Procedure for determining airborne particulate radionuclides in air near the ground by gamma spectrometry – Standard Procedure –

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Authors:
W. Kiesewetter
H. Diedrich
W. Dyck
T. Steinkopff
H. Ulbricht

Federal coordinating office for air and precipitation
(Leitstelle für Luft und Niederschlag)
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1 Scope

The procedure described here facilitates the nuclide-specific identification of gamma-emitters in samples of suspended matter by measurement of the sample without prior chemical preparation. It conforms to the requirements for measuring programmes of the AVV-IMIS (1) and the guidelines for the monitoring of emissions and immissions of nuclear installations (REI) (2).

2 Sampling

Guidelines for sampling airborne particulate radionuclides in the framework set for the monitoring of radioactivity in the air are summarised, amongst others, in DIN Standard 25423 (3) and VDI Guideline 2463 (4). Complementary procedures for the collection of representative samples are described in detail in the following chapters (5, 6).

2.1 General criteria for the selection of the sampling location

In general, collecting samples will alter the size distribution of aerosol particles in the air sample compared to their original distribution in the outside air. Air currents, turbulences in the air, the spatial structure of the entire measuring point and its influences on the airflow, the distribution of particle sizes, the ratio of particle surface to mass, and the geometry of the sampling devices and the flow of the volume of sampled air may exert major influences on the precipitation of airborne particulate.

The aim of the procedure described here is to measure the activity concentration of radioactive airborne particulate in the outside air in a representative manner. This necessitates a complete deposition of all aerosol particles on a filter. Sampling is preferably effected at a level 1,5 m to 5 m above the ground. The distance of the point of sampling to the nearest obstacle should be at least equal to ten times the height of the obstacle (7).

The characteristics of the sampling location (coordinates, type of vegetation, relative arrangement of obstacles) need to be documented prior to commencing the measurements.

If measurements are to be taken in the surroundings of emitters, like for example as part of the monitoring process of nuclear installations, the sampling location should be chosen to lie within the sector of the predominant wind direction (2).
2.2 Alignment of the sampling nozzle

The sampling equipment consists of a pump for sucking in air, a support unit for the filter that is to collect aerosols, a short inlet air duct, and the sampling nozzle. The latter should in general be installed at least 1.5 m above ground level and be free all around so that air can be sucked in without any obstructions. Compromising influences from walls, roofs, pipes etc. need to be avoided.

If the sampler is installed in a building, setting up the sampling nozzle outside on a flat roof in a vertical and axisymmetric manner that is independent on the direction of the wind offers an optimal solution. In order to minimize the possible influence of turbulences along the edges of the roof, the sampling nozzle is best installed in the centre of the roof.

Setting up a sampling nozzle on a roof may in many cases be impossible due to details of the construction. In such a case, mounting the sampling nozzle to an outside wall can be justified in the context of monitoring radioactivity in the environment. If sampling is to take place at a distance of 1 km to 2 km from the emission source, an outside wall should be selected that is positioned parallel to the imagined connecting line from the location of the emission source to the building that serves as the sampling location. Collecting samples on the far side (lee-side) relative to the location of the emitter is not recommendable due to the risk of turbulences.

The sampling nozzle should be shielded against the intrusion of rain or snow. It furthermore needs to be secured against sucking in insects by covering it with a corrosion-resistant grille with a mesh width of ca. 5 mm. This grille should be heated in winter in order to keep it from freezing shut; alternatively, it is advisable to remove the grille in winter.

In order to avoid condensation caused by high humidity, at least the sampling duct, and if necessary also the collection and measuring components, should be heated so that their temperatures remain above dew point.

A sufficiently large distance between the sampling nozzle and the air outlet is required to prevent exhaust air from being sucked into the sampler once more.

