Comparison of different sampling methods for the determination of low-level radionuclides in air

MA. Duch*, I. Serrano, V. Cabello, A. Camacho

Institut de Tècniques Energètiques, Universitat Politènica de Catalunya (INTE-UPC), Diagonal 647, 08028 Barcelona, Spain

Abstract

The aim of this work is to check the consistency of results given by different air dust samplers (flow-rates between 2 and 700 m^3/h) and measurement protocols at a single location. The study is focussed on ^{210}Pb since is the only nuclide that can be easily assessed through all the studied sampler types. Results from high- and mid-volume samplers agreed well to within the associated uncertainties. Gross beta activity from low-volume samplers can be used as a good indicator of the evolution of ^{210}Pb concentration in air.

Keywords: Air samplers, Natural radionuclides, Gamma-spectrometry, Calibration

* Corresponding author. Tel.: + 34 93 401 19 69; fax: + 34 93 401 71 49.
E-mail address: maria.amor.duch@upc.edu (MA. Duch).

1. Introduction

Natural radionuclides such as ^{7}Be and ^{210}Pb are commonly used for atmospheric radioactivity studies with different aims (Azahra et al, 2004; Vallés et al, 2009) and very often involve different locations and sampling methods (Bourcier et al, 2011; Tossiti et al, 2004). Comparing results from different sampling stations and laboratories is difficult since there is a lack of a widely accepted protocol for aerosol sampling, and, consequently, different types of samplers, filter materials and protocols are used.

The most commonly used samplers have flow-rates of approximately 2 m^3/h, followed by mid-volume samplers with a flow-rate up to 90 m^3/h, and finally there are available high-volume samplers with average flow-rates between 300-1000 m^3/h. Due to the small amount of collected dust by low-volume samplers, filters from these stations usually undergo rapid measurement methods using proportional counters providing gross beta activity results. Afterwards, samples composed of various filters can be assessed by gamma-spectrometry (Dueñas, 2004), but just providing an average concentration value of the whole studied period (usually 1-3 months). On the other hand, filters from mid- and high-volume samplers are usually assessed by gamma-spectrometry. ^{7}Be is the radionuclide that shows the highest concentration in air (1-5 mBq/m^3) and can be easily measured through all types of air samplers, as well as ^{210}Pb (0.2-0.7 mBq/m^3), but it is worth mentioning that ^{7}Be disintegrates by electron capture with subsequent gamma emission of ^{7}Li and cannot be assessed by beta counting. Other beta-gamma nuclides, such as ^{22}Na, ^{40}K, ^{137}Cs are also present in the atmosphere but at lower concentrations in such a way that require high-volume samplers to be measured. Thus, only ^{210}Pb (beta-gamma emitter) could be measured

either by using beta counters and gamma-spectrometry in filters from all air sampler types.

The quantity commonly measured in inter-laboratory comparison exercises for quality control purposes is the total activity of a particular filter (Wätjen et al, 2008; Wershofen et al, 2008). However, even in this case, production of suitable reference sources remains a challenge (Montsanglant et al, 2015). In addition, these inter-laboratory exercises do not take into account the whole process, since in a realistic case, meteorological conditions or properties of the sampler can change during the sampling period, thus influencing the results.

The aim of this work is to check the consistency of results given by different air dust samplers (flow-rates between 2 and 700 m^3/h) and measurement protocols (gamma-spectrometry and gross beta assessment). The samplers were exposed to the same meteorological conditions and airborne particulates concentration at a single location, at the premises of the Radioactivity Analysis Laboratory of the INTE-UPC. The study is focussed on ^{210}Pb since is the only nuclide that can be assessed using either gamma-spectrometry or gross beta results. ^{210}Pb is usually assessed by using gamma-spectrometry through its 46.5 keV gamma-line, and thus the applied calibration method could strongly influence the results. This work describes a comparison of different calibration methods for the determination of ^{210}Pb in filters by gamma-spectrometry.

