Monitoring of the radioactivity of airborne particles by γ-ray-spectrometry

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1 Purpose

Regular monitoring of the radioactivity of airborne particles (formerly aerosols) is prescribed in both, the measurement programs of the AVV-IMIS [1] and the Directive on the Monitoring of Emissions and Immissions of Nuclear Plants (REI) [2]. The measurements serve to determine quality and quantity of artificial radioactive nuclides in the environment, and to control the emissions of nuclear plants. The present measurement instructions meet the specific requirements of program point 1.2 of the REI for normal operation. This point requires continuous sampling of radioactive airborne particles in the environment of a nuclear plant and, as a monitoring measure to be carried out every two weeks by the license holder, evaluation by γ-spectrometry of the dust-covered filters in the laboratory. According to program point 1.2, quarterly analyses of mixed samples is a task of the independent measurement centers.

2 Measurand, unit of measurement and limit of detection to be required

2.1 Measurand

The measurand is the activity concentration of long-lived radionuclides attached to airborne particles, averaged over the sampling period.

2.2 Unit of measurement

The unit of measurement is the radioactivity concentration in $Bq \cdot m^{-3}$.

2.3 Required limit of detection

As limit of detection, the REI [2] requires 0.4 $Bq \cdot m^{-3}$, related to ^{60}Co.

3 Method of measurement

3.1 Sampling

The requirements to be met by the site and the sampler for airborn particles are described in LB 3.4.6. The filter used for suspended particles is a filter of class E 12 according to DIN 25184 [3], 6 cm in diameter, through which – during the sampling time (two weeks) – ambient air is continuously pumped with a mean volume flow rate of 1.2 m^3/h (total air flow rate: approx. 400 m^3). The pump maybe, a rotary vane pump for giving an example.

3.2 Sample preparation

To reduce the natural background, caused by short-lived natural radionuclides, they are recommended for decaying. After a decay time of at least three days, the dust-covered filter is measured by direct γ-ray-spectrometry in an appropriate packaging, without any additional treatment. Figure 1 shows the spectra of an aerosol sample measured for approximately 54 000 s after decay times of 1 h, 1 d and after 5 d. The example of the 46 keV line of the ^{210}Pb clearly shows the increase of measurement sensitivity, caused by the decay of the natural radioactivity.

3.3 Measurement by γ-ray-spectrometry and analyzes

The measurement is carried out with a high-purity germanium detector of about 30% relative efficiency (compared with the efficiency of a 3" x 3" NaI(TI)-detector and a ^{60}Co point source at a distance of 25 cm). Filter (sample) and detector are shielded against ambient radiation by a lead coating of approx. 10 cm thickness. A commercial analyzing program is, for example, offered by the manufacturers of the germanium detectors and the multi channel analyzers, can be used to analyze the spectrum, assign the nuclides and determine the radioactivity concentration. The spectrometer has to be calibrated with a multi line calibration source. In addition coincidence summation correction factors have to be taken into account. A measurement time of 54 000 s is scheduled. The value stated by the filter manufacturer is used as the filter collection efficiency. The radioactivity concentration is related to the middle of the sampling period (two weeks). If more detailed information about time and duration of the occurrence of a radioactivity concentration at the place of sampling is available, the measurement results have to be corrected accordantly.

The activity concentration c_r related to the middle of the collection time is calculated according to equation (1):

$$c_r = \frac{N_n}{\varepsilon_r \cdot p_\gamma \cdot t_m \cdot V} \cdot f_1 \cdot f_2 \cdot f_3 \qquad (1)$$

or

$$c_r = \frac{R_n}{\varepsilon_r \cdot p_\gamma \cdot V} \cdot f_1 \cdot f_2 \cdot f_3 \qquad (1a)$$

with

$$R_n = \frac{N_n}{t_m} \qquad (1b)$$

where is:

c_r : activity concentration of the radionuclide r in $Bq \cdot m^{-3}$
N_n : net counts
R_n : net counting rate
ε_r : detection probability for radionuclide r in $Bq^{-1} \cdot s^{-1}$
p_γ : emission probability per nuclear transition
t_m : measuring time for the sample in s

V : air volume passed in m^3
λ_r : decay constant of the radionuclide r
t_A : time between sampling (related to the middle of the sampling time) and start of the measurement in s
t_r : half-life of the radionuclide r in s
f_1 : summation correction factor
f_2 : correction factor for the decay of the radionuclides in the time between sampling and start of the measurement
f_3 : correction factor for the decay of the radionuclides during the measurement

In case of impulse losses and with a point or area source positioned directly on the top of the detector, the summation correction factor f_1 normally reaches from 1.1 to 1.5.

