 4. The numbers of events registered by each of the gross counts are labelled by I_1, I_2 and I_3.

Table 4 – Time scheme for the method of Thomas [57]

Sampling	Waiting	First count	Waiting	Second count	Waiting	Third count
		I_1		I_2		I_3
300 s	120 s	180 s	60 s	840 s	60 s	540 s

The activity concentrations of ^{218}Po, C_{Po-218}, ^{214}Pb, C_{Pb-214}, and ^{214}Bi, C_{Bi-214}, collected onto the filter are given by the formulas:

$$C_{Po-218} = \frac{1}{\varepsilon \cdot v}\left(1{,}0418 \cdot 10^{-4} \cdot I_1 - 5{,}0567 \cdot 10^{-5} \cdot I_2 + 4{,}7810 \cdot 10^{-5} \cdot I_3\right) \quad (22)$$

$$C_{Pb-214} = \frac{1}{\varepsilon \cdot v}\left(7{,}5233 \cdot 10^{-7} \cdot I_1 - 1{,}2685 \cdot 10^{-5} \cdot I_2 + 3{,}0272 \cdot 10^{-5} \cdot I_3\right) \quad (23)$$

$$C_{Bi-214} = \frac{1}{\varepsilon \cdot v}\left(-1{,}3887 \cdot 10^{-5} \cdot I_1 + 2{,}0461 \cdot 10^{-5} \cdot I_2 - 2{,}3254 \cdot 10^{-5} \cdot I_3\right) \quad (24)$$

where

ε is the detection and sampling efficiency, and

v is the volume flow rate of air sampled in the unit cubic metre per second ($m^3 \cdot s^{-1}$).

The potential alpha energy concentration C_p in the unit $J \cdot m^{-3}$ (Joule per cubic metre) follows from:

$$C_p = \left(13{,}69 \cdot C_{Po-218} + 7{,}69 \cdot \left(C_{Pb-214} + C_{Bi-214}\right)\right) \cdot 1{,}60 \cdot 10^{-13} \quad (25)$$

THOMAS quotes measurement uncertainties of C_{Po-218} with about 12 %, and of C_{Pb214} and C_{Bi214} with about 4 %.

A further modification of this method was undertaken by Hartley [59].

7.3.3.3 Method of MARKOV

The method of MARKOV [63] is based on the simplified assumptions that in the first 10 min after the cessation of sampling, the increase of the activity of new generated ^{214}Pb and the decrease of the sampled activity of ^{214}Bi on the filter mutually compensate each other. The sampling and counting time scheme is given in Table 5.

Analogously to the method of THOMAS, two different count intervals are used for determining the activity concentrations of the short-lived radon decay products. Because this method achieved a particular importance for the determination of the potential alpha energy

concentration, only that formula is presented. For information about the determination of activity concentrations of the decay products, it is referred to literature.

Table 5 – Time scheme for the method of MARKOV [63]

Sampling	Waiting	First count	Waiting	Second count
		I_1		I_2
300 s	60 s	180 s	180 s	180 s

Investigations of atmospheres with different activity concentrations of short-lived decay products have demonstrated that the potential alpha energy concentration is proportional to the number I_2 of alpha events registered in the second gross count and can be calculated using the empirical formula:

$$C_p = e_m \frac{I_2}{\varepsilon \cdot v} \tag{26}$$

where

C_p is the potential alpha energy concentration in the micro joule per cubic metre ($\mu J \cdot m^{-3}$),

ε is the detection and sampling efficiency,

v is the volume flow rate of the air sampled in the unit cubic metre per second ($m^3 \cdot s^{-1}$), and

e_m is the coefficient of MARKOV given by $1{,}07 \cdot 10^{-7}$ $\mu J \cdot s^{-1}$.

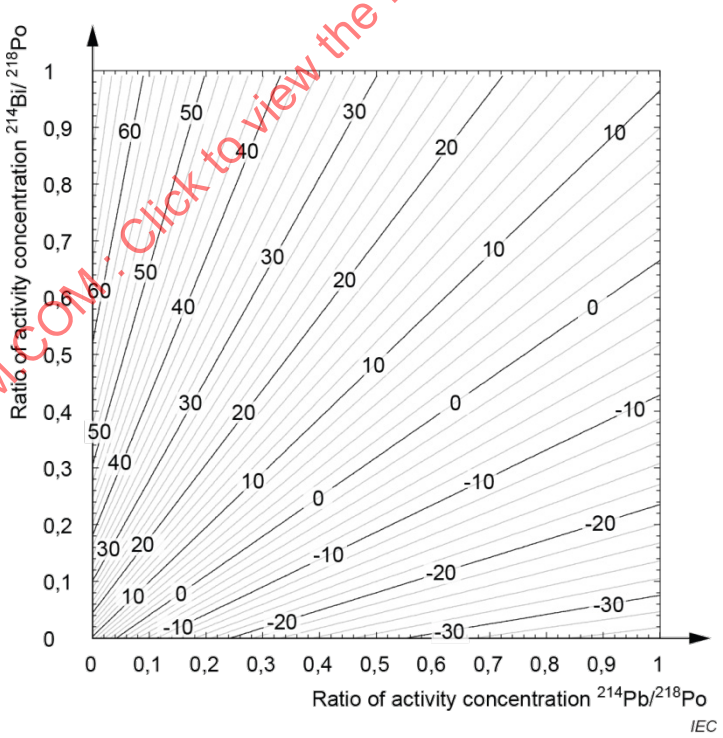


Figure 9 – Measurement error of the method of MARKOV given in percent for different ratios of decay products in the air sampled

Figure 9 shows the resulting measurement error in percent for different nuclide concentrations in the air sampled. In real atmospheres not every ratio of activity concentrations calculated in figure occur. The measurement errors in real atmospheres in homes or at workplaces are estimated to be in the range of 10 % to 15 %.

7.3.3.4 Method of multiple successive countings

The abovementioned methods are restrictive with respect to the number and duration of counting intervals. The method of multiple successive countings does not have such restrictions. The measurement procedure is delineated in Figure 10. After the cessation of sampling, the abatement of activity is registered by multiple countings. The countings can be taken to different times with periods over different durations. The counting periods can be partly overlapping, consecutive as shown in Figure 10 or with interruptions.

The number of each ^{222}Rn and ^{220}Rn short-lived decay product sampled and deposited onto the filter, and the activity concentration of the decay products in the air sampled is calculated using a least square regression analysis. In the first step, the numbers of the five unknown ^{222}Rn and ^{220}Rn decay products, $N_i(t_s)$, are determined from various count determinations, D_k, after cessation of sampling at time t_s (given in formula (B.23)). For that purpose, n count determinations within different time intervals (α_k,β_k) are to be taken.

