Applied Radiation and Isotopes 81 (2013) 284–289

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Remarks on representative ground-level air monitoring

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HIGHLIGHTS

- Activity ratios of ^{7}Be, ^{22}Na, ^{40}K, ^{137}Cs, and ^{210}Pb are determined.
- Results variability is discussed by use of two samples per sampling period.
- Comparability and reliability are extended to ^{22}Na, ^{40}K, ^{137}Cs, and ^{210}Pb.

ARTICLE INFO

Available online 21 March 2013

Keywords:
Representativeness
^{7}Be
^{22}Na
^{40}K
^{137}Cs
^{210}Pb

ABSTRACT

The methods applied and the typical influences affecting representative measurements of radionuclides contained in ground-level aerosols are summarized. The activity concentrations of ^{7}Be, ^{22}Na, ^{40}K, ^{137}Cs, and ^{210}Pb were determined in weekly collected samples and the ratio "sample A/sample B" was calculated over a period of 5 years. The results are compared with findings from literature. The estimation of the limits of comparability and reliability were extended to ^{22}Na, ^{40}K, ^{137}Cs, and ^{210}Pb measurements.

1. Introduction

The trace survey station of Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig for monitoring ground-level aerosols is operated within the framework of the German Federal Precautionary Radiation Protection Act "Strahlenschutzvorsorgegesetz" (BGBl, 2003) which prescribes an "Integrated Measurement and Information System" (IMIS). Within the scope of IMIS, "trace survey stations" for measurements of airborne radioactivity are in operation in order to measure mean activity concentrations of less than 100 μBq/m^3. The German environmental monitoring program is described in Bundesanzeiger, 2006.

After the Fukushima Daiichi Nuclear Power Plant was hit by the Tohoku earthquake and the following tsunami on 2011-03-11, the radionuclides released there arrived at ground-level air in Germany on 2011-03-23. On 2011-03-22, the trace survey station at PTB and three more German trace survey stations changed the operation mode to an intensified mode with daily aerosol sampling and daily reporting. The daily results of the γ-ray spectrometric measurements elaborated at PTB were jointly reported to the public on an internet site operated by BfS (BfS, 2011). More results can be found on the internet (DWD, 2011; PTB, 2011) or in the literature (Bieringer et al., 2011; Masson et al., 2011).

Lively public interest resulted in the results themselves but also in the methods and materials applied. Later on, some experts as well as concerned citizens wondered about both the reliability and the agreement of the results elaborated at different institutions located at such large distances to each other. That discussion motivated a re-evaluation of a measurement series available at PTB – actually intended for internal purposes only – with respect to reasons and effects influencing the representativeness/variability of ground-level aerosol samples collected at a specific sampling site. That was achieved by means of two individual routine weekly samples which had been collected simultaneously on the same site under the same operational and environmental conditions over a period of five years.

Representative measurements require ideal operating conditions, e.g. unhindered air flow from all directions and 100% aerosol collection efficiency. In most cases, however, these conditions are not given in reality. The technical design of the aerosol samplers is not the same and different types of filter materials are in use. Therefore, within the framework of the German IMIS, regular participation in intercomparisons (e.g. Wershofen et al., 2008) is requested.

Previous investigations on the reliability of ground-level aerosol monitoring can be found in the literature (e.g. Vintersved, 1994; Irshad et al., 2006; Le Petit et al., 1998). Vintersved investigated the use

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0969-8043/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.apradiso.2013.03.030

of a mobile high-volume reference aerosol sampler for an intercomparison under operating conditions at nine different sampling sites. The acceptable deviation between the samples collected simultaneously at a given site was set to ±15%. ^{7}Be was found to be the most suitable radionuclide for such intercomparisons. Most of the deviations observed were in the range ±50% but reached −80% or +55% in very few cases.

The PTB case study presented below was elaborated by means of routine weekly data from a period of about five years. A comparison of the results originating from two individual samples which were collected simultaneously under nearly the same operating conditions on the same sampling site was performed. The way from raw data to a final set of plausible data which was elaborated by rejection of data affected by either known malfunctions (e.g. of the volume counting devices) or by observed deviations caused e.g. by agricultural or technological activities close to the sampling site. If one has only one sample, there is no chance to compare and, therefore, to get any idea on any influences affecting the representativeness or variability of the samples. Especially, short-range, local effects cannot be seen. This might become important if samples need to be compared with other samples which were taken on the other sampling sites e.g. operated within a national monitoring network or if one wants to compare samples for radioecological investigations.