2.3 Geometry of the sampling nozzle and mounting of the filter

As far as the geometry of the sampling nozzle is concerned, there are two options that are in principle suitable (Figure 1):

(A) Sampler with an integrated filter bracket

(B) Sampler with a spatially separated filter bracket

A representative, reproducible detection of the total dust load, i.e., the airborne particulate collective, with aerodynamic equivalent-diameters (AED) of 0.001 µm to 150 µm is difficult to realize in practice. The size spectrum of airborne particulate matter extracted from the air is largely determined by the shape of the sampling nozzle as well as the air intake speed in the sampling nozzle and the wind speed at the time of sampling. The minimum diameter of aerosol particles recorded increases with rising air intake speeds and decreasing wind speeds. In order to be able to capture aerosol particles larger than 1 µm AED even at higher wind speeds, a minimum air intake speed of 0.12 m·s⁻¹ to 3 m·s⁻¹ needs to be maintained (5) depending on the type of sampling nozzle used.
Fig. 1: Schematic diagram of samplers

In order to capture so-called respirable dust (i.e., airborne particulate matter < 10 µm AED) independent of wind speeds, suitable prefilters are used to separate coarse dust particles.

To avoid losses in separation effectiveness, the length of the air inlet duct should not exceed 3 m and it should be installed without bends and changes in diameter. If bends cannot be avoided, the radius of the curvature should not be smaller than three times the duct’s diameter.

An inlet duct made from corrosion-resistant steel with a smooth surface should be given preference over other materials. Plastic pipes have the disadvantage of potentially becoming electrostatically charged. Both corrugated hoses and hoses with wire mesh reinforcement act as airborne particulate matter traps. Welded pipes need to be checked for smooth welding seams. This applies in particular to flange joints. Flanges that are nearly perfectly free of burrs and connect pipes without gaps are commercially available.

The inlet duct needs to be heatable in order to prevent condensation effects on the pipes and the filter at times of temperatures below the dew point (see section 2.2). The corrosion-resistant filter bracket has to be shaped to sufficiently support the filter and should be fitted with a seal that excludes the possibility of secondary air being taken in.

An example of the mechanical structure of a sampling nozzle can be found in the VDI Guideline (8).

2.4 Filter

The selection of the filter medium depends on a number of criteria. It has to have a mechanical stress resistance that allows for using it in conditions of high air throughput rates for extended periods of time. The separation of aerosol particles with an AED in the range of up to 10 µm has to be ensured by using filters with corresponding specifications, i.e., those of the S Class according to DIN 24184 (9) or DIN EN 1822-1 (10), for example by choosing a fibreglass filter with a high degree of separation. The choice of filter medium should also take into consideration that it might need to be suitable for radiochemical processing subsequent to its gamma-spectrometric examination, as may be required for the identification of alpha and beta emitters. A cellulose fibre filter whose mechanical stability has been improved by reducing its fibreglass content may be appropriate in these instances.
2.5 Determination of air volume throughput

To determine the activity concentration it is necessary to precisely ascertain the total air volume throughput or the air volume throughflow and the sampling period. The air volume throughflow should always be measured behind the filter. Measuring instruments often suitable for this purpose may be simple aerosol throughflow meters or small turbine- and impeller-driven instruments.

Particularly accurate measuring techniques are based on recording differential pressures. These make use of an aperture situated behind the filter and measure the differential between the pressure above the aperture and the absolute barometric pressure in conjunction with the air temperature and the collection period and so make it possible to exactly quantify the air volume throughput via the ideal gas law. Alternatively, calorimetric, ultrasonic and vibration measuring techniques are used.

Volumetric flow meters are in general designed for use within narrow measuring limits that will usually span less than two orders of magnitude. Selecting a suitable volumetric flow meter therefore requires great care, considering that volumetric flows from as little as 1 m³·h⁻¹ to more than 500 m³·h⁻¹ may be encountered in practice.

With the procedures for the quantification of air volume throughputs depending on temperature and barometric pressure, it is necessary to record whether operational cubic metres or air in its regular state, i. e., at 0 °C and 1013 hPa (standard cubic metre), are used. The computation of standard cubic metres requires recording the temperature of the airflow and the absolute pressure.

