

AN EXPERIMENTAL SEARCH FOR A CORRELATION BETWEEN OUTDOOR ^{222}Rn CONCENTRATION AND ^{210}Pb ACTIVITY IN AIR PARTICULATE SAMPLES

by

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This work was aimed to search for a correlation between outdoor ^{222}Rn air concentration and ^{210}Pb activity in particulate samples collected on a filter. The existence of a correlation could support the hypothesis that both ^{222}Rn and its long-lived product ^{210}Pb are embedded into the same air masses and undergo the same air transport phenomena. Lead-210 activity was determined by means of gamma ray spectrometric measurement carried out a few days after the sampling whereas ^{222}Rn concentration was measured through a commercial monitor. Experimental tests allowed to obtain a weak correlation between ^{222}Rn and ^{210}Pb air concentration as a preliminary result due to high uncertainties of outdoor ^{222}Rn concentration measurements.

Key words: ^{222}Rn , ^{210}Pb , air filtration, atmospheric particulate, gamma ray spectrometry

INTRODUCTION

Radioactivity measurement of air particulates collected on filter is a standard method used in environmental monitoring to evaluate air concentration of both natural and artificial gamma-emitting radionuclides. Over the last years, our interest was aimed to study techniques for sampling and measurement of air particulate matter, with reference to artificial radionuclides from nuclear accidents or in resuspended dusts as well as natural species produced by cosmic-ray interactions with the atmosphere and ^{222}Rn decay products [1-9].

A particular interest has been focussed on ^{210}Pb air concentration evaluation both as a tracer to study atmospheric aerosol transport and for its contribution to population dose from internal irradiation [5, 10-12]. Airborne activity of ^{210}Pb (half-life, $T_{1/2} = 22.3$ year), a daughter product in the ^{238}U decay series, is mainly produced by the short-lived decay products of ^{222}Rn ($T_{1/2} = 3.8$ d), inert radioactive gas emanates from soils and building materials into the atmosphere. Once produced, ^{222}Rn short lived daughters (^{218}Po , $T_{1/2} = 3.05$ min; ^{214}Pb , $T_{1/2} = 26.8$ min; ^{214}Bi , $T_{1/2} = 19.9$ min) and ^{210}Pb become attached to the aerosols particles and their concentrations change as a result of the movement of air masses.

Due to its half-life, ^{210}Pb can be practically considered the final product of ^{222}Rn decay, and therefore its activity can be correlated with the outdoor ^{222}Rn concentration. In a previous work [10] a relationship between airborne ^{210}Pb concentration and wind direction was highlighted. Indeed, the lowest average values of ^{210}Pb concentration were observed in correspondence to wind directions bringing maritime air masses. The existence of a correlation could support the hypothesis that both ^{222}Rn and its long-lived product ^{210}Pb are embedded into the same air masses and thus undergo the same air transport phenomena.

Some correlations were already established for indoor air, where it is fairly easy to find reliable ^{222}Rn measurement instruments [13-17], while few studies were concerned outdoor environments [e. g., 18].

The most widespread measurement techniques adopted to evaluate ^{210}Pb air activity concentration differ in analysis and sample preparation methods, and some of them involve complex chemical separation processes of ^{210}Pb from its daughters [19-22]. Such techniques permit the ^{210}Pb activity determination by detecting 1.2 MeV α -emission of daughter ^{210}Bi ($T_{1/2} = 5.0$ days), in equilibrium with ^{210}Pb after about 30 days, or 5.3 MeV α -emission of its grand daughter ^{210}Po ($T_{1/2} = 138.4$ days). However, the high detection limit of the β -counting technique needs of high level of activity, while α -spectrometry has the disadvantage of a long delay period between sampling and measure-

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ment to reach equilibrium between activities of ^{210}Pb and ^{210}Po [19-22].

The availability of a high-volume air sampler and a low background planar HPGe detector, allows evaluation of ^{210}Pb activity on filter through a direct γ -ray spectrometric measurement carried out a few days after the sampling. A description of γ -ray spectrometric system besides sampling and measurement methods have already been reported in previous works [1-6, 9, 10].

The aim of this study is to verify a possible correlation between measurements of ^{210}Pb activity in particulate samples collected on filter with outdoor ^{222}Rn air concentration. Experimental tests have allowed to obtain a weak correlation between ^{222}Rn and ^{210}Pb concentration in outdoor air as a preliminary result due to high uncertainties of outdoor ^{222}Rn concentration measurements.

MATERIALS AND METHODS

Air particulate samplings

Outdoor particulate samplings were collected on cellulose filter paper by means of a high-volume air sampler (HVAS) located on the roof of department building, about 20 m above the ground level [3]. Particulate collection was performed by suction atmospheric air through a 45 cm \times 45 cm Sofiltra Poelman HYN-75 (Bleu type) cellulose filter paper mounted vertically in HVAS, with a flow-rate of about 900 m³h⁻¹. The filter, after the sampling, was cut into strips, folded and pressed to obtain a „packet-sample” geometry with 6 cm \times 6 cm \times 0.7 cm dimensions [1-6].