The unknown variables $N_i(t_s)$ result from the minimum of the function f defined by:

$$\text{Min } f = \text{Min}\left(\sum_{k=1}^{n}\left(\sum_{i=1}^{5} a_{k,i}(\alpha_k,\beta_k)N_i(t_s) - D_k\right)^2\right) \quad (27)$$

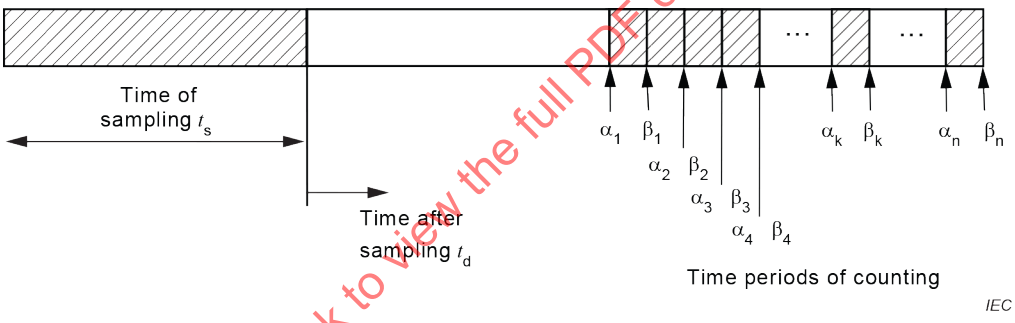


Figure 10 – Method of multiple successive countings

where

$a_{k,i}(\alpha_k,\beta_k)$ refers to the i^{th} coefficient for the count determination k within the limits α_k and β_k,

D_k is the gross number of alpha disintegrations on the filter measured during the count determination k within the limits α_k and β_k.

The minimization is accomplished by partial derivatives with respect to the unknown variables $N_i(t_s)$. It follows the normal formula written in matrix form as:

$$\mathbf{A}^T\mathbf{A}\,\mathbf{N} = \mathbf{A}^T\mathbf{D} \quad (28)$$

where

$$\mathbf{A} = \begin{vmatrix} a_{1,1}(\alpha_1,\beta_1) & a_{1,2}(\alpha_1,\beta_1) & a_{1,3}(\alpha_1,\beta_1) & a_{1,4}(\alpha_1,\beta_1) & a_{1,5}(\alpha_1,\beta_1) \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ a_{k,1}(\alpha_k,\beta_k) & a_{k,2}(\alpha_k,\beta_k) & a_{k,3}(\alpha_k,\beta_k) & a_{k,4}(\alpha_k,\beta_k) & a_{k,5}(\alpha_k,\beta_k) \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ a_{n,1}(\alpha_n,\beta_n) & a_{n,2}(\alpha_n,\beta_n) & a_{n,3}(\alpha_n,\beta_n) & a_{n,4}(\alpha_n,\beta_n) & a_{n,5}(\alpha_n,\beta_n) \end{vmatrix} \quad (29)$$

is the matrix of coefficients $a_{k,i}(\alpha_k,\beta_k)$. The vector of the gross counts of alpha disintegrations, D_k, measured at n count determinations, and the vector of the unknown number of atoms of the five short-lived decay products, $N_i(t_s)$, which are onto the filter at the time when the sampling has finished, are denoted by:

$$\mathbf{D} = \begin{vmatrix} D_1 \\ \vdots \\ \vdots \\ D_k \\ \vdots \\ D_n \end{vmatrix} \text{ and } \mathbf{N} = \begin{vmatrix} N_1(t_s) \\ N_2(t_s) \\ N_3(t_s) \\ N_4(t_s) \\ N_5(t_s) \end{vmatrix} \quad (30)$$

From formula (28), the number of atoms finally results from:

$$\mathbf{N} = \left(\mathbf{A}^T\mathbf{A}\right)^{-1}\mathbf{A}^T\mathbf{D} \quad (31)$$

Once the number of atoms $N_i(t_s)$ has been calculated, the purpose of the following calculations is the determination of the mean activity concentrations C_1 (^{218}Po), C_2 (^{214}Pb), C_3 (^{214}Bi), C_4 (^{212}Pb) and C_5 (^{212}Bi) in the air investigated during the sampling period. The mean activity concentrations C_i of the five short-lived decay products in air are calculated by the matrix formula:

$$\mathbf{C} = \frac{1}{\varepsilon_s v}\mathbf{M}\,\mathbf{N} \quad (32)$$

where

$$\mathbf{C} = \begin{vmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \\ C_5 \end{vmatrix} \quad (33)$$

represents the vector of mean activity concentrations C_i. The coefficients in the matrix $\mathbf{M}$ result from resolving the formulas (B.2), (B.4), (B.6), (B.8) and (B.10) for the corresponding C_i. The matrix $\mathbf{M}$ is given by:

$$\mathbf{M} = \begin{vmatrix} \lambda_1 \kappa_1 & 0 & 0 & 0 & 0 \\ -\lambda_2 \kappa_1 \left[1 - \dfrac{\lambda_2 \eta_{2,1}}{(\lambda_1 - \lambda_2)}\right] & \lambda_2 \kappa_2 & 0 & 0 & 0 \\ -\lambda_3 \kappa_1 \left[\begin{array}{l} \dfrac{\lambda_2 \eta_{2,1}}{(\lambda_1 - \lambda_2)} \\ + \dfrac{\lambda_2 \lambda_3 \eta_{3,2}(\eta_{2,1} + 1)}{(\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} \\ + \dfrac{\lambda_2 \lambda_3 \eta_{3,1}}{(\lambda_1 - \lambda_3)(\lambda_1 - \lambda_2)} \end{array}\right] & -\lambda_3 \kappa_2 \left[1 + \dfrac{\lambda_3 \eta_{3,2}}{(\lambda_3 - \lambda_2)}\right] & \lambda_3 \kappa_3 & 0 & 0 \\ 0 & 0 & 0 & \lambda_4 \kappa_4 & 0 \\ 0 & 0 & 0 & -\lambda_5 \kappa_4 \left[1 - \dfrac{\lambda_5 \eta_{5,4}}{(\lambda_4 - \lambda_5)}\right] & \lambda_5 \kappa_5 \end{vmatrix} \quad (34)$$

with the substitutions:

$$\eta_{k,l} = \frac{\left(e^{-\lambda_k t_s} - e^{-\lambda_l t_s}\right)}{\left(1 - e^{-\lambda_k t_s}\right)} \quad (35)$$

$$\kappa_k = \frac{\lambda_k}{\left(1 - e^{-\lambda_k t_s}\right)} \quad (36)$$

For the accurate determination of the radionuclides $N_i(t_s)$, the observations of alpha counts are made within the first 20 min after the sampling has ended. The sampled quantities of ^{218}Po, ^{214}Pb and ^{214}Bi as given in formula (31) make their greatest relative changes with respect to each other within the first 20 min. After 20 min has elapsed, the changes of activity of ^{218}Po and of ^{214}Pb are lower. Even the change of activity of ^{214}Bi is no longer as distinctly different as it is in the first 20 min.

The potential alpha energy concentration of ^{222}Rn short-lived decay products can be calculated from the activity concentrations C_1, C_2 and C_3, and that of ^{220}Rn short-lived decay products from the activity concentrations C_4 and C_5.

8 Quality assurance

8.1 Definition and purpose

Quality assurance includes all planned and systematic actions that are necessary to provide adequate confidence in the results of measurements.

Quality assurance shall include validation of methods and verification of results, which in turn involves all the actions by which the adequacy of equipment, instruments and procedures are assessed against specified requirements. It shall ensure that equipment and instruments function correctly, the procedures are correctly established and followed, quantifiable errors are within acceptable limits, and records are correctly and promptly maintained. The quality assurance programme and the regular checks made for quality control shall be fully documented [24][66].