2.6 Influence of “hot particles”

The analysis of samples collected at intervals of days or even weeks and the subsequent preparation of counting sources, for example, by compacting the filter, does not provide information about whether the noted increased activity of the sample is a result of having captured “hot particles” or whether it is due to the presumed continual collection of minuscule aerosol particles. If the presence of “hot particles” (11) is suspected, the processing of the sample (pressing, ashing) needs to be preceded by a procedure that provides spatial resolution on the filter. In the simplest scenario and a large filter surface, this can be effected by means of a surface contamination monitor. More detailed information is obtained from an auto-radiographic examination or from measuring the sample with a thin-layer scanner or a location-sensitive multi-wire gas proportional counter (digital autoradiography). These procedures are much more elaborate, however.

2.7 Sampling installations at the monitoring stations for radioactivity of the Deutscher Wetterdienst (DWD)

The weather stations of the synoptic climatological data collection network of the DWD are arranged in accordance with the directives of the World Meteorological Organisation (WMO) and meant to provide meteorological data that are representative of an extended geographical area. It is for this reason that the stations monitoring radioactivity in the environment have been set up at selected weather stations, too.

The sampling installations for the collection of suspended particles are always accommodated inside the respective station buildings. The sampling nozzle is, in most instances, mounted to an outside wall, and the air is channelled to the filter...
via a short supply pipe. The sampling intervals are based on the directives relating to routine and intensified monitoring programmes as stipulated in the appendices of the AVV-IMIS (1).

The following table provides an overview of the sampling installations at the locations for monitoring radioactivity of the DWD:

**Tab. 1:** Sampling installations at the locations for monitoring radioactivity of the DWD

<table>
<thead>
<tr>
<th>Type of sampler</th>
<th>Filter</th>
<th>Volume metering</th>
<th>Volume throughflow in m³·h⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>Annular duct blower</td>
<td>Fibreglass, Class S d = 257 mm</td>
<td>Thermo-electrical</td>
<td>65-95</td>
</tr>
<tr>
<td>Rotary piston blower</td>
<td>Cellulose fibre (80 %), fibreglass (20 %), 420 mm x 380 mm</td>
<td>Operating period, pressure, temperature</td>
<td>145-160, 200-300</td>
</tr>
<tr>
<td>Radial blower</td>
<td>PVC with cotton support grille, 400 mm x 400 mm</td>
<td>Thermo-electrical</td>
<td>260-750</td>
</tr>
</tbody>
</table>

The volume throughflow of the radial blower amounts to 95 m³·h⁻¹ with a freshly installed filter and decreases to 65 m³·h⁻¹ towards the end of a dust-collection interval of one week.

3 **Analysis**

3.1 **Principle of the method**

Airborne particulate matter is collected on a filter. The filter is processed and then subjected to gamma spectrometric analysis.

3.2 **Sample preparation**

The filters are folded and moulded in a hydraulic press into a solid right-parallelepiped shape of little height. The compacted pellet obtained thus is sealed into polyethylene foil, fixed in a petri dish, and positioned on a germanium detector (DWD: the use of a cellulose fibre filter produces a surface of 2 cm x 2 cm and a thickness of ca. 0,7 cm).

3.3 **Radiochemical separation**

Radiochemical separation is not required for the procedure outlined here.
4 Measuring the activity

4.1 General

The pellets produced according to section 3.1 are examined by gamma-spectrometry. In order to obtain a detection limit in the range of ca. 10 μBq·m⁻³, a high-purity germanium detector with a relative detection efficiency of at least 40 %, should be employed. Here, the detection efficiency of 40 % refers to a 3 x 3” NaI(Tl) crystal and a half-width of the 1332 keV gamma line of the radionuclide Co-60 of less than 2,0 keV. The shielding of the detector should be made of minimum-radiation lead at least 100 mm thick. A sheet of copper needs to be installed between the lead shield and the detector in order to suppress the Roentgen radiation of the lead at 75 keV and 85 keV, and Perspex that is meant to absorb the beta particles that are dispersed on the sheet of copper. Measuring processes at lower resolutions may require a less elaborate outfitting.

In this context, reference is made to the basics of gamma spectrometry outlined in chapter IV.1.1 of this procedures manual.

4.2 Calibration

Calibration is achieved by using a surface source that contains several radionuclides with known activities. This surface source is measured in a petri dish and, if necessary, mounted on a spacer, with a detector at a distance, \( d \). The distance, \( d \), corresponds to half the thickness of a pellet plus the thickness of the bottom of the petri dish. This supposes that the activity is homogeneously distributed in the pellet.