2. Materials and methods

2.1 Sampling procedures

Samples of air aerosols were collected weekly at the premises of our Institute, located in Barcelona (Spain), latitude 41º23'05.8''N, longitude 2º7'02.3''E. Three different types of samplers were tested.

The nominal airflow rate at each sampling station was 700 m^3/h (ASS-500 station), 60 m^3/h (MCV sampler) and 2 m^3/h respectively (F&J Sampler). Both high- and mid-volume samplers are equipped with a calibrated internal flow-meter and the total air volume sampled is recorded automatically by the system. The flow-rate of the low-volume station was daily checked with an external flow-meter, and the total air volume sampled was calculated taking into account the sampling time.

G3 polypropylene filters were used both for high- and mid-volume samplers, whereas Whatman GF/A glass microfiber filters were used for the low-volume sampler. The corresponding filter size was (44 x 44) cm^2 for the high-volume sampler, (20 x 25) cm^2 for the mid-volume sampler and a 4.7 cm diameter filter was used for the low-volume sampler.

2.2 Measurement system

Filters from the high-volume samplers were folded and pressed to obtain a surface area of about 8x8 cm^2 with the active area facing inwards. The filters from mid-volume samplers had a final surface area of about 6x6 cm^2. In both cases, the filters were fitted onto square plastic boxes. Measurements were done by gamma spectrometry using two Canberra hyperpure germanium (HPGe) coaxial detectors model GX4020 and GX3020, equipped with a cryostat with a Carbon Epoxy window and a cryostat with a Be window, respectively. The detectors are located in a room

with 1-m-thick walls, and are shielded with 10.5 cm of lead plus 2 mm of copper (GX4020 detector), and 14.4 cm of iron (GX3020 detector). Their nominal efficiencies are 41 % and 33 % respectively, and the resolutions are 1.86 and 1.77 keV at 1.33 MeV of ^{60}Co. The acquisition times ranged from 2 to 4 days.

Gross beta activity of the filters from the low-volume sampler was measured with an argon-methane gas flow low-level proportional counter (Berthold, model LB770-2), calibrated using a ^{90}Sr/^{90}Y standard (dust-loaded filter), with a counting time of 1 day.

2.3 Calibration procedures for ^{210}Pb determination

Different standards were set up for (44 x 44) cm^2 filters for the determination of ^{210}Pb content:

- Type SPK1: Two filters were spiked with a mixed-gamma-ray standard solution containing ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{137}Cs, ^{88}Y, ^{60}Co, energy range 59.5 to 1332.5 keV. Subsequently, the mean full energy peak efficiency was calculated as a function of gamma-ray energy using Genie® 2000 software from the two filters, and a logarithmic polynomial fit was established and validated (Camacho et al, 2014):

$$\ln \varepsilon(E) = \sum_{i=0}^{N} a_i \ln(E)^i$$

where ε is the efficiency, E is energy, a_i are the fitted parameters and N is the degree of the polynomial. Two such equations are used (one for the low energy range and another one for the high energy range). Summing-coincidence corrections are noticeable for some high-energy peaks (^{88}Y, ^{60}Co),

but were considered to be negligible in the low-energy range up to 122 keV (Gilmore, 2008).

- Type SPK2: Two filters were spiked with a standard solution of ^{210}Pb, and the corresponding efficiency was calculated as a mean of the results from the two filters.
- Type PRM: A representative amount of a phosphogypsum reference material (PRM) available in powder form was spread onto a filter previously sprayed with an adhesive aerosol.

For mid-size filters (20 x 25) cm^2, Type SPK1 and Type PRM standards were set up.

The uncertainties assigned to the efficiency results were calculated in accordance with the "Guide to the Expression of Uncertainty in Measurement" (GUM) (JCGM 100:2008). They include the uncertainties of the nuclear decay data, background and blank correction, counting statistics, uncertainty of the fit and the uncertainty of the activity concentration of the standard. The global uncertainties assigned to the results of aerosol measurements include the uncertainties of measurement of air volume, nuclear decay data, background and blank correction, counting statistics, counting geometry and efficiency calibration data.

