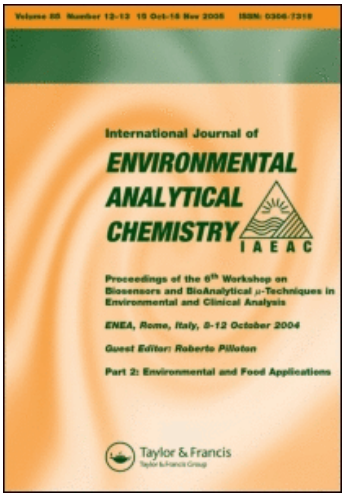

International Journal of Environmental Analytical Chemistry

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713640455

Determination of Natural Gamma Emitters in Surface Air

E. Gómez[a]; F. Garcías[b]; M. Casas[b]; V. Cerdá[a]
[a] Departament de Química, Universital de les Illes Balears, Palma de Mallorca, Spain [b] Departament de Física, Universital de les Illes Balears, Palma de Mallorca, Spain

To cite this Article Gómez, E. , Garcías, F. , Casas, M. and Cerdá, V.(1994) 'Determination of Natural Gamma Emitters in Surface Air', International Journal of Environmental Analytical Chemistry, 56: 4, 327 — 335
To link to this Article: DOI: 10.1080/03067319408034111
URL: http://dx.doi.org/10.1080/03067319408034111

Intern. J. Environ. Anal. Chem., Vol. 56, pp. 327-335
Reprints available directly from the publisher
Photocopying permitted by license only

DETERMINATION OF NATURAL GAMMA EMITTERS IN SURFACE AIR

E. GÓMEZ*, F. GARCÍAS†, M. CASAS† and V. CERDÁ*

**Departament de Química, Universitat de les Illes Balears, E-07071 Palma de Mallorca, Spain; †Departament de Física, Universitat de les Illes Balears, E-07071 Palma de Mallorca, Spain*

(Received, 16 April 1993; in final form, 3 March 1994)

Using cellulose and charcoal filters simultaneously, we propose a very simple method to estimate the attached and unattached fractions of some airborne radionuclides. We have systematically controlled by gamma-ray spectrometry the concentrations in surface air of some primordial radionuclides: ^{40}K, ^{226}Ra, ^{212}Pb, ^{214}Pb, ^{208}Tl and ^{214}Bi. We have also determined the concentration of the cosmogenic radionuclide ^{7}Be.

All the activities of these elements keep lower than the accepted mean values, and in most cases are around the background activity. The results show that ^{214}Bi, ^{212}Pb, ^{214}Pb and ^{208}Tl are always attached to aerosols, whereas ^{7}Be, ^{40}K and ^{226}Ra have also a fraction unattached.

Using a scanning electronic microscope we have determined the main components of the aerosols, which are found to be Si, Ca, Al, Cl and K.

KEY WORDS: Natural gamma emitters, surface air.

INTRODUCTION

The knowledge of the concentrations in air of the natural radioactive elements is of particular importance, not only because natural radiation is the largest contributor to the collective dose of the world population, but also because it is possible to use this radioactivity as a tracer for the study of atmospheric circulation.

The natural radioactive elements in the environment can be classified, according to their origin, into cosmogenic and primordial ones. Cosmogenic radionuclides (i.e. ^{3}H, ^{7}Be, ^{14}C, etc.) are mainly produced by secondary cosmic rays, and contribute to the total external gamma radiation dose at ground level by as much as 40%. The activity of the main primordial radionuclides (^{40}K, ^{70}Rb and the elements of the radioactive series 4n and 4n+2) depends on the concentration of these radionuclides in soil, and gives the most relevant contribution to outdoor exposure. In spite of variations in soil composition, UNSCEAR-82 [1] estimates the outdoor terrestrial absorbed dose rate in air from gamma radiation as 4.4×10^{-8} Gy/h and the relative contributions of ^{40}K, ^{238}U and ^{232}Th to be about 35%, 25% and 40% respectively.

For comparison, the annual absorbed dose from cosmogenic radionuclides is found to be 2.8×10^{-4} Gy at ground level.