General requirements for the competence of laboratories are set out by the international standard ISO/IEC 17025 [66], which shall be adopted to services carrying out radon measurements. The competence of a radon service can be formally recognized by an authorized body working under a national accreditation scheme.

8.2 Quality control

Radon service shall maintain an appropriate monitoring system to prove that specified requirements are kept and processes are under control. The modality of recording relevant process parameters shall ensure the repeatability of measurements.

From an analysis of the measurement process the relevant parameters should be deduced and monitored by control charts to reveal early trends and prevent malfunctions of procedures and equipment. A QA plan shall specify all activities, measurements and documentations to be carried out. The activities can include regular calibrations, cross-checks of measurement results, duplicates or collocated measurements, laboratory and field background measurements [67].

Through the provision of objective evidences that specified requirements have been fulfilled, quality control is an essential prerequisite for verification of processes and accomplish confidence in results.

8.3 Validation and traceability of measurements

8.3.1 Validation of methods

Validation is the confirmation that requirements for a specific intended use have been fulfilled. Standard validation procedures are type tests of instruments, interlaboratory comparisons and calibrations. The validation shall be executed as extensive as necessary to meet the needs for a given application.

8.3.2 Type test of radon instruments

By a type test, one or more radon instruments representative for the production are checked for compliance with requirements committed to the intended use. It covers mechanical, electrical and radiological examinations which can be carried out by the manufacturer himself or by an approved testing laboratory [68]. Specific requirements for radon measuring instruments are laid down in the standard series IEC 61577.

8.3.3 Interlaboratory comparison

An interlaboratory radon comparison includes the organisation, examination and evaluation of instruments exposed to radon. As a rule, comparisons are conducted by recognised reference laboratories. Interlaboratory radon comparisons contribute to ensure a uniform quality standard and will preferably be organized for passive radon instruments (nuclear track detectors, activated charcoal detectors, electrets). The comparisons are often used for the purposes of determining the performance and the surveillance of approved radon services.

Intercomparisons are designed for instruments using solid state nuclear track detectors, electrets or activated charcoal and run with similar procedures [69]: Radon services submit a sufficient number of instruments of the same type to the provider. Depending on the applied test scheme devices are randomized and grouped according to the number of exposures. The number of instruments to be submitted depends on the number of exposures and the need for additional transfer instruments being used for measurement effects during storage and delivery. After exposure, radon instruments are delivered to radon services in order to determine the exposures to radon and report the results to the provider of the intercomparison. The provider prepares a report with the measurements and reference data.

Radon services being interested in a radon intercomparison can get further information about providers and organisational conditions from the European Information System on Proficiency Testing Schemes (eptis) available via Internet.

8.3.4 Measurement traceability and calibration

A radon service shall ensure the traceability of its measurements to relevant primary standards by an unbroken chain of calibrations [70]. Resulting from calibration, the relationship between values indicated by the measuring instrument and the corresponding known values of activity concentration of radon is ascertained. The relationship is expressed as calibration factor or function. Calibrations should be regularly repeated after two years at least, but also after technical modifications or maintenance procedures, or when the instrument will be used under conditions not covered by previous calibrations.

The International Bureau of Weights and Measures (BIPM) maintains a database on Calibration and Measurement Capabilities (CMC) on its web page. The database provides information about national metrological institutes and their capability of performing metrological quantities. By international comparisons between national metrology institutes, the calibration and measurement certificates issued are mutually recognised.

Concerning radon measurements, some national metrological institutes offer gas standards [71]. These standards consist of glass ampoules or stainless steel containers containing a certified quantity of ^{222}Rn within a carrier gas. Owing to radioactive decay, the gas standard is to be used for calibration purposes without significant delay after assembling and delivery. As an alternative to radon gas standards, also solid radium standards with a constant certified emanation rate are possible [72].

Calibrations of radon instruments are carried out in reference atmospheres and should be undertaken by approved calibration laboratories. A reference atmosphere is a radioactive atmosphere with parameters being controllable during the calibration procedure. Reference atmospheres can be set up in small containers with several tens of litres up to chambers with volumes of several cubic metres. The quantity ^{222}Rn activity concentration is established by transferring ^{222}Rn from a certified standard into a well-known volume. In many cases radon monitors provided by national metrological institutes and calibrated against a radon primary standard are used for transferring radon quantities as well. A reference atmosphere of ^{222}Rn shall be free of ^{220}Rn (thoron), otherwise the activity concentration of ^{220}Rn shall be measured [73][74][75].

Besides the radon activity concentration, relevant parameters are aerosol particle concentration and size distribution as well as climatic parameters. The parameters of the reference atmosphere should be corresponded to the location at which the instrument is intended to be used (e.g. in houses). Special requirements on reference atmospheres are set out in IEC 61577-4.

9 Determination of the measurement uncertainty, detection threshold, detection limit

9.1 General

The detection threshold, detection limit and confidence intervals play an important role to prove the applicability for a specific purpose of measurement, and to ensure the quality of the measurement. On their basis the measurement results can be interpreted and validated.

In the following, guidelines for the application of the uncertainty analysis according to ISO/IEC Guide 98-3:2008 [76] are given. These guidelines shall appropriately be applied to radon and radon decay product measurements. The detection threshold and the detection limit are determined according to ISO 11929 [77]. Since there are different measurement methods, the uncertainty analysis will exemplarily be described by the application to selected measurements. The procedure can analogously be transposed to other measurement methods. Only the analytical method using a linear model function is presented here. More information, particularly the application of the Monte-Carlo-Method, is given in literature [76] and IEC TR 62461:2015.

9.2 Procedure for the determination

In this document the determination of the measurement uncertainty is carried out according to IEC TR 62461:2015. The following steps are to be executed sequentially:

a) A linear mathematical model function is to be set up describing the relation of the input quantities X_j and the output quantity M,

$$M = h(X_1, \ldots, X_T) \tag{37}$$

where

T is the number of input quantities;

X_j is an input quantity;

M is the output quantity.

The model function should contain every quantity, including all corrections and correction factors that can contribute a significant component of the uncertainty to the result of the measurement.

b) The available information for the input quantities X_j has to be collected.

c) The standard uncertainty of the output quantity has to be calculated. The usage of an appropriate computer program can facilitate the calculations.

1) For each input quantity, X_j with $j = 1 \ldots T$, the best estimate, $\hat{x}_j$, and its standard uncertainty, $u_{rel}(\hat{x}_j)$, have to be obtained.

2) The sensitivity coefficient, c_j, i.e. the partial derivate of the output quantity with respect to each input quantity, has to be calculated:

$$c_j = \frac{\partial h}{\partial x_j} \tag{38}$$

3) The uncertainty contribution to the output due to each input quantity has to be calculated by multiplying the sensitivity coefficient and the standard uncertainty:

$$u_j(\hat{m}) = |c_j| \cdot u_{rel}(\hat{x}_j) \tag{39}$$

4) The combined standard uncertainty for the output quantity is computed as the square root of the squared uncertainty contributions:

$$u_c(\hat{m}) = \sqrt{\sum_{j=1}^{T} \{u_j(\hat{m})\}^2} \ . \tag{40}$$

In the case that some input quantities are correlated with one other, i.e. they depend on each other, further terms need to be added to the sum under the square root sign.