5 Calculation of the results

The calculation of the activity concentration, \( c \), relative to the mean collection period follows the equation (1):

\[
c_r = \frac{N_n}{\varepsilon_r \cdot p_r \cdot t_m \cdot V \cdot f_1 \cdot f_2 \cdot f_3}
\]

with

\[
f_2 = e^{\lambda_r \cdot t_m}
\]

and

\[
f_3 = \frac{\lambda_r \cdot t_m}{1 - e^{-\lambda_r \cdot t_m}}
\]

wherein:

- \( c_r \) activity concentration of the radionuclide, \( r \), in Bq·m⁻³;
- \( N_n \) number of net counts;
- \( \varepsilon_r \) detection efficiency of the radionuclide, \( r \), in Bq·m⁻³;
- \( p_r \) emission probability of gamma radiation;
- \( t_m \) duration of counting source measurement, in s;
- \( V \) processed air volume, in m³;
- \( \lambda_r \) decay constant of the radionuclide, \( r \), in s⁻¹;
\( t_A \)  period from the middle of the sampling interval to the commencement of measuring, in s;
\( f_1 \) summation correction factor;
\( f_2 \) correction factor for the decay of radionuclides within the period from the middle of the sampling interval to the commencement of measuring;
\( f_3 \) correction factor for the decay of the radionuclides during the measurement.

The summation correction factor, \( f_1 \), is determined according to chapter IV.1, section 5, of this procedures manual.

If the period \( t_A \) or \( t_m \), as may be the case, is much smaller than the half-life of the radionuclide to be measured, \( f_2 = 1 \) or \( f_3 = 1 \) will apply, respectively.

According to chapter IV.5, section 2.2.3, of this procedures manual, the standard deviation, \( s(R_n) \) of the net count rate, \( R_n \), is calculated as follows:

\[
s(R_n) = \sqrt{\frac{R_n + b \cdot R_0(E)}{t_m}} \cdot \left(1 + \frac{b}{2 \cdot L}\right)
\]  \hspace{1cm} (4)

where
\( s(R_n) \) standard deviation of the net count rate, in s\(^{-1}\)
\( R_n \) net count rate, in s\(^{-1}\)
\( b \) base width of the gamma line in the number of channels
\( R_0(E) \) mean background count rate per channel, in s\(^{-1}\)
\( E_\gamma \) energy of the gamma line
\( L \) number of channels used for determining the background effect

For the standard deviation of the activity concentration, \( s(c_r) \), the following equation applies:

\[
s(c_r) = s(R_n) \cdot \frac{c_r}{R_n} = \frac{1}{\varepsilon_r \cdot p_r \cdot V} \cdot f_1 \cdot f_2 \cdot f_3 \cdot s(R_n)
\]  \hspace{1cm} (5)

5.1 Worked example

The following are two examples of calculating the activity concentration of Cs-137 and I-131 in different samples.

a) Calculation of the activity concentration of the airborne particulate radionuclide Cs-137:

The calculation of the activity concentration of Cs-137 in the air is based on filling the equation (1) with the following values:

\( N_g = 3415; \)
\( N_0 = 2083; \)
\( N_n = 1332; \)
\( R_n = 1,85 \cdot 10^{-2} \text{ s}^{-1}; \)
\( \varepsilon_{\text{Cs-137}} = 0,03361 \text{ Bq}^{-1} \cdot \text{s}^{-1}; \)
\[ p_{\gamma} = 0.851; \]
\[ b = 10; \]
\[ L = 5; \]
\[ R_0(E_{\gamma}) = 2,89 \cdot 10^{-3} \text{ s}^{-1}; \]
\[ E_{\gamma} = 662 \text{ keV}; \]
\[ t_m = 72000 \text{ s}; \]
\[ V = 25752 \text{ m}^3; \]
\[ t_A = 3,02 \cdot 10^5 \text{ s}; \]
\[ \lambda_{\text{Cs-137}} = 7,26 \cdot 10^{-10} \text{ s}^{-1}; \]
\[ f_1 = 1,00. \]

with

\[ f_2 = e^{7,26 \cdot 10^{-10} \cdot 3,02 \cdot 10^5} = 1,00 \]

and

\[ f_3 = \frac{7,26 \cdot 10^{-10} \cdot 72000}{1 - e^{-7,26 \cdot 10^{-10} \cdot 72000}} = 1,00 \]