The separation between the "unattached" fraction, constituted by free molecular atoms or ions of one isotope, from the "attached" one, composed of a cluster mode aerosol, becomes of great significance given that a considerable number of unattached atoms may deposit in the trachea and the bronchial region, while attached atoms deposit mainly in the pulmonary region. This results in a dose into the bronchial area which is considerably higher than that into the pulmonary epithelium.

Of course, the concomitant literature about the subject of airborne radioactivity is so extensive that no attempt can be made to list a comprehensive set of references (for an excellent recent review see, for example, ref. 2). In particular, the behaviour of the short-lived ^{222}Rn and ^{220}Rn daughters (reported as ^{214}Pb and ^{212}Pb) and their health physics implications have been recently the subject of different works[2-9]. The investigation of the unattached fraction of short-lived Rn decay products using wire screens has been largely addressed[2-4]. In these references the Rn progeny is usually measured by α-spectrometry, which requires a previous chemical process for the sample preparation.

In this work, using cellulose and charcoal filters simultaneously, we propose a very simple method that allows us to estimate by gamma-ray spectrometry, without any previous chemical process, the attached and unattached fractions of the radionuclides considered. We have systematically controlled (every week from September 1992 to April 1993) the concentrations in surface air of some primordial radionuclides: ^{40}K, ^{226}Ra, ^{212}Pb, ^{214}Pb, ^{208}Tl and ^{214}Bi. We have also controlled the activity of ^{7}Be. The samples were collected at the Physics Department of the University of the Balearic Islands (Palma de Mallorca, Spain).

EXPERIMENTAL

Instrumentation

The experimental set-up was composed of :

- RADECO AVS-28A low flow pump (30 l/m) with a base for cellulose and charcoal filters
- Schleicher-Schuell ME-27 membrane filters (diameter 47 mm, porous size 0.8 μm)
- RADECO CP-100 charcoal filters
- CANBERRA GR 250-7500SL germanium detector
- BERTRAN 315 high-voltage source
- CANBERRA 2021 amplifier
- 2048-channel analyzer as an interface to an IBM PS/2/30 microcomputer
- scanning electronic microscope (Hitachi 530) with probe for elemental analysis by X-ray fluorescence (Kevex 2000).

The experimental set-up was located at the University of the Balearic Islands (lat. 39° 38′ N, long. 2° 39′ E, elevation above sea level ≅ 70 m). The pump base with filters was placed outdoor at 4.5 m from the ground level.

Measurements

To concentrate the gamma emitters in the air, we have employed a membrane filter (porous size of 0.8 μm) followed by a charcoal filter, both placed in the base of the low flow pump (30 l/m) working continuously during a week. Aerosols (attached fraction) were retained on the cellulose filter, whereas the charcoal filter was used for gas concentration (unattached fraction).

Four days after the concentration process, the activity of both filters was measured using the gamma spectrometry system. The set-up for gamma-ray spectrometry was calibrated using a ^{152}Eu, ^{137}Cs, ^{133}Ba and ^{60}Co source of the same geometry as the samples. The resolving power of the chain was 5.0 keV.

To give an example, in Figure 1a we show the gamma-ray spectrum obtained from the charcoal filter exposed during the week 8–15 Sept. 1992, and for comparison the spectrum of the cellulose filter corresponding to the same period is displayed in Figure 1b. Both spectra were obtained by measuring the radiation emitted by the samples for 24 h. For each filter, the activities of the radionuclides detected were calculated from the areas of the peaks, after subtraction of the corresponding background spectrum.