The activity prevailing at a reference time earlier than the start of the measurement by a time t_A, is obtained by multiplication by f_2 according to equation (2):

$$f_2 = e^{\lambda_r \cdot t_A} \tag{2}$$

The following is valid for the correction factor f_3 according to equation (3):

$$f_3 = \frac{\lambda_r \cdot t_m}{1 - e^{-\lambda_r \cdot t_m}} \tag{3}$$

If the time between sampling and measuring time (or the measuring time itself) is much shorter than the half-life of the radionuclides to be measured, the following is valid: $f_2 = 1$ or $f_3 = 1$.

3.4 Analysis of the standard uncertainty of measurement and of the achievable limit of detection

3.4.1 Combined standard measurement uncertainty

The relative combined standard measurement uncertainty of the activity concentration s_G is mainly composed - according to the following equation - of the uncertainty contribution of the sampling process (determination of the air flowrate, perfect filter fit) s_p, the contribution of the calibration s_K and the statistical measurement uncertainty s_S:

$$(s_G)^2 = (s_P)^2 + (s_K)^2 + (s_S)^2 \tag{4}$$

The uncertainty contribution of the sampling process is estimated to be approx. 15%.

The calibration uncertainty is evaluated from the results of the inter-comparisons organized for external quality assurance. It is assumed to be approx. 5 % on average over the whole energy range and should not exceed this value.

The relative statistical uncertainty contribution (s_S) assigned to the activity concentration ensues from equation (5) and depends on the standard measurement uncertainty $s(R_n)$ of the net counting rate R_n.

$$s_s = \frac{s(c_r)}{c_r} = \frac{s(R_n)}{R_n} \tag{5}$$

The following equation is valid for the calculation of the standard measurement uncertainty $s(Rn)$ of the net counting rate R_n:

$$s(R_n) = \sqrt{\frac{R_n}{t_m} + \frac{b \cdot \overline{R_0}(E_\gamma)}{t_m} \cdot \left(1 + \frac{b}{2L}\right)} \qquad (6)$$

where is:

$s(R_n)$: standard deviation of the net counting rate in s^{-1}
R_n : net counting rate in s^{-1}
b : base of the gamma line in number of channels
$R_0(E\gamma)$: mean counting rate of the background per channel in s^{-1}
$E\gamma$: energy of the gamma line
L : number of channels for background determination

3.4.2 Limit of detection and limit of decision

The limit of detection achievable with the measurement procedure is calculated according to DIN 25482, part 5 [4], until implementation of the international standard ISO 11929 [5] is realized. The measurement instructions of the Federal Ministry of the Environment, Nature Conservation and Nuclear Safety [6] and the rules of KTA contain simplified equations which will be used here.

- If the mean counting rate for the background per channel $R_0(E_\gamma)$ provides sufficiently great counting rates

and

- if the following is valid for the relation between base b of the line and width L of the ranges from which the background is determined on the right and left of the line,

$$2L = b, \qquad (7)$$

the limit of detection $G_{N,r}$ for the radioactivity concentration of the radionuclide r is determined according the following equation:

$$G_{N,r} = \frac{1}{\varepsilon_r \cdot p_\gamma \cdot V} \cdot f_1 \cdot f_2 \cdot f_3 \cdot \left(k_{1-\alpha} + k_{1-\beta}\right) \cdot \sqrt{\frac{2b \cdot \overline{R_0}(E_\gamma)}{t_m}} \qquad (8)$$

where is:

$G_{N,r}$: limit of detection for the activity concentration of the radionuclide r in air in Bq/m^3
$k_{1-\alpha}$, $k_{1-\beta}$: quantils of the standardized normal distribution
ε_r : detection probability for the radionuclide r in $Bq^{-1} \cdot s^{-1}$
p_γ emission probability for the gamma line with energy E_γ of the radionuclide r observed
V . air volume having passed through the filter in m^3
$R_0(E_\gamma)$: mean counting rate of the background per channel
b : base of the line
L : width of the ranges from which the background is determined on the left and right of the line
t_m : measuring time in s
$G_{E,r}$: limit of decision for the activity concentration of the radionuclide r in air in Bq/m^3

The limit of decision $G_{E,r}$ of the activity concentration of the radionuclide r is calculated according to equation (9) as follows:

$$G_{E,r} = [k_{1-\alpha}/(k_{1-\alpha} + k_{1-\beta})] \cdot G_{N,r} \quad (9)$$

3.4.3 Arithmetic example

For the γ-ray-spectrometer described above, the limit of detection and the limit of decision of the reference nuclide ^{60}Co can be calculated with the following parameters:

$k_{1-\alpha}$	:	3.0 (99.9 % statistical certainty of the normal distribution)
$k_{1-\beta}$	:	1.65 (95 % confidence level of the normal distribution)
ε_r	:	$1.96 \cdot 10^{-2}$ (detection probability for ^{60}Co)
p_γ	:	0.999826 (emission probability for the line with the energy of 1332.50 keV of nuclide ^{60}Co)
V	:	400 m^3 (air flowrate through the filter)
t_m	:	54 000 s (measuring time)
$R_o(E_\gamma)$	:	$1.5 \cdot 10^{-3}$ pulses per second and channel, estimated
b	:	4 channels (base of the line, with 2 k spectra)
L	:	2 channels (width of the ranges from which the background is determined)
f_1	:	1
f_2	:	1
f_3	:	1

The following results for the limit of detection according to equation (8):

$$G_{N,Co\text{-}60} = 0.28 \text{ mBq/m}^3$$

The following results for the limit of decision according to equation (9)

$$G_{E,Co\text{-}60} = 0.18 \text{ mBq/m}^3$$

4 Evaluation of the method

The procedure proposed allows a relatively straightforward metrological determination of the radionuclide concentration for artificial radionuclides. For higher air flow rates, the limit of detection can be reduced. Use of a larger filter may, however, negatively influence the measurement geometry. This must be checked in each individual case.

An indispensable prerequisite for this monitoring measure is the installation of a sampling system for airborne particles. The required γ-ray-spectrometer set-up should be designed that way to be used for all other measurement tasks.

In case of sampling systems which do not continuously measure the air flow rate, the uncertainty due to the air flow rate through the filter which is not always constant may contribute the greatest contribution to the combined standard measurement uncertainty. The contribution to the uncertainty assigned to the sampling process can basically be attributed to the varying humidity and to different dust contents of the air.

The working time required mainly results from the measuring times for both the sample and the background. Compared with this time, the time required for filter change and measurement evaluation is negligible.

The ubiquitous activity caused by natural occurring radionuclides is normally much higher than the activity caused by artificial radionuclides. To achieve a lower limit of detection a previous decay of the short-lived natural activity is recommended. The decay time can differ from the three days according to section 3.2 for meeting operational requirements. This has an effect on the achievable limit of detection and therefore has to be properly taken into account.

As an example, Table 1 shows the limits of detection for some radionuclides in Bq/sample obtained after measuring times of 54 000 s. The measurements were started after decay times of 1 h, 1 d and 5 d, respecively.

The proposed measuring time of 54 000 s as used in the example is an empirical value. According to the above calculation, a somewhat shorter measuring time would also be sufficient to obtain the limit of detection required according to REI.

The filter may also be used to determine the α- and β- radioactivity concentration.

Table 1: Limits of detection in Bq/sample, achieved by the measurements shown in Figure 1.

Radionuclide	Measurement after 1 h	Measurement after 1 d	Measurement after 5 d
^{51}Cr	3.28	1.65	1.21
^{51}Mn	0.30	0.12	0.079
^{60}Co	0.37	0.14	0.081
^{131}I	0.49	0.27	0.22
^{137}Cs	0.39	0.17	0.060
^{40}K	4,3	1.8	1.4
^{210}Pb	49	23	12

5 Documentation

The sampling place, the date and time of filter change, the air volume, the decay time, the date and time of the measurement have to be recorded, for instance on an accompanying protocol sheet or in a dedicated pocketbook.

According to REI, the quarterly reports must contain place, start and end of the sampling period, decay time and measurement result, including the statistical standard measurement uncertainty.

The filters should be kept available until the end of the subsequent year for potential control measurements. The inscription on the retained sample must allow for positive identification of the sample stored, including storage date and activity concentrations of the radionuclides detected, e. g. sample number, sampling period, activity concentrations at reference time. With regard to the disposal of the retained particle filters, it is recommended keeping them separate from other samples which need to be permanently stored.

As far as the safekeeping period of particle filters is concerned, the following concept is usually applied:

Particle filters from the calendar year 2000, for example, are discarded in January 2002. This guarantees that particle filters from December 2000 will be available for one more complete year, whereas particle filters from January 2000 would be stored for almost two years.

According to REI, the recorded measurement results must be stored for 30 years.