5) The expanded uncertainty for the output quantity has to be calculated by multiplying the standard uncertainty with the appropriate coverage factor (usually $k = 2$):

$$U_c(\hat{m}) = 2\,u_c(\hat{m}) \tag{41}$$

If the probability distribution of the output quantity is not approximately Gaussian (or normal), the coverage factor may have another value.

An example of an uncertainty analysis for the method of multiple successive countings is given in Annex C.

Annex A
(informative)

Tables and figures

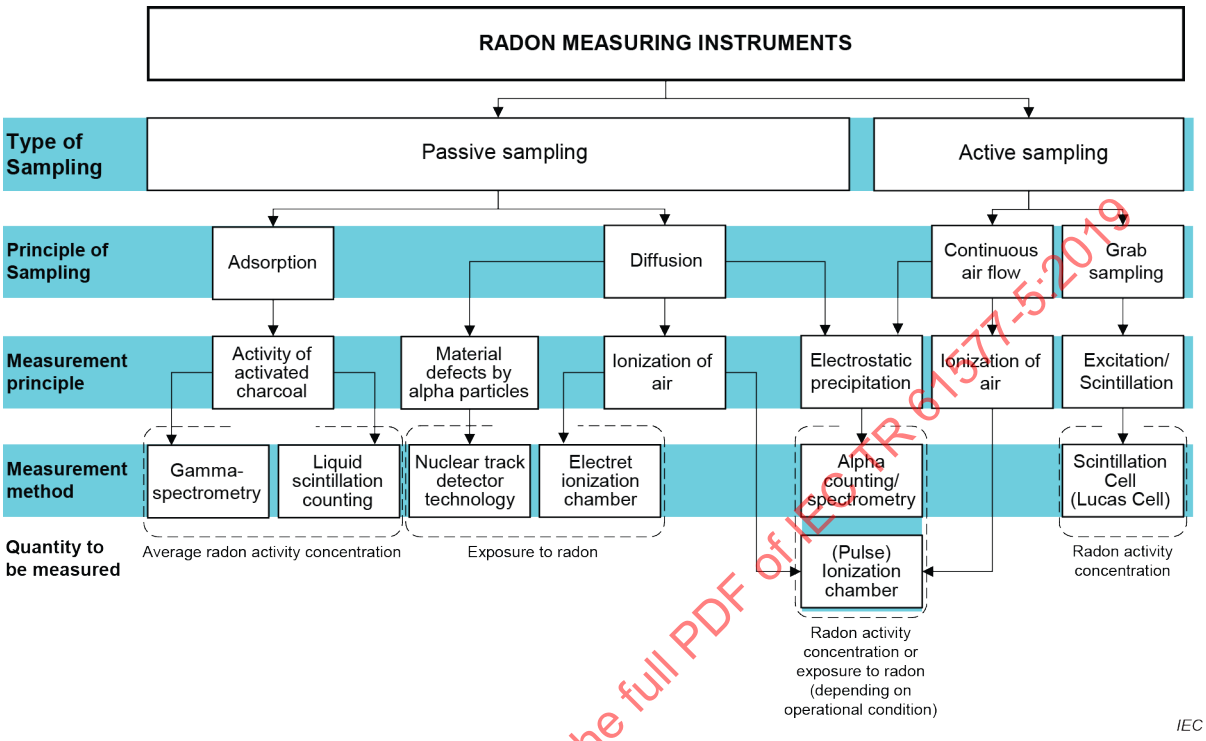


Figure A.1 – Sampling and measurement procedures commonly used for radon instruments

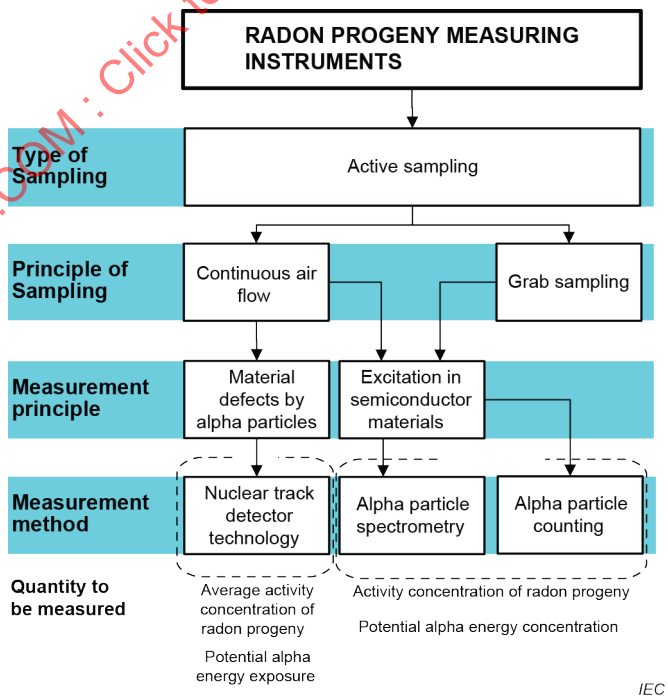


Figure A.2 – Sampling and measurement procedures commonly used for radon progeny instruments

Table A.1 – Physical and chemical characteristics [29]

	Specific gravity	Melting point	Boiling point
	$kg \cdot m^{-3}$	°C	°C
Radon (^{222}Rn, ^{220}Rn)	9,73	-71	- 61,7
Polonium (^{218}Po, ^{214}Po, ^{216}Po, ^{212}Po, ^{210}Po)	9 320	254	962
Bismuth (^{214}Bi, ^{212}Bi, ^{210}Bi)	9 747	271,4	1 564
Lead (^{214}Pb, ^{212}Pb, ^{210}Pb)	11 350	327,46	1 749
Thallium (^{208}Tl)	11 850	304	1 473

Table A.2 – ^{226}Ra, ^{222}Rn and radionuclides of the ^{222}Rn decay chain [37]

Symbol	Half-life	Major radiation energies					
		Alpha		Beta		Gamma	
		Energy	Intensity	Energy (average)	Intensity	Energy	Intensity
		[MeV]	[%]	[MeV]	[%]	[MeV]	[%]
$^{226}_{88}Ra$	1 600 y	4,601 4,784	6,16 93,84			0,186	3,64
$^{222}_{86}Rn$	3,8235 d	5,489	99,9				
$^{218}_{84}Po$	3,098 min	6,002	100				
$^{214}_{82}Pb$	26,8 min			0,205 0,226	45,9 40,2	0,295 0,352	18,4 35,6
$^{214}_{83}Bi$	19,9 min			0,492 0,525 0,539 1,269	8,1 17,0 17,6 19,1	0,609 1,120 1,764	45,5 14,9 15,3
$^{214}_{84}Po$	163,6 µs	7,686	100				
$^{210}_{82}Pb$	22,2 y			0,004 0,016	84 16	0,046	4,25
$^{210}_{83}Bi$	5,012 d			0,389	100		
$^{210}_{82}Po$	138,38 d	5,304	100				
$^{206}_{82}Pb$	stable						