The background activity was obtained by measuring the radiation emitted during 24 hours by the filters before aerosol concentration, assuming the same air volume pumped as in the samples. From the background measurements we have computed the Low Limit of Detection

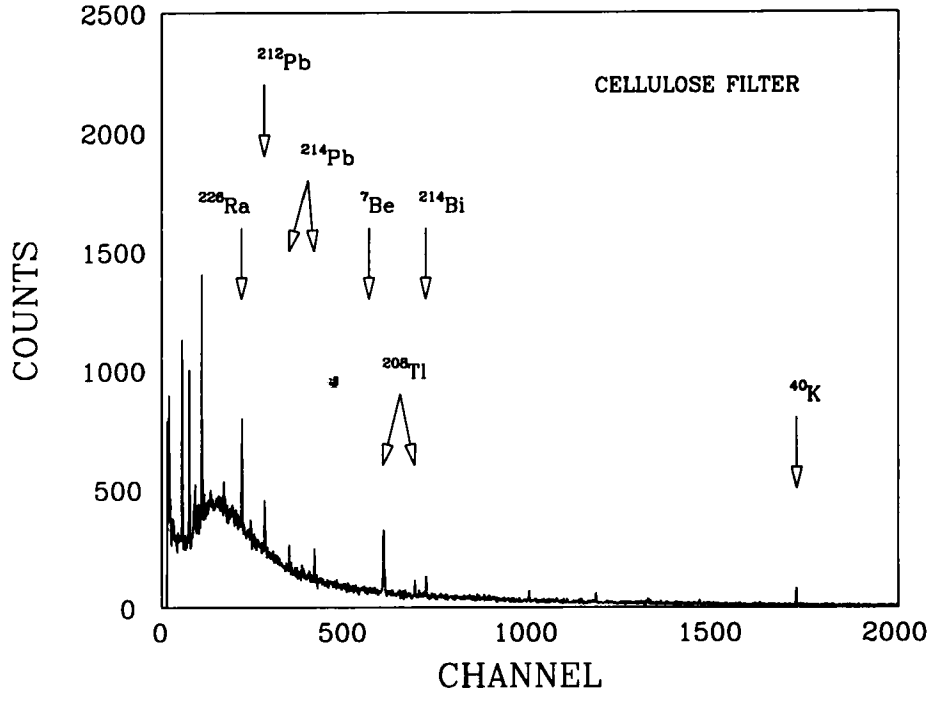

Figure 1 Gamma spectra of filters corresponding to the week 8–15 Sept. 1992. a) charcoal filter

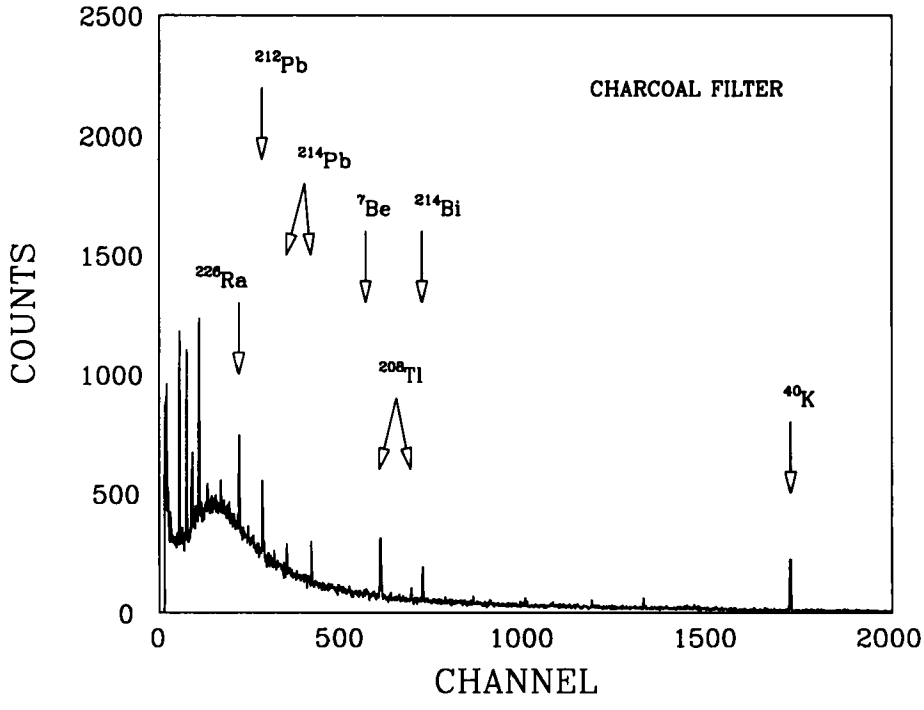

Figure 1 Gamma spectra of filters corresponding to the week 8–15 Sept. 1992. b) cellulose filter.