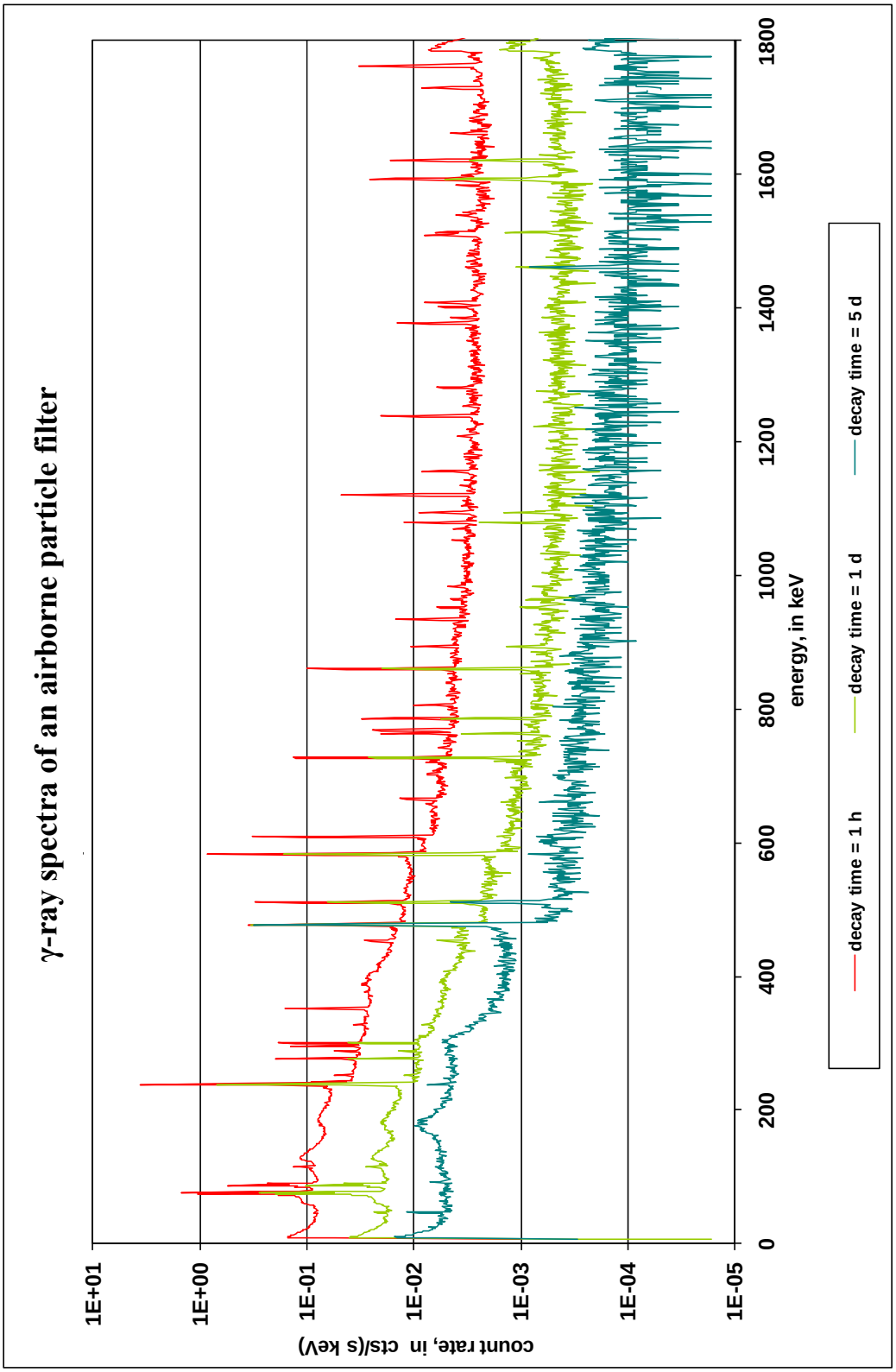

Figure 1: Spectra of an airborne particle filter measured for 54 000 s after decay times of 1 h, 1 d and 5 h.

6 Literature

[1] Federal Ministry for the Environment, Nature Conservation and Nuclear Safety: General administrative instruction regarding the Integrated Measuring and Information System according to the Law on Preventive Radiation Protection (AVV-IMIS) with Annexes 1 and 2, Federal Law Gazette, volume 47, number 200a, October 24, 1995. (only in German language)

[2] Federal Ministry for the Environment, Nature Conservation and Nuclear Safety: Directive for emission and immission monitoring of nuclear plants (REI), GMBI. 2006 No. 14-17 p. 254 et sqq. (only available in German language)

[3] DIN EN 1822-1 Schwebstofffilter (EPA, HEPA und ULPA) - Teil 1: Klassifikation, Leistungsprüfung, Kennzeichnung; Deutsche Fassung EN 1822-1:2009 (in German language)

[4] DIN 25 482, part 5: Limit of detection and limit of decision in nuclear radiation measurement – Counting high-resolution gamma-spectrometry measurements, leaving the influence of sample treatment out of account, June 1993 (in German language)

[5] ISO 11929: Determination of the characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation – Fundamentals and application

[6] Federal Ministry for the Environment, Nature Conservation and Nuclear Safety: Measurement instructions for the monitoring of radioactivity in the environment and external radiation, ISSN 1865-8725, online available at http://www.bmu.de/strahlenschutz/ueberwachung_der_umweltradioaktivitaet_/messanleitungen/doc/42042.php

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Please refer to the note in the loose leaf 1.3 "Notes to the loose-leaf collection".