To compare the different calibration procedures, the ^{210}Pb activity of the standards containing ^{210}Pb was subsequently assessed according to the established calibration procedures and compared with the reference value.

3. Results

Table 1 and Table 2 show the ^{210}Pb activity determined according to the different calibration procedures studied and the corresponding reference value. Comparison of the different calibration approaches for gamma spectrometry of the filters led to differences of up to 14% between the activities calculated using a calibration curve using a mix of different radionuclides (59.5 to 1332.5 keV) versus the use of a single ^{210}Pb nuclide standard. As regards the use of the phosphogypsum reference material, there was good agreement (differences < 3%) with the efficiency calculated with a ^{210}Pb single isotope standard solution. However, special care should be taken when using the phosphogypsum standard for higher energies (García-Talavera et al, 2001).

In addition, results from standards spiked with a mixed-gamma-ray standard solution revealed that the measurement efficiency for mid-size filters was 16% higher than for the large filters, mainly due to a higher self-attenuation of high-volume filters.

Results of ^{210}Pb concentration in air from high- and mid-volume samplers are shown in Figure 1. As can be observed, there was good agreement between the two sampling systems, within the associated combined uncertainties (20%, k=2).

Figure 2 shows the gross beta results obtained from the low-volume sampler compared with ^{210}Pb activity measured from the high-volume sampler.

The measured gross beta activity showed a strong correlation (Coefficient of determination, R-squared=0.99) with ^{210}Pb activity, but the gross beta activity was systematically higher due to the presence of other beta emitters such as ^{22}Na, ^{40}K, ^{137}Cs or ^{210}Bi. The obtained results are in agreement with the results of other authors that suggest that about 61-87 % of the gross beta activities are contributed by ^{210}Pb (Dueñas et al, 2004; Huang et al, 2009) . Nevertheless, gross beta results can be used as a good indicator of ^{210}Pb concentration.

4. Conclusions

The use of an adequate calibration method indicates that the results from different aerosol samplers agree to within associated uncertainties. However, special care should be taken for low-energy gamma emitters such as ^{210}Pb. For a suitable determination of ^{210}Pb the use of specific calibration standards is recommended employing filters of the same geometry, as is typical in routine measurements. The use of traceable nuclide solutions or reference materials containing ^{210}Pb or ^{133}Ba is also recommended.

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Figure captions:

Fig.1. ^{210}Pb activity concentration in air determined with high- and mid-volume samplers. Error bars indicate combined standard uncertainties of measurement results (k=2). A linear fit has been added to help the reader

Fig. 2. ^{210}Pb activity concentration in air determined through a high-volume sampler versus gross beta activity determined through a low-volume sampler. Error bars indicate combined standard uncertainties of measurement results (k=2). A linear fit has been added to help the reader

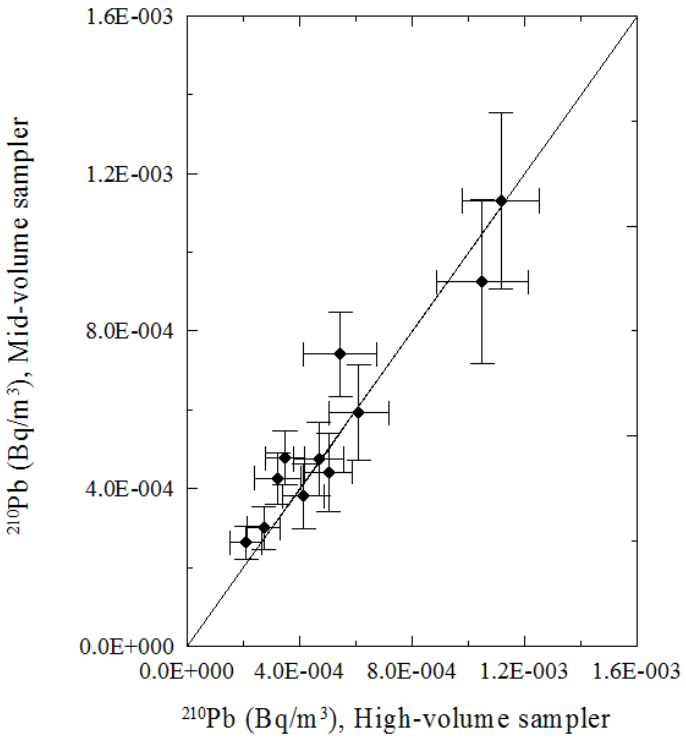
1.6E-003
1.2E-003
8.0E-004
4.0E-004
0.0E+000
0.0E+000
4.0E-004
8.0E-004
1.2E-003
1.6E-003
210Pb (Bq/m3), Mid-volume sampler
210Pb (Bq/m3), High-volume sampler