(LLD) as 3 times the standard deviation of the spectrum basis line [10] for each radionuclide and for each kind of filter.

On the other hand, we have controlled the influence of the porous size of the membrane filter on the retention of aerosols. Using cellulose filters with porous sizes from 0.1 to 0.8 μm we have found that ^{226}Ra, ^{214}Pb and ^{214}Bi are best retained in the filters with porous size about 0.2 μm, but the activities of ^{212}Pb and ^{208}Tl remain more or less insensitive to the porous size. These results seem to be in contradiction with the predictions of ref. 5 where a mean aerodynamic diameter of 0.16 μm for ^{214}Pb and 0.13 μm for ^{212}Pb is assumed. Nevertheless, all the results obtained are of the same order of magnitude. In what follows we present the results obtained using cellulose filters with a porous size of 0.8 μm, which are appropriate in health physics because 0.8 μm is approximately the size of the pulmonary alveolus.

The cellulose filters were also analyzed using a scanning electronic microscope (Hitachi 530) with probe for elemental analysis by X-ray fluorescence (Kevex 2000). The composition of the cellulose filter before being submitted to the aerosol concentration process previously described is only organic matter, and the main components, with $Z>10$, in the cellulose filter after the aerosol concentration process are Si, Ca, Al, Cl and K, as was expected according to the soil composition (Majorcan soil is essentially calcareous).

Table 1 Mean background activities (MBA) and low limits of detection (LLD) obtained in cellulose and charcoal filters for the weekly concentrations of the natural gamma emitters.

		Ra-226	Pb-212	Pb-214	Be-7	Tl-208	Bi-214	K-40
MBA	CF	12.5	0.84	N.D.	N.D.	0.15	N.D.	4.2
(mBq/m^3)	CHF	20.3	0.72	2.4	N.D.	0.32	2.0	30.5
LLD	CF	150	19	22	90	13	35	140
($\mu Bq/m^3$)	CHF	1150	170	100	310	66	63	400

N.D. = Not detected
CF = Cellulose filter.
CHF = Charcoal filter.

RESULTS

The mean background activity together with the Low Limit of Detection (LLD) for each element in membrane and charcoal filters are shown in Table 1. Notice that in all cases the LLD of cellulose filters are below those of charcoal filters, according to the background measurements.

The weekly activities (exceeding the LLD) of the radionuclides retained on charcoal filters from September 1992 up to April 1993 are displayed in Figure 2, as well as the rate of rainfall by week. A quick glance at this figure shows that only ^{7}Be, ^{40}K and ^{226}Ra present