Table A.3 – ^{224}Ra, ^{220}Rn and radionuclides of the ^{220}Rn decay chain [37]

Symbol	Half-life	Major radiation energies					
		Alpha		Beta		Gamma	
		Energy	Intensity	Energy (average)	Intensity	Energy	Intensity
		[MeV]	[%]	[MeV]	[%]	[MeV]	[%]
$^{224}_{88}Ra$	3,66 d	5,449 5,685	5,1 94,9				
$^{220}_{86}Rn$	55,6 s	6,288	99,9				
$^{216}_{84}Po$	0,145 s	6,778	100				
$^{212}_{82}Pb$	10,64 h			0,041 0,093 0,172	5,1 83,1 11,9	0,239	43,6
$^{212}_{83}Bi$	60,55 min	6,051 6,090	69,9 27,1	0,193 0,231 0,533 0,834	1,9 1,4 4,5 55,4	0,727	10,4
$^{212}_{84}Po$	0,299 µs	8,784	100 [b]				
$^{208}_{81}Tl$	3,053 min			0,441 0,535 0,649	24,2 [a] 22,2 [a] 49,1 [a]	0,511 0,583 0,861 2,614	22,6 [a] 85,0 [a] 12,5 [a] 99,8 [a]
$^{208}_{82}Pb$	stable						

[a] For intensity related to the parent when in equilibrium, multiply by 0,3594.

[b] For intensity related to the parent when in equilibrium, multiply by 0,6406.

Table A.4 – CSDA-Range of alpha particles emitted by Radon-222 and Radon-220 decay products in different materials [38]

	Alpha energy	CSDA-Range			
		Water	Air	Cellulose Nitrate (LR115)	Poly-carbonate (Makrofol, CR39)
	MeV	10^{-3} g·cm^{-2}	10^{-3} g·cm^{-2}	10^{-3} g·cm^{-2}	10^{-3} g·cm^{-2}
^{222}Rn	5,489	4,331	5,032	4,985	4,499
^{218}Po	6,002	4,970	5,775	5,708	5,170
^{214}Po	7,687	7,353	8,536	8,394	7,669
^{220}Rn	6,288	5,344	6,209	6,130	5,563
^{216}Po	6,778	6,014	6,986	6,886	6,265
^{212}Bi	6,057*	5,041	5,857	5,788	5,244
^{212}Po	8,785	9,131	10,591	10,395	9,532

* Average value

Densities: Water ρ = 1,00 g·cm^{-3}

Air, Dry, Sea level ρ = 1,20484·10^{-3} g·cm^{-3}

Cellulose Nitrate ρ = 1,49 g·cm^{-3}

Polycarbonate ρ = 1,20 g·cm^{-3}

Table A.5 – Solubility of radon in organic components [31]

Organic component		Ostwald coefficient	Temperature °C
Name	**Formula**		
Water	H_2O	0,245	20
Ethanol	C_2H_6O	6,03	20
Acetone	C_3H_6O	6,10	20
Ethyl acetate	$C_4H_8O_2$	7,16	18
Benzene	C_6H_6	12,82	18
Toluene	C_7H_8	13,24	20
Ethyl ether	$C_4H_{10}O$	14,80	20
Chloroform	$CHCl_3$	14,60	20
		15,10	18
Hexane	C_6H_{14}	14,70	20
		16,56	18

Table A.6 – Diffusion coefficients and diffusion lengths for radon in different materials [79]

Material	Diffusion coefficient 10^{-6} m^2·s^{-1}	^{222}Rn Diffusion length 10^{-3} m	^{220}Rn Diffusion length 10^{-3} m
Air	10	2 200	28
Water	10-3	22	0,3
Water-saturated soil	0,01 – 0,1	70 – 200	1 – 3
Wet clays and limes	0,1 – 5	200 – 1 550	3– 9
Dry sands and gravels	5 – 8	1 550 – 1 950	9– 25
Gypsum	2,35	1 100	
Pumice	1,50	850	
Slag stone	0,38	400	
Lime sandstone	0,34	400	
Porphyry	0,05	150	
Bricks	0,35	400	
Light concrete	1,30	800	
Standard concrete	0,007	60	
Polymer concrete	0,005	50	
Concrete, epoxy coated	0,002	30	
Cement-Silicate	< 10-6	< 0,7	
Plaster, plastic-containing	0,70	650	
Natural lacquer	0,001	20	
Artificial resin	< 10^{-6}	< 0,7	

Material	Diffusion coefficient 10^{-6} $m^2 \cdot s^{-1}$	^{222}Rn Diffusion length 10^{-3} m	^{220}Rn Diffusion length 10^{-3} m
Bitumen	< 10^{-6}	< 0,7	
PEHD Sealing foil	< 10^{-6}	< 0,7	
Bitumen rubber	< 10^{-6}	< 0,7	
Silicone rubber	< 10^{-6}	< 0,7	
Rubber sealing foil	10^{-5}	2	
Polypropylene foil	< 10^{-6}	0,3	
Thermofoil DBB (one-sided aluminum coated)	< 10^{-6}	0,5	
Polyamide paint or foil	< 10^{-6}	< 0,7	
Polymeric bitumen foil	< 10^{-6}	< 1,5	
Epoxy resin coating	< 10^{-6}	< 0,7	
Polyurethane sealing	< 10^{-6}	< 0,7	

Annex B
(informative)

Radioactive decay formulae

B.1 General

In this annex the analytical formulas describing the build-up and the decay of radioactivity are developed. According to the application to the decay of radon, a decay chain of three consecutive radionuclides representing ^{222}Rn decay products, and of two consecutive radionuclides representing ^{220}Rn decay products are considered. The formulas are also derived elsewhere [80]. It is assumed that radon decay products containing in the ambient air are collected by a particle filter. Starting with solving the differential equation system for the sampling, the buildup of the radioactivity on a filter over the sampling time is calculated. After cessation of sampling, the differential equation system describing the abatement of radioactivity taking the decay chain into consideration is solved. The analytical solutions can be used for the determination of the activity concentrations of radon decay products in the ambient air.

B.2 Symbols

Besides the symbols, which have already been introduced, the following symbols are used for this annex:

t_d	Time after cessation of sampling in seconds (s)
D	Number of alpha disintegrations observed during the time interval Δt
r_a, r_b, r_c, r_d	Substitutions defined in the text

B.3 Preliminary considerations and assumptions

Although ^{222}Rn and its decay products are the most important radionuclides that cause health effects due to inhalation, it cannot be excluded that environmental air samples also contain radonuclides from the ^{220}Rn decay chain or other airborne long-lived radionuclides. The formulas derived will restrict to the case of the determination of ^{222}Rn and ^{220}Rn decay products in air.

NOTE 1 In cases in which decay products of ^{220}Rn and/or other airborne long-lived radionuclides can be disregarded, the formulas given in this document may be reasonably simplified.

Samples of radon decay products being attached or unattached to aerosol particles are commonly collected by aerosol filters or wire screens, but even other collection methods might be employed. These samples are collected over some period of time, usually shorter than 1 h. After the collection of the air samples has completed, the alpha activity on each sample is observed by counting over various intervals of time. Any counting technique measuring gross counts from alpha disintegrations over a finite time interval is suitable.