The final set of plausible results allows estimation of the limits of representativeness of measured activity concentrations of ^{7}Be, ^{22}Na, ^{40}K, ^{137}Cs, and ^{210}Pb in ground-level air.

2. Materials and methods

2.1. Aerosol sampling

The two weekly aerosol samples were collected simultaneously with two high-volume aerosol samplers located at a distance of about 10 m to each other. They have practically the same aerosol collection properties (effective filter area: 54 cm × 54 cm; nominal air-flow rate: 900 m^3/h) resulting in routine sample volumes between 120.000 m^3 and 140.000 m^3. The operating data are recorded and stored hourly. The filter material has an average aerosol collection efficiency of about 95% for the aerosol size fraction of interest (Wershofen, 2001). The real particle size distribution of the air dust was not determined. The ash content of the filter material is 0.1% by mass (about 10 mg per filter).

2.2. Measurement procedure

Details on the procedures are described in Wershofen and Arnold (2005). For measurements of radioactive iodine isotopes, the dust-loaded filter is folded to fit into a 1-l Marinelli beaker. Typical limits of detection achieved for ^{131}I are in the range of 1–5 μBq/m^3. After that, the sample is ashed at 450 °C and the aerosol ash is pressed to pellets. A subsequent γ-ray spectrometric measurement allows the determination of mean weekly activity concentrations of ^{137}Cs between 0.01 μBq/m^3 and 0.1 μBq/m^3. For the purpose of this investigation, the nuclide-specific results of the ash measurements of sample A and sample B were compared by the calculation of the ratio “sample A/sample B”.

2.3. Measurement uncertainties

The combined standard measurement uncertainties were calculated according to ISO/BIPM (2008). The main contribution when measuring a dust-loaded aerosol filter in a Marinelli beaker is about 10% caused by the difference between the aqueous calibration source and a real aerosol filter. When measuring aerosol ash pellets in a well-type detector, efficiency transfer calculation, self-attenuation correction and true coincidence correction factors e.g. for radionuclides like ^{22}Na or ^{134}Cs need to be calculated with the software GESPECOR (Arnold and Sima, 2006). The uncertainties of these corrections strongly depend on the precise knowledge of the detector properties. Unfortunately, these data of the up to 20-year-old detectors were not known very precisely. Therefore, these uncertainties were estimated to be 5–7% by comparing calculated and measured correction factors. Regular cross-checks by measurement of a particular aerosol ash sample with both the spectrometers revealed agreement within the limits of uncertainties for the more easily measurable

Table 1
Typical relative standard measurement uncertainties (%).

Contribution	Sample A	Sample B
Aerosol collection efficiency under operating conditions	4.2	4.2
Air volume measurement under operating conditions	0.8	2.7
Air volume correction to reference volume (use of external meteorological data)	1.5	1.5
Difference in geometry “calibration/aerosol filter” in 1-l Marinelli beaker (only ^{131}I and daily filter measurements!)	10.0	10.0
Statistical measurement uncertainty of ^{131}I in Marinelli beaker	0.5–10	0.5–10
Yield of aerosol ashing and pelletizing	1.5	1.5
Statistical measurement uncertainty in well-type detector depending on radionuclide, activity and acquisition time	0.5–20	0.5–20
Efficiency transfer “calibration/real sample”	1.5	1.0
True coincidence summing correction	5.0	4.0
Self-attenuation correction	1.5	1.0
Decay data; background subtraction	2–5	2–5

Table 2
Range of activity concentration and relative standard measurement uncertainty observed.

Radionuclide	Activity concentration in air (μBq/m^3)			Combined uncertainty *A* (%)	Combined uncertainty *B* (%)
^{7}Be	(940 ± 68)	≤a(^{7}Be)	≤(7821 ± 339)	2.3–8.9	1.8–9.2
^{22}Na	(0.08 ± 0.02)	≤a(^{22}Na)	≤(1.01 ± 0.05)	3.1–34	4.2–32
^{40}K	(0.85 ± 0.03)	≤a(^{40}K)	≤(117.9 ± 0.3)	2.9–15	2.9–16
^{137}Cs	(0.05 ± 0.01)	≤a(^{137}Cs)	≤(4.6 ± 0.4)	2.2–19	2.1–22
^{210}Pb	(49 ± 0.03)	≤a(^{210}Pb)	≤(2443 ± 117)	2.6–14	2.1–11