The activity concentration of Cs-137 is therefore:

\[ c_{\text{Cs-137}} = \frac{1332}{0,03361 \cdot 0,851 \cdot 72000 \cdot 25752} \cdot 1,00 \cdot 1,00 \cdot 1,00 \text{ Bq} \cdot \text{m}^{-3} = 2,51 \cdot 10^{-5} \text{ Bq} \cdot \text{m}^{-3} \]

and the standard deviation of the net count rate yields:

\[ s(R_n) = \sqrt{\frac{1,85 \cdot 10^{-2}}{72000} + \frac{10 \cdot 2,89 \cdot 10^{-3} \cdot 2}{72000}} \text{ s}^{-1} = 1,03 \cdot 10^{-3} \text{ s}^{-1} \]

This means that the standard deviation of the activity concentration of Cs-137 is:

\[ s(c_{\text{Cs-137}}) = 1,03 \cdot 10^{-3} \cdot \frac{2,51 \cdot 10^{-5}}{1,85 \cdot 10^{-2}} \text{ Bq} \cdot \text{m}^{-3} = 1,40 \cdot 10^{-6} \text{ Bq} \cdot \text{m}^{-3} \]

The final result of this worked example for measuring the activity concentration of Cs-137 therefore reads:

\[ c_{\text{Cs-137}} = (2,51 \pm 0,14) \cdot 10^{-5} \text{ Bq} \cdot \text{m}^{-3} \]
b) Calculation of the activity concentration of the airborne particulate radionuclide I-131:

The calculation of the activity concentration of I-131 in air is based on putting following values in equation (1):

\[ N_g = 5830; \]
\[ N_0 = 3198; \]
\[ N_n = 2632; \]
\[ R_n = 1,07 \cdot 10^{-2} \text{ s}^{-1}; \]
\[ E_{I-131} = 0,0803 \text{ Bq} \cdot \text{s}^{-1}; \]
\[ p_{\gamma} = 0,816; \]
\[ b = 10; \]
\[ L = 5; \]
\[ R_0(E_{\gamma}) = 1,29 \cdot 10^{-3} \text{ s}^{-1}; \]
\[ E_{\gamma} = 364 \text{ keV}; \]
\[ t_m = 247083 \text{ s}; \]
\[ V = 24033 \text{ m}^3; \]
\[ t_A = 655416 \text{ s}; \]
\[ \lambda_{I-131} = 1,00 \cdot 10^{-6} \text{ s}^{-1}; \]
\[ f_1 = 1,00. \]

with

\[ f_2 = e^{1,00 \cdot 10^{-6} \cdot 6,55 \cdot 10^{-5}} = 1,93 \]

and

\[ f_3 = \frac{1,00 \cdot 10^{-6} \cdot 247083}{1 - e^{-1,00 \cdot 10^{-6} \cdot 247083}} = 1,13 \]

The activity concentration of I-131 therefore yields:

\[ c_{I-131} = \frac{2632}{0,08 \cdot 0,816 \cdot 247083 \cdot 24033} \cdot 1,00 \cdot 1,925 \cdot 1,13 \text{ Bq} \cdot \text{m}^{-3} = 1,48 \cdot 10^{-5} \text{ Bq} \cdot \text{m}^{-3} \]

The standard deviation of the net count rate amounts to:

\[ s(R_n) = \sqrt{\frac{1,07 \cdot 10^{-2}}{247083} + \frac{10 \cdot 1,29 \cdot 10^{-3}}{247083}} \cdot 2 \text{ s}^{-1} = 3,84 \cdot 10^{-4} \text{ s}^{-1} \]