Because radon decay products comprise a relatively fast decaying chain of radionuclides, the composition of a sample rapidly changes during the collection as well as during the counting periods. Multiple different observations of the alpha activity are necessary to ascertain the character of the decay of each sample in order to calculate the amounts of the various decay products, which were originally collected.

NOTE 2 Since ^{222}Rn and ^{220}Rn are gaseous and their decay products are solids, dust sampling techniques collect only the decay products.

Since alpha particles are the only radiation, which will be considered, the nuclides which decay by alpha emission are of primary interest.

^{222}Rn decay product chain: 99,98 % of the decays of ^{218}Po result in the emission of alpha particles that is, therefore, reasonably considered a pure alpha emitter. ^{214}Pb is not an alpha emitter, but it contributes to the appearance of alpha particle in the subsequent decay chain. Although ^{214}Bi is not an alpha emitter, it effectively behaves as one, since it primarily decays to ^{214}Po, which instantaneously emits an alpha particle. ^{210}Pb is not an alpha emitter. In comparison to the other radionuclides, it has a long half-life of about 20 years, which makes this radionuclide of no importance for the analytical technique described in the following.

^{220}Rn decay product chain: Because of the short half-life of ^{216}Po, it decays during sampling to ^{212}Pb, and thus evades it from counting. ^{212}Pb is not an alpha emitter, but it is important because it decays relatively slowly to ^{212}Bi, which is effectively an alpha emitter in that 36 % decays by alpha emission and 64 % decays by beta emission to ^{212}Po which also instantaneously emits an alpha particle. The resulting ^{208}Pb is stable and needs not be considered.

B.4 Build-up of filter activity during sampling

The mathematical analysis is based on the following assumptions:

a) The concentrations of the radionuclides in the atmosphere to be sampled are considered as being unchanged during the sampling period or can satisfactorily be described by averages.

b) The volumetric rate of sampling is constant over the sampling period or can be satisfactorily considered as an average volumetric rate.

c) The efficiency of sampling is equal for all radionuclides ($\varepsilon_s = \varepsilon_{s1} = \varepsilon_{s2} = \varepsilon_{s3} = \varepsilon_{s4} = \varepsilon_{s5}$).

The build-up of atoms attributed to the ^{222}Rn and the ^{220}Rn decay chain on the filter during sampling are described by the following formulas:

At first, the ^{222}Rn decay chain is considered. The build-up of ^{218}Po is described by the differential formula:

$$dN_1 = \frac{\varepsilon_s v}{\lambda_1} C_1 \, dt - \lambda_1 N_1 \, dt \, . \tag{B.1}$$

With the initial condition $N_1(t_s = 0) = 0$, it yields:

$$N_1(t_s) = \varepsilon_s \, v C_1 \frac{\left(1 - e^{-\lambda_1 t_s}\right)}{\lambda_1^2} \, . \tag{B.2}$$

The build-up of ^{214}Pb is decribed by the differential formula:

$$dN_2 = \frac{\varepsilon_s v}{\lambda_2} C_2 \, dt - \lambda_2 N_2 \, dt + \lambda_1 N_1 \, dt \, . \tag{B.3}$$

With the initial condition $N_2(t_s = 0) = 0$, it yields:

$$N_2(t_s) = \varepsilon_s \, v C_1 \left[\frac{\left(1 - e^{-\lambda_2 t_s}\right)}{\lambda_1 \lambda_2} - \frac{\left(e^{-\lambda_2 t_s} - e^{-\lambda_1 t_s}\right)}{\lambda_1 (\lambda_1 - \lambda_2)} \right] + \varepsilon_s \, \dot{v} C_2 \left[\frac{\left(1 - e^{-\lambda_2 t_s}\right)}{\lambda_2^2} \right]. \tag{B.4}$$

The build-up of ^{214}Bi is described by the differential formula:

$$dN_3 = \frac{\varepsilon_s \, v}{\lambda_3} C_3 \, dt - \lambda_3 \, N_3 \, dt + \lambda_2 \, N_2 \, dt \, . \tag{B.5}$$

With the initial condition $N_3(t_s = 0) = 0$, it yields:

$$N_3(t_s) = \varepsilon_s \, v C_1 \left[\frac{\left(1 - e^{-\lambda_3 t_s}\right)}{\lambda_1 \lambda_3} - \frac{\left(e^{-\lambda_2 t_s} - e^{-\lambda_3 t_s}\right)}{\lambda_1 (\lambda_3 - \lambda_2)} - \frac{\lambda_2 \left(e^{-\lambda_2 t_s} - e^{-\lambda_3 t_s}\right)}{\lambda_1 (\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} + \frac{\lambda_2 \left(e^{-\lambda_3 t_s} - e^{-\lambda_1 t_s}\right)}{\lambda_1 (\lambda_1 - \lambda_3)(\lambda_1 - \lambda_2)} \right] + \tag{B.6}$$
$$+ \varepsilon_s \, v C_2 \left[\frac{\left(1 - e^{-\lambda_3 t_s}\right)}{\lambda_2 \lambda_3} - \frac{\left(e^{-\lambda_2 t_s} - e^{-\lambda_3 t_s}\right)}{\lambda_2 (\lambda_3 - \lambda_2)} \right] + \varepsilon_s \, v C_3 \frac{\left(1 - e^{-\lambda_3 t_s}\right)}{\lambda_3^2}$$

Next up, the ^{220}Rn decay chain is considered. The formulas for the build-up are equivalent to the formulas for the first and second nuclide of the ^{222}Rn chain. Because of the short half-life of ^{216}Po, the first relevant nuclide to be considered is ^{212}Pb. The build-up of ^{212}Pb is described by the differential formula:

$$dN_4 = \frac{\varepsilon_s \, v}{\lambda_4} C_4 \, dt - \lambda_4 \, N_4 \, dt \, . \tag{B.7}$$

With the initial condition $N_4(t_s = 0) = 0$, it yields:

$$N_4(t_s) = \varepsilon_s \, v C_4 \frac{\left(1 - e^{-\lambda_4 t_s}\right)}{\lambda_4^2} . \tag{B.8}$$

The build-up of ^{212}Bi is described by the differential formula:

$$dN_5 = \frac{\varepsilon_s \, v}{\lambda_5} C_5 \, dt - \lambda_5 \, N_5 \, dt + \lambda_4 \, N_4 \, dt \, . \tag{B.9}$$

With the initial condition $N_5(t_s = 0) = 0$, it yields:

$$N_5(t_s) = \varepsilon_s \, v C_4 \left[\frac{\left(1 - e^{-\lambda_5 t_s}\right)}{\lambda_4 \lambda_5} - \frac{\left(e^{-\lambda_5 t_s} - e^{-\lambda_4 t_s}\right)}{\lambda_4 (\lambda_4 - \lambda_5)} \right] + \varepsilon_s \, v C_5 \left[\frac{\left(1 - e^{-\lambda_5 t_s}\right)}{\lambda_5^2} \right] . \tag{B.10}$$

As t_s refers to the time at the end of the sample collection, $N_i(t_s)$ is the number of atoms of each of the radionuclides on the sample at the end of collection.

