

**Procedure for determining the activity
concentrations of gaseous iodine compounds
in air near the ground
by gamma spectrometry**

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1 Scope

The procedure described here facilitates determining the nuclide-specific activity concentration of gaseous iodine radionuclides via their gamma emission by means of directly measuring a sample following its enrichment on an adsorbent. In the simplest case, no difference needs to be made between gaseous, elementary iodine (I_2) and organic iodine compounds (e. g., CH_3I). The procedure conforms to the requirements for measuring programmes of the AVV-IMIS [1] and the directive for the monitoring of emissions and immissions of nuclear installations (REI) [2].

2 Sampling

A pump is used to extract airborne particulate from the air by first channelling the airborne particulate over a filter for suspended particles and then, for the enrichment of the gaseous radionuclides of iodine, over activated charcoal impregnated with potassium iodine. The adsorbent is housed in a cylindrical stainless steel cartridge (Fig. 1).

The air throughflow amounts to ca. $1 \text{ m}^3 \cdot \text{h}^{-1}$ to $4 \text{ m}^3 \cdot \text{h}^{-1}$. This low throughflow is necessary to facilitate a sufficiently long period of contact of more than 0,2 seconds between the iodine compounds contained and the adsorbent. This is intended to ensure their being completely adsorbed. In order to prevent condensation and to improve the degree of effectiveness of the filter, the supply air needs to be heated. The degree of separation for iodine and methyl iodide is $> 99,9 \%$ at a temperature of $45 \text{ }^\circ\text{C}$ and a relative humidity of 77% . In order to reduce adsorption losses, a short inlet pipe made from a suitable material (e. g., stainless steel piping or plastic hose with a smooth surface) is to be used.

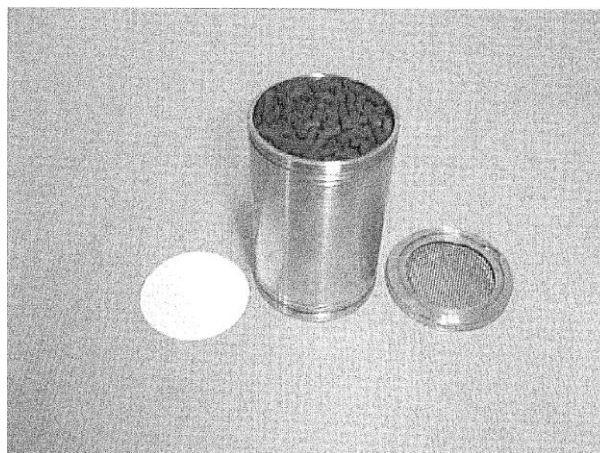


Fig. 1: Iodine adsorber cartridge

If the collection period is long, at least two cartridges should be installed in series. The posterior cartridges are only analysed if active iodine has been detected in the first cartridge. This serves to determine the degree of retention of the adsorbent used.

Employing adsorbents of different selectivity enables both activity concentration of gaseous elementary iodine and the activity concentration of gaseous organically compounded iodine to be determined

3 Analysis

3.1 Principle of the procedure

The gaseous iodine compounds are selectively collected on the surface of an adsorbent. Radionuclides attached to airborne particulate have previously been extracted on a filter for suspended particles.

3.2 Sample preparation

The contaminated adsorbent is transferred to a suitable measuring container (with dimensional stability to 60 °C) and homogenised. This needs to be done with care, as the cylindrical stainless steel cartridge may become hot.

3.3 Radiochemical separation

No radiochemical separation is required for the procedure described here.

4 Measuring the activity

4.1 General

The activity of the iodine isotopes attached to the adsorbent is measured by means of gamma spectrometry. An ultrapure germanium detector with a relative efficiency of at least 40 % should be employed. Here, the 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 < 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 X-ray radiation of lead at 76 keV and 85 keV, and Perspex that is meant to absorb the beta particles that disperse on the sheet of copper.

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

Measuring processes with lower sensitivities may require a less elaborate outfitting.

4.2 Calibration

Calibration is effected with a special gelatinous standard mix calibration source that contains several radionuclides of known activity. This standard mix calibration source has to have the same geometry and size as the sample container accommodating the adsorbent. Owing to the differences between the chemical composition of the calibration source and that of the sample material, a self-absorption correction needs to be applied.