This means that the standard deviation of the activity concentration of I-131 is:

\[ s(c_{I-131}) = 3,84 \cdot 10^{-4} \cdot \frac{1,47 \cdot 10^{-5}}{1,07 \cdot 10^{-2}} \text{ Bq} \cdot \text{m}^{-3} = 5,31 \cdot 10^{-7} \text{ Bq} \cdot \text{m}^{-3} \]

The final result of this worked example for measuring the activity concentration of I-131 therefore reads:

\[ c_{I-131} = (1,48 \pm 0,05) \cdot 10^{-5} \text{ Bq} \cdot \text{m}^{-3} \]
5.2 Consideration of uncertainties

The overall uncertainty of individual measuring results is composed of statistical uncertainties in the counting of pulses during the measurement and uncertainties related to calibration and measuring the air volume throughput. The largest contributor will typically be the uncertainty arising from sampling.

Uncertainties resulting from non-representative sampling can only be estimated (see section 2).

6 Characteristic limits of the procedure

In gamma spectrometric analyses of activity, the detection limit of the activity concentration, \( g \), is given by (provided that the background effect pulse count rates are not too small and \( t_0 = t_m \)):

\[
g = \frac{1}{\varepsilon_r \cdot p_r \cdot V} \cdot f_1 \cdot f_2 \cdot f_3 \cdot (k_{1-\alpha} + k_{1-\beta}) \cdot \sqrt{\frac{2 \cdot b \cdot R(\bar{E})}{t_m}}
\]

Applying the quantiles of the normal distribution, \( k_{1-\alpha} = 3,0 \) and \( k_{1-\beta} = 1,645 \), and the values given in section 5.1, equation (6) is evaluated as follows:

a) for Cs-137

\[
g = \frac{1}{0,03361 \cdot 0,851 \cdot 25752} \cdot 1,00 \cdot 1,00 \cdot 1,00 \cdot (3 + 1,645) \cdot \sqrt{\frac{2 \cdot 10 \cdot 2,89 \cdot 10^{-3}}{72000}} \text{ Bq} \cdot \text{m}^{-3}
\]

\[
= 5,65 \cdot 10^{-6} \text{ Bq} \cdot \text{m}^{-3}
\]

This yields a detection limit for Cs-137 of \( 5,65 \cdot 10^{-6} \text{ Bq} \cdot \text{m}^{-3} \).

b) for I-131

\[
g = \frac{1}{0,0803 \cdot 0,816 \cdot 24033} \cdot 1,00 \cdot 1,925 \cdot 1,13 \cdot (3 + 1,645) \cdot \sqrt{\frac{2 \cdot 10 \cdot 1,29 \cdot 10^{-3}}{247083}} \text{ Bq} \cdot \text{m}^{-3}
\]

\[
= 2,07 \cdot 10^{-6} \text{ Bq} \cdot \text{m}^{-3}
\]

This yields a detection limit for I-131 of \( 2,07 \cdot 10^{-6} \text{ Bq} \cdot \text{m}^{-3} \).

The filter will have a high content of radon progeny immediately after sampling. This creates a more intense background. Leaving the filter to cure for some time can reduce this content substantially. For such a case, more examples of achievable detection limits are given in the following table.
Radionuclide | Detection limit, $g$, in Bq·m$^{-3}$
--- | ---
Zr-95 | $4,3 \cdot 10^{-6}$
Nb-95 | $3,1 \cdot 10^{-6}$
Ru-103 | $3,1 \cdot 10^{-6}$
Cs-134 | $2,7 \cdot 10^{-6}$
Ce-141 | $4,9 \cdot 10^{-6}$
Ce-144 | $2,1 \cdot 10^{-5}$

7 Catalogue of chemicals and equipment

7.1 Chemicals
- Glass fibre filter according to DIN 24184;
- Cellulose fibre filter (80 % cellulose / 20 % glass fibre) by Luwa according to DIN 24184;
- PCV filter with cotton support grille;
- Surface filter for calibration.

7.2 Equipment
- Dust sample collector with filter bracket and air volume throughput counter;
- Hydraulic press up to 300 kN;
- Bag sealer;
- Petri-dishes, diameter 110 mm;
- Gamma spectrometry workstation.

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