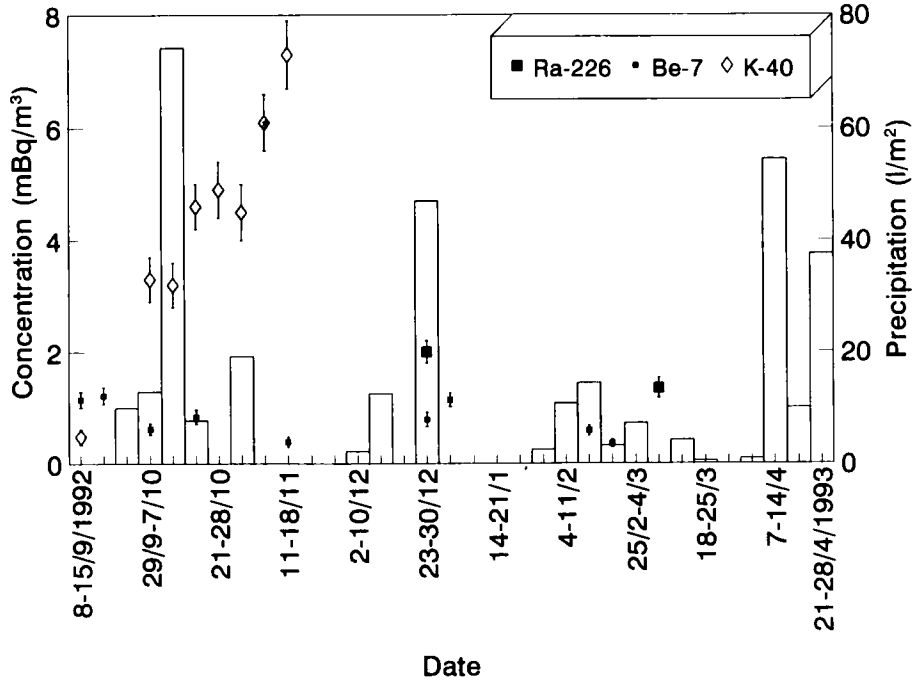

Figure 2 Weekly concentrations (only those exceeding LLD) of the natural gamma emitters retained on charcoal filters. The histogram shows the rate of rainfall by week.

an unattached fraction of the activity. The activities of ^{208}Tl, ^{212}Pb, ^{214}Pb and ^{214}Bi in charcoal filters always correspond to the background one (Bg) because these nuclei are always attached to aerosols and retained by the cellulose filters.

This is clearly shown in Table 2a where some selected weekly activities of charcoal and cellulose filters are compared. Notice that, as expected, there is a clear deviation from secular equilibrium. The figures show that all the elements of the 4n and 4n+2 series have been preferentially found in a cluster mode. For all these radionuclides the attached fraction is always much greater than the unattached one, only ^{226}Ra having a significant gaseous fraction in some cases. The results concerning the unattached fraction of ^{40}K are in contradiction with the usual criterion that the large-lived elements must be preferentially found in a particle-bound mode. Our work confirms that the activity of both fractions of ^{40}K at ground level clearly depends on the air dust concentration. The same comments apply to Table 2b where the monthly mean concentrations are shown. We want to remark that these activities give the absolute radionuclide concentrations on the filters at the measuring date.

There is not a clear correlation between the total rainfall, which is also shown in Tables 2a and 2b, and the unattached activity of ^{7}Be, ^{40}K and ^{226}Ra. Nevertheless, during the most dry periods the mean concentration of the radionuclides increases but remains less than the accepted mean values [1], as it corresponds to a calcareous soil where the concentration of elements from the 4n and 4n+2 series is rather small.

CONCLUSIONS

We have proposed a very simple method, using simultaneously cellulose and charcoal filters, that allows us to estimate the attached and unattached fractions of the radionuclides analyzed. The systematic control during eight months of some gamma emitters in surface air at the Balearic Islands shows that ^{214}Bi, ^{212}Pb, ^{214}Pb and ^{208}Tl are always attached and ^{7}Be, ^{40}K and ^{226}Ra have a fraction attached and a fraction unattached. In all the cases analyzed the concentration of primordial radionuclides is less than the accepted mean value.

The analysis of the cellulose filters by a scanning electronic microscope shows that the main components of the aerosols are Si, Ca, Al, Cl, and K, in agreement with the Majorcan soil composition.

Acknowledgments

We wish to thank F. Hierro (*Servei de Microscopia Electrònica* of the University of the Balearic Islands) for his technical help, and J. H. E. Cartwright (*Departament de Física* of the University of the Balearic Islands, on leave from the University of London) for a careful reading of the manuscript. This work has been partially supported by the REVIRA program between CSN and the University of the Balearic Islands. Additional financial support from DGICYT (PB92-0021-C02-02 and PB90-0359 grants) is also gratefully acknowledged.

Table 2a Weekly concentrations of the natural radionuclides analyzed.