5 Calculation of the results

The activity concentration, c_r in the centre of the accumulation period is calculated according to equation (1):

$$c_r = \frac{N_n}{\varepsilon_r \cdot p_\gamma \cdot \eta_\gamma \cdot t_m \cdot V} \cdot f_0 \cdot f_1 \cdot f_2 \cdot f_3 \quad (1)$$

with

$$f_2 = e^{\lambda_r \cdot t_m} \quad (2)$$

and with

$$f_3 = \frac{\lambda_r \cdot t_m}{1 - e^{-\lambda_r \cdot t_m}} \quad (3)$$

where:

- c_r activity concentration of the radionuclide, r , in $\text{Bq} \cdot \text{m}^{-3}$;
- N_n number of net pulses;
- ε_r detection efficiency of the radionuclide, r , in $\text{Bq}^{-1} \cdot \text{s}^{-1}$;
- p_γ emission probability per nuclear transformation;
- η_γ degree of retention by the adsorbent;
- t_m duration of sample measurement, in s;
- V processed air volume, in m^3 ;
- λ_r decay constant of the radionuclide, r , in s^{-1} ;
- t_A period from the centre of the sampling period to the commencement of measuring, in s;
- f_0 correction factor for the differences in self-absorption between the calibration source and the sample;
- f_1 summation correction factor;
- f_2 correction factor for the decay of radionuclides within the period from the middle the sampling period to the commencement of measuring;
- f_3 correction factor for the decay of the radionuclides during the measuring process.

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

If the period between the sampling and the measurement or the measurement period 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 (4):

$$s(R_r) = \sqrt{\frac{R_n}{t_m} + \frac{b \cdot \overline{R}_0(E_\gamma)}{t_m} \cdot \left(1 + \frac{b}{2 \cdot L}\right)} \quad (4)$$

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 number of channels;
 E_γ energy of the gamma line;
 $R_0(E_\gamma)$ mean background count rate per channel around gamma energy E_γ , in s^{-1} ;
 L number of channels used for determining the background effect.

For the standard deviation of the activity concentration, $s(c_r)$, equation (5) applies:

$$s(c_r) = s(R_n) \cdot \frac{c_r}{R_n} = s(R_n) \cdot \frac{1}{\varepsilon_r \cdot p_\gamma \cdot \eta_\gamma \cdot V} \cdot f_0 \cdot f_1 \cdot f_2 \cdot f_3 \quad (5)$$

5.1 Worked example

For calculating the activity concentration of the totality of the radionuclide I-131 content that is present in gaseous form, the following numerical values of a sample, collected over a period of 7 days, are inserted:

N_n	= 1200;	$\bar{R}_0(E_\gamma)$	= 0,022 s^{-1} ;
R_n	= 0,014 s^{-1} ;	ε	= 0,0299 $Bq^{-1} \cdot s^{-1}$;
p_γ	= 0,816;	η_γ	= 1;
b	= 10;	L	= 5;
E_γ	= 364,5 keV;	t_m	= 86400 s;
V	= 431 m^3 ;	t_m	= 302404 s;
λ	= $1,00 \cdot 10^{-6} s^{-1}$;	f_0	= 0,872;
f_1	= 1.		

with

$$f_2 = e^{1,00 \cdot 10^{-6} \cdot 302404} = 1,35$$

and

$$f_3 = \frac{1,00 \cdot 10^{-6} \cdot 86400}{1 - e^{-1,00 \cdot 10^{-6} \cdot 86400}} = 1,044$$

The activity concentration of I-131 activity therefore shows as:

$$\begin{aligned} c_{I-131} &= \frac{1200}{0,0299 \cdot 0,816 \cdot 1 \cdot 86400 \cdot 431} \cdot 0,872 \cdot 1 \cdot 1,35 \cdot 1,044 \text{ Bq} \cdot m^{-3} = \\ &= 1,62 \cdot 10^{-3} \text{ Bq} \cdot m^{-3} \end{aligned}$$

and the standard deviation of the net count rate amounts to:

$$s(R_n) = \sqrt{\frac{0,014}{86400} + \frac{10 \cdot 0,022}{86400} \cdot 2} \text{ s}^{-1} = 2,29 \cdot 10^{-3} \text{ s}^{-1}$$