B.5 Decay of the filter activity after cessation of sampling

After the end of sampling no further accumulation of radionuclides on the filter takes place. The sampled radionuclides decay according to the formulas below. The time variable t_d is the time elapsed from the end of sampling. After cessation of sampling at time t_s, the number of atoms $N_i(t_s)$ on the filter is the initial condition for the system of decay formulas.

The decay of atoms on the filter after the end of sampling is described by the following formulas:

At first the ^{222}Rn decay chain is considered. The decay of ^{218}Po is described by the differential formula:

$$dN_1 = -\lambda_1 N_1 dt \,. \tag{B.11}$$

With the initial condition $N_1(t_d = 0) = N_1(t_s)$, it yields:

$$N_1(t_d) = N_1(t_s) e^{-\lambda_1 t_d} \,. \tag{B.12}$$

The decay of ^{214}Pb is described by the differential formula:

$$dN_2 = -\lambda_2 N_2 dt + \lambda_1 N_1 dt \,. \tag{B.13}$$

With the initial condition $N_2(t_d = 0) = N_2(t_s)$, it yields:

$$N_2(t_d) = N_1(t_s) \frac{\lambda_1 \left(e^{-\lambda_2 t_d} - e^{-\lambda_1 t_d}\right)}{\lambda_1 - \lambda_2} + N_2(t_s) e^{-\lambda_2 t_d} \,. \tag{B.14}$$

The decay of ^{214}Bi is described by the differential formula:

$$dN_3 = -\lambda_3 N_3 dt + \lambda_2 N_2 dt \,. \tag{B.15}$$

With the initial condition $N_3(t_d = 0) = N_3(t_s)$, it yields:

$$N_3(t_d) = N_1(t_s) \left[\frac{\lambda_1 \lambda_2 \left(e^{-\lambda_2 t_d} - e^{-\lambda_3 t_d}\right)}{(\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} - \frac{\lambda_1 \lambda_2 \left(e^{-\lambda_3 t_d} - e^{-\lambda_1 t_d}\right)}{(\lambda_1 - \lambda_3)(\lambda_1 - \lambda_2)} \right] + N_2(t_s) \frac{\lambda_2 \left(e^{-\lambda_2 t_d} - e^{-\lambda_3 t_d}\right)}{(\lambda_3 - \lambda_2)} + N_3(t_s) e^{-\lambda_3 t_d} \tag{B.16}$$

Next up, the ^{220}Rn decay chain is considered. The formulas for the decay are equivalent to the formulas for the first and second nuclide of the ^{222}Rn chain. The decay of ^{212}Pb is described by the differential formula:

$$dN_4 = -\lambda_4 N_4 dt \,. \tag{B.17}$$

With the initial condition $N_4(t_d = 0) = N_4(t_s)$, it yields:

$$N_4(t_d) = N_4(t_s) e^{-\lambda_4 t_d} \quad . \tag{B.18}$$

The decay of ^{212}Bi is described by the differential formula:

$$dN_5 = -\lambda_5 N_5 \, dt + \lambda_4 N_4 \, dt \quad . \tag{B.19}$$

With the initial condition $N_5(t_d = 0) = N_5(t_s)$, it yields:

$$N_5(t_d) = N_4(t_s) \frac{\lambda_4 \left(e^{-\lambda_5 t_d} - e^{-\lambda_4 t_d} \right)}{\lambda_4 - \lambda_5} + N_5(t_s) e^{-\lambda_5 t_d} \quad . \tag{B.20}$$

B.6 Number of alpha disintegrations registered after sampling

Considering a filter on which ^{222}Rn and ^{220}Rn decay products have been deposited during the sampling period. After cessation of sampling, the alpha activity of the filter is measured. The alpha activity, represented by the gross number of alpha disintegrations per unit time at any time t_d elapsed from the end of sampling, is given by the summation of contributing radionuclide: ^{218}Po indicated by N_1, ^{214}Bi/^{214}Po indicated by N_3 and ^{212}Bi/^{212}Po indicated by N_5. It should be noted that ^{212}Bi can emit an alpha particle directly or via ^{212}Po. The activity of the filter at time t_d is given by:

$$A = \lambda_1 N_1 + \lambda_3 N_3 + \lambda_5 N_5 \quad , \tag{B.21}$$

The integration of formula (B.21) over the time interval from $t_d = \alpha$ to $t_d = \beta$ yields the gross number *D* of alpha disintegrations on the filter within this time period with:

$$D = \int_{\alpha}^{\beta} A \cdot dt \quad . \tag{B.22}$$

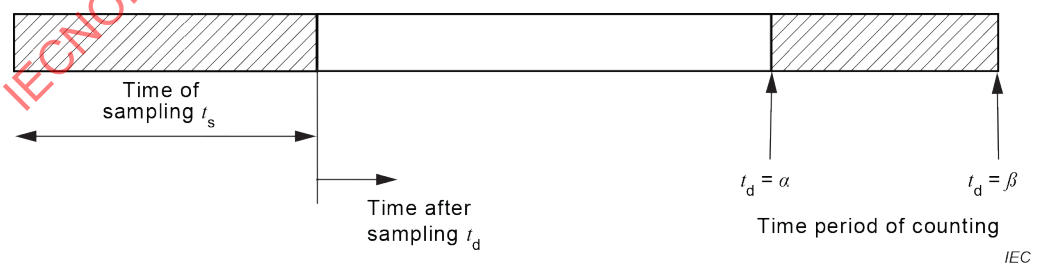


Figure B.1 – Scheme for sampling and counting

The measurement is delineated in Figure B.1. Taking into account that alpha disintegrations will be registered by an instrument with a nuclide-specific counting efficiency ε_{ci}, the gross number *D* of alpha disintegrations on the filter within the time period of counting is given by:

$$
\begin{aligned}
D = {} & N_1(t_s)\,\varepsilon_{c3}\left[-\left(\frac{\varepsilon_{c1}}{\varepsilon_{c3}}+\frac{r_b}{\lambda_1}\right)\left(e^{-\lambda_1\beta}-e^{-\lambda_1\alpha}\right)-\frac{r_a}{\lambda_2}\left(e^{-\lambda_2\beta}-e^{-\lambda_2\alpha}\right)+\frac{(r_a+r_b)}{\lambda_3}\left(e^{-\lambda_3\beta}-e^{-\lambda_3\alpha}\right)\right] \\
& + N_2(t_s)\,\varepsilon_{c3}\left[-\frac{r_c}{\lambda_2}\left(e^{-\lambda_2\beta}-e^{-\lambda_2\alpha}\right)+\frac{r_c}{\lambda_3}\left(e^{-\lambda_3\beta}-e^{-\lambda_3\alpha}\right)\right] \\
& + N_3(t_s)\,\varepsilon_{c3}\left[-\left(e^{-\lambda_3\beta}-e^{-\lambda_3\alpha}\right)\right] \\
& + N_4(t_s)\left(0{,}36\,\varepsilon_{c5}+0{,}64\,\varepsilon_{c5'}\right)\left[\frac{r_d}{\lambda_4}\left(e^{-\lambda_4\beta}-e^{-\lambda_4\alpha}\right)-\frac{r_d}{\lambda_5}\left(e^{-\lambda_5\beta}-e^{-\lambda_5\alpha}\right)\right] \\
& + N_5(t_s)\left(0{,}36\,\varepsilon_{c5}+0{,}64\,\varepsilon_{c5'}\right)\left[-\left(e^{-\lambda_5\beta}-e^{-\lambda_5\alpha}\right)\right]
\end{aligned}
\qquad .(B.23)
$$