	Activity ($\mu Bq/m^3$)													
	8-15 Sept Rf= 0.0 l/m^2		14-21 Octob Rf= 7.7 l/m^2		2-10 Decemb Rf= 2.2 l/m^2		14-21 Jan Rf= 0.0 l/m^2		11-18 Febr Rf=14.4 l/m^2		11-18 March Rf=4.2 l/m^2		7-14 April Rf=54.4 l/m^2	
Element	CF	CHF	CF	CHF	CF	CHF	CF	CHF	CF	CHF	CF	CHF	CF	CHF
Be-7	Bg	1150±140	Bg	840±110	Bg	Bg	500±40	Bg	Bg	590±90	<LLD	Bg	1100±90	Bg
K-40	410±90	480±140	Bg	4600±400	<LLD	Bg	<LLD	Bg	Bg	Bg	Bg	Bg	<LLD	Bg
Tl-208	Bg	<LLD	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	68±6	<LLD
Pb-212	21±3	Bg	<LLD	Bg	21±3	Bg	<LLD	Bg	<LLD	Bg	48±4	Bg	116±7	Bg
Bi-214	140±10	Bg	1020±140	Bg	90±10	Bg	270±20	Bg	130±10	Bg	160±10	Bg	250±20	Bg
Pb-214	180±20	Bg	310±20	Bg	160±20	Bg	160±20	Bg	40±5	Bg	48±5	Bg	190±10	Bg
Ra-226	280±30	<LLD	<LLD	Bg	520±40	Bg	520±40	<LLD	360±30	Bg	710±50	Bg	400±30	Bg

Rf = weekly rainfall rate.
CF = Cellulose filter.
CHF = Charcoal filter.
LLD low limit of detection.
Bg background.

Table 2b Monthly mean concentrations of the natural radionuclides analyzed.

	Activity (μBq/m^3)															
	September Rf= 10.0 l/m^2		October Rf= 94.8 l/m^2		November Rf= 19.2 l/m^2		December Rf= 61.6 l/m^2		January Rf= 0.0 l/m^2		February Rf= 30.9 l/m^2		March Rf= 12.0 l/m^2		April Rf=102.8 l/m^2	
Element	CF	CHF	CF	CHF	CF	CHF	CF	CHF	CF	CHF	CF	CHF	CF	CHF	CF	CHF
Be-7	<LLD	870±100	100±10	390±70	110±10	130±40	560±40	200±30	190±20	310±30	140±20	230±40	210±20	<LLD	<LLD	Bg
K-40	<LLD	160±50	380±60	4000±400	120±30	3600±300	Bg	Bg	270±50	Bg	290±50	Bg	<LLD	Bg	230±50	Bg
Tl-208	<LLD	<LLD	36±5	<LLD	<LLD	Bg	Bg	<LLD	<LLD	Bg	<LLD	<LLD	Bg	Bg	<LLD	<LLD
Pb-212	10±1	Bg	<LLD	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg
Bi-214	380±70	Bg	140±10	Bg	<LLD	Bg	<LLD	Bg	<LLD	Bg	Bg	Bg	Bg	Bg	Bg	Bg
Pb-214	37±4	Bg	78±7	Bg	59±5	Bg	<LLD	Bg	47±7	Bg	Bg	Bg	Bg	Bg	Bg	Bg
Ra-226	160±20	450±70	<LLD	Bg	<LLD	170±20	350±30	510±50	360±30	Bg	590±50	Bg	<LLD	<LLD	270±30	Bg

Rf = monthly rainfall rate.
CF = Cellulose filter.
CHF = Charcoal filter.
LLD low limit of detection.
Bg background.

References

1. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), *Ionizing Radiation: Sources and Biological effects* (United Nations publ., New York, 1982).
2. A. Reineking and J. Porstendörfer, *Health Phys.*, **58,** 715–727 (1990).
3. M. Ramamurthi and P. K. Hopke, *Health Phys.*, **56,** 189–194 (1989).
4. C. Papastefanou and E. A. Bondietti, *J. Envir. Radioactivity*, **13,** 1–11 (1991).
5. C. Papastefanou and E. A. Bondietti, *Health Phys.*, **53,** 461–472 (1987).
6. F. Raes, *Health Phys.*, **49,** 1177–1187 (1985).
7. H. M. Y. Leung and C. R. Phillips, *Radiat. Prot. Dosim.*, **22,** 97–105 (1988).
8. W. W. Nazaroff, D. Kong and A. J. Gadgil, *J. Aerosol Sci.*, **23,** 339–352 (1992).
9. H. Kojima, S. Abe and K. Fujitaka, *Radiat. Prot. Dosim.*, **46,** 103–109 (1993).
10. L. A. Currie, *Anal. Chem.*, **40,** 586 (1968).