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

$$s(c_{\text{I-131}}) = 2,29 \cdot 10^{-3} \cdot \frac{1,62 \cdot 10^{-3}}{1,39 \cdot 10^{-2}} \text{ Bq} \cdot \text{m}^{-3} = 2,67 \cdot 10^{-4} \text{ Bq} \cdot \text{m}^{-3}$$

The activity concentration of I-131 therefore yields

$$c_{\text{I-131}} = (1,6 \pm 0,3) \cdot 10^{-3} \text{ Bq} \cdot \text{m}^{-3}$$

5.2 Consideration of uncertainties

The overall uncertainty of individual measuring results is mainly composed of statistical counting errors and uncertainties related to calibration and measuring the volume throughput. The total uncertainty for the above calculation example is about 16 % with regard to statistical counting errors.

6 Characteristic limits of the procedure

In gamma spectrometric analyses of activity, the detection limit of the activity concentration, g , is given by equation (6):

$$g = \frac{(k_{1-\alpha} + k_{1-\beta})}{\varepsilon_r \cdot p_\gamma \cdot \eta_\gamma \cdot V} \cdot f_0 \cdot f_1 \cdot f_2 \cdot f_3 \cdot \sqrt{\frac{b \cdot \bar{R}_0(E_\gamma)}{t_m} \cdot \left(1 + \frac{b}{2 \cdot L}\right)} \quad (6)$$

where:

g detection limit of the activity concentration, in $\text{Bq} \cdot \text{m}^{-3}$;

$k_{1-\alpha}$ quantile of the normal distribution;

$k_{1-\beta}$ quantile of the normal distribution.

Applying the quantiles of the normal distribution, $k_{1-\alpha}=3$ and $k_{1-\beta}=1,645$, and the numerical values used in section 5.1, the following detection limit for the activity concentration of I-131 is obtained:

$$g = \frac{(3 + 1,645)}{0,02987 \cdot 0,816 \cdot 1 \cdot 431} \cdot 0,872 \cdot 1 \cdot 1,35 \cdot 1,044 \cdot \sqrt{\frac{2 \cdot 10 \cdot 0,022}{86400}} \text{ Bq} \cdot \text{m}^{-3} =$$

$$= 1,2 \cdot 10^{-3} \text{ Bq} \cdot \text{m}^{-3}$$

The detection limit for the activity concentration of I-131 amounts to $1,2 \cdot 10^{-3} \text{ Bq} \cdot \text{m}^{-3}$.

7 Catalogue of chemicals and equipment

7.1 Chemicals

Activated charcoal KI3-P, granulate of small rods with a diameter of 3,5 mm and a length of 4 mm to 10 mm, coated with potassium iodide 3 %.

Filter for suspended particles: solidification agent-free fibreglass filter, Class S according to DIN 24184 or superior to H12 according to DIN EN 1822-1, degree of separation for elementary iodine < 1 % and for methyl iodide likewise < 1 %.

7.2 Equipment

- Pump with preheating for supply air with a capacity of ca. $1 \text{ m}^3 \cdot \text{h}^{-1}$ to $3 \text{ m}^3 \cdot \text{h}^{-1}$ in throughflow volume
- Cylindrical stainless steel cartridge with an inner volume of 170 ml (diameter: 50 mm, length: 200 mm) for accommodating the iodine adsorbent, with support grille for a pre-filter
- 250 ml-measuring vessel from temperature-tolerant plastic
- Gamma spectrometry station including data evaluation computer

References

- (1) German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (ed.): Allgemeine Verwaltungsvorschrift zum Integrierten Mess- und Informationssystem nach dem Strahlenschutzvorsorgegesetz (AVV-IMIS) (Common Administrative Procedure on the Integrated Measuring and Information System according to the Precautionary Radiation Protection Act), Federal Gazette 47, Number 200a dated 24. October 1995
- (2) German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (ed.): Richtlinie zur Emissions- und Immissionsüberwachung kerntechnischer Anlagen (Guideline concerning Emission and Immission Monitoring of Nuclear Installations) GMBI. 44, dated 19. August 1993
- (3) Standard DIN 24184 Typprüfung von Schwebstofffiltern (Type testing of high efficiency particulate air filters). 1990-12
- (4) Standard DIN EN 1822-1 Schwebstofffilter (HEPA und ULPA) (High efficiency air filters (HEPA and ULPA)). 1998-07