In formula (B.23) the following substitutes are used:

$$r_a = \frac{\lambda_1\,\lambda_2\,\lambda_3}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)}, \qquad (B.24)$$

$$r_b = \frac{\lambda_1\,\lambda_2\,\lambda_3}{(\lambda_1-\lambda_2)(\lambda_1-\lambda_3)}, \qquad (B.25)$$

$$r_c = \frac{\lambda_2\,\lambda_3}{(\lambda_3-\lambda_2)}, \qquad (B.26)$$

$$r_d = \frac{\lambda_4\,\lambda_5}{(\lambda_4-\lambda_5)}. \qquad (B.27)$$

The parameter ε_{c1} represents the counting efficiency of alpha particles emitted by ^{218}Po with an energy of about 6 MeV, and ε_{c3} is that of alpha particles emitted by ^{214}Po with an energy of about 7,7 MeV. The term $(0{,}36\,\varepsilon_{c5}+0{,}64\,\varepsilon_{c5'})$ takes into account the two possible branches of the decay of ^{212}Bi. The parameter ε_{c5} is the counting efficiency of alpha particles emitted directly by ^{212}Bi with an energy of about 6,05 MeV, and $\varepsilon_{c5'}$ is that of the other branch, and represents alpha particles emitted by ^{212}Po with an energy of about 8,8 MeV.

Annex C
(informative)

Uncertainty analysis for the method of multiple successive countings to determine the activity concentrations of radon and thoron decay products

C.1 Symbols

Besides the symbols, which have already been introduced, the following symbols are used for this annex:

t_0	Measurement time for the determination of background in seconds (s)
t_c	Time interval of counting in seconds (s)
$D_{k,b}$	Coefficients of vector $\mathbf{D}_b$: Number of gross alpha disintegrations observed during the time interval t_c
D_0	Coefficients of vector $\mathbf{D}_0$: Number of background count events observed during the time interval t_0
$\mathbf{U}^2(\ )$	Diagonal matrix of variances (squared uncertainties)
$\mathbf{D}_b$, $\mathbf{D}_0$, $\mathbf{G}$	Matrices and vectors
$g_{i,k}$	Coefficients of matrix $\mathbf{G}$:
ω	Coefficient to gather the correction factors and the flow rate in hours per cubic metre (h·m^{-3})
$\tilde{C}_j$	True value of the activity concentration of the j^{th} radionuclide in Becquerels per cubic metre (Bq·m^{-3})
C_j^*	Detection threshold of the activity concentration of the j^{th} radionuclide in Becquerels per cubic metre (Bq·m^{-3})
$C_j^{\#}$	Detection limit of the activity concentration of the j^{th} radionuclide in Becquerels per cubic metre (Bq·m^{-3})
$\hat{C}_j$	Best estimate of the activity concentration of the j^{th} radionuclide in Becquerels per cubic metre (Bq·m^{-3})
$C_j^{\triangleleft}$, $C_j^{\triangleright}$	Lower and upper confidence limit of the activity concentration of the j^{th} radionuclide in Becquerels per cubic metre (Bq·m^{-3})
k_q	Quantiles of the standardized normal distribution for the probability q, respectively (for instance $q = 1-\alpha, 1-\beta, 1-\gamma/2$)

C.2 Uncertainties of the parameter of the model function

The uncertainty analysis is carried out for the activity concentrations of radon and thoron decay products in air. It is assumed that an air stream passes a filter during a time period t_s. The air containing particles is deposited on the filter. After cessation of sampling, the gross alpha disintegrations are registered by multiple countings within consecutive time periods t_c. From the numbers of count events, the activity concentration of the ^{222}Rn and ^{220}Rn decay products are estimated. The measurement method is identical to the method described in 7.3.3.4 but with using equivalent periods of counting times, t_c.

According to formulas (C.1) and (C.2), the model for the analysis is:

$$\mathbf{C} = \frac{1}{\varepsilon_c \, \varepsilon_s \, v} \mathbf{M} \left(\mathbf{A}^{\mathsf{T}} \mathbf{A} \right)^{-1} \mathbf{A}^{\mathsf{T}} \mathbf{D} \tag{C.1}$$

where $\mathbf{D}$ is the vector of the net count events, D_k, within the time period $t_c = \beta_k - \alpha_k$ for $k = 1, \ldots, n$. Taking into account the gross count events, $D_{k,b}$, and the background count events, D_0, measured within a time period t_0, it yields:

$$\mathbf{D} = \begin{vmatrix} D_1 \\ \vdots \\ \vdots \\ D_k \\ \vdots \\ D_n \end{vmatrix} = \begin{vmatrix} D_{1,b} - \frac{t_c}{t_0} D_0 \\ \vdots \\ \vdots \\ D_{k,b} - \frac{t_c}{t_0} D_0 \\ \vdots \\ D_{n,b} - \frac{t_c}{t_0} D_0 \end{vmatrix} \tag{C.2}$$

The matrix of the squared uncertainties $\mathbf{U}^2(\mathbf{D})$ attributed to the countings during the time periods $i = 1 \ldots n$ contains contributions from the uncertainties of the gross counts and the background counts. The matrix representing the uncertainties of the gross counts is obtained from the derivative of formula (C.2) with respect to the vector of the gross counts, $\partial / \partial \mathbf{D}_b$. Since no covariances exist, a diagonal matrix with unity elements results. Its multiplication with the uncertainties of the gross counts, $\Delta \mathbf{D}_b$, which in turn is a diagonal matrix, and squaring down yields:

$$\begin{aligned} \mathbf{U}^2(\mathbf{D}_b) &= \left(\Delta \mathbf{D}_b \left(\frac{\partial}{\partial \mathbf{D}_b} \mathbf{D} \right) \right)^T \left(\Delta \mathbf{D}_b \left(\frac{\partial}{\partial \mathbf{D}_b} \mathbf{D} \right) \right) \\ &= \left[\begin{pmatrix} \Delta D_{1,b} & 0 & \cdots & 0 \\ \vdots & \Delta D_{2,b} & \cdots & 0 \\ 0 & 0 & \ddots & \vdots \\ 0 & 0 & \cdots & \Delta D_{n,b} \end{pmatrix} \begin{pmatrix} 1 & 0 & \cdots & 0 \\ \vdots & 1 & \cdots & 0 \\ 0 & 0 & \ddots & \vdots \\ 0 & 0 & \cdots & 1 \end{pmatrix} \right]^2 \\ &= \begin{pmatrix} \Delta D_{1,b}^2 & 0 & \cdots & 0 \\ \vdots & \Delta D_{2,b}^2 & \cdots & 0 \\ 0 & 0 & \ddots & \vdots \\ 0 & 0 & \cdots & \Delta D_{n,b}^2 \end{pmatrix} \end{aligned} \tag{C.3}$$