Fig. 1

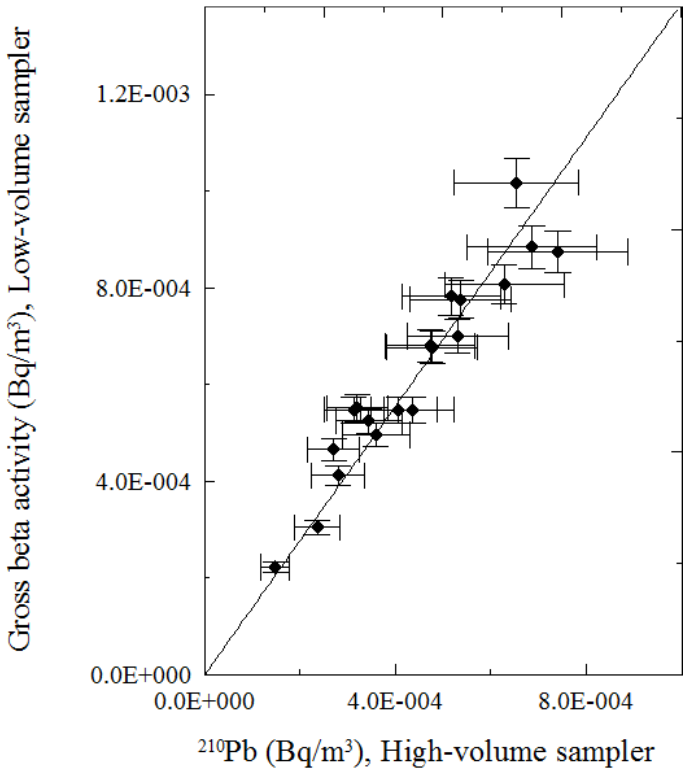
Gross beta activity (Bq/m³), Low-volume sampler
1.2E-003
8.0E-004
4.0E-004
0.0E+000
0.0E+000
4.0E-004
8.0E-004
²¹⁰Pb (Bq/m³), High-volume sampler

Fig. 2

Table 1: ^{210}Pb activity determined according to different calibration procedures for polypropylene (44 x 44) cm^2 filters with spiked ^{210}Pb (SPK2) or phosphogypsum reference material (PRM). Uncertainties are expanded with a coverage factor k=2.

Filter no.	Reference value (Bq/filter)	Polynomial fit		Single isotope calibration	
		Measured value (Bq/filter)	Bias (%)	Measured value (Bq/filter)	Bias (%)
1 (SPK2)	46.8±0.5	52.1±6.8	11	45.6 ± 1.4	-3
2 (SPK2)	44.5±0.5	50.8±6.6	14	44.5 ± 1.4	-
3 (PRM)	5.01±0.17	5.72±0.97	14	5.13±0.62	2

Table 2: ^{210}Pb activity determined according to different calibration procedures for (21 x 26) cm^2 filters with the phosphogypsum reference material (PRM). Uncertainties are expanded with a coverage factor k=2.

Filter no.	Reference value (Bq/filter)	Polynomial fit		Single isotope calibration	
		Measured value (Bq/filter)	Bias (%)	Measured value (Bq/filter)	Bias (%)
1 (PRM)	2.19±0.07	2.35±0.50	7	2.16±0.40	-1
2 (PRM)	1.64±0.05	1.80±0.52	10	1.69±0.45	3