Sampling on almost daily basis (14 h, from 7 p. m. to 8 a. m. the following day, GMT time, about 12,000 m³ volume of air filtered) was carried out for a period of about 6 months, mainly in the Winter and Spring months.

Air-flow rate was determined through a Flow Meter Turbine calibrated by the manufacturer (Flow Technologies Inc.) in the measurement range with a specification of a combined uncertainty of no more than 1 % in the air volume sampled. However, air flow rate varied with the suction time and to check the variation, the flow rate was measured using a microprocessor-based electronic circuit designed for detecting and conditioning impulse signals from a magnetic pick-off device mounted on the Flow Meter Turbine. The output low-amplitude signals were converted to 10 V pulses and recorded by means of a PC-based multi-channel scaler (MCS Silena EMCAPLUS) with a 30-second dwell time per channel [3].

Gamma ray spectrometric measurements

The air particulate samples were analyzed by a gamma-ray spectrometric system with an ORTEC™

GLP Series Planar HPGe LEPS detector, 1000 mm² active area and 7 mm depth, with a 0.254 mm thick beryllium window. The energy resolution (FWHM) was 440 eV at 5.9 keV and 620 eV at 122 keV.

The detector was coupled with an ORTEC™ 672 amplifier and an ORTEC™ 919E EtherNIM multi-channel Buffer connected into an Ethernet environment. Data analysis was performed using EG&G ORTEC™ GammaVision® version7 software [23]. Lead-210 was identified through its gamma emission of 46.5 keV, with an intensity of 4.23 \pm 0.05 % [24]. To reduce the background level under 46.5 keV photopeak, spectrometric measurements were carried out a few days after the sampling to allow for the decay of short-lived products of ^{222}Rn and ^{220}Rn .

The 46.5 keV efficiency evaluation was performed by measuring a calibrated disk source of ^{210}Pb - ^{210}Bi , 2.5 cm active diameter, as described in [25]. Efficiency value of 2.5 \pm 0.12 $\cdot 10^{-3}$ counts per Bq (5.9 \pm 0.3 $\cdot 10^{-2}$ counts per 46.5 keV photon) was very close to the value of 2.27 $\cdot 10^{-3}$ counts per Bq obtained with other techniques [8].

To determine detection limit (DL) for 46.5 keV photons and ^{210}Pb Minimum Detectable Activity (MDA), a blank filter was counted for the time generally adopted for filter analysis with LEPS detector (8 $\cdot 10^4$ s). The relation for paired sample and blank measurements reported by Currie [26] was adopted and a value of DL= 27 counts and a MDA = 0.14 Bq were determined for ^{210}Pb activity.

Radon concentration measurements

During the 14-h air samplings, the ambient ^{222}Rn concentration level (C_{Rn}) was also monitored by a Genitron AlphaGuard P2000Q Monitor based on a ionization chamber able to detect low values of ^{222}Rn concentrations in the sampled air (active volume: 0.56 L; minimum detectable concentration, MDC: 2 Bq m⁻³). The equipment was placed on the roof of our Department, close to the HVAS. Since the counting interval cannot be set for more than 1 h, the 14 values corresponding to the filter sampling period were added to find an ^{222}Rn average concentration in the same reference period.

To perform some comparative analyses it was useful to know the equilibrium equivalent concentration (EEC), *i. e.*, the ^{222}Rn concentration, in equilibrium with the short-lived daughters, that would have the same potential alpha energy per unit volume as the existing mixture. EEC evaluation can be carried out through the knowledge of the equilibrium factor (F_{EQ}) according to

$$\text{EEC} = C_{\text{Rn}} F_{\text{EQ}} \quad (1)$$

To this goal, simultaneous measurements of ^{222}Rn concentration by a SILENA PRASSI (Portable

Radon gas Survey meter) Model 5S, and of Working Level (WL), a quantity related to the concentration of the decay products, by a SILENA Radon/Thoron Daughters Meter (Model 4S), were performed near the HVAS. PRASSI'S built-in detector is a 2-liter Lucas cell, with internal walls coated with ZnS(Ag) coupled with a low gain-drift photomultiplier. The SILENA 4S Meter is provided with a silicon solid-state detector able to spectrometric differentiate the alpha emission of ^{222}Rn and ^{220}Rn daughters. For both detectors, an integration time setting was 4 hours.

RESULTS AND DISCUSSION

The high sampled air volume and use of LEPS HPGe detector allow to turn out values of 46.5 keV photopeak count-rate larger than the detection limit of spectrometric analysis system in direct gamma-ray measurements performed a few days after the sampling. Figure 1 shows a typical gamma ray spectrum detected on a particulate sample highlighting 46.5 keV gamma emission and Bi X-rays.

Outdoor air ^{222}Rn concentration was evaluated in the same 14 h sampling period by computing, as above described, an arithmetic average of the values furnished by AlphaGuard P2000Q during the same filter sampling time.

For the aims of this work, ^{222}Rn measurements with overall uncertainty of more than 50 % have not been taken into account, as well as the evaluation of ^{222}Rn concentration lower than MDC that were considered unreliable. This resulted in the selection of 118 values that, together with the corresponding ^{210}Pb filter activity evaluations, formed the experimental basis for the test on the correlation. With reference to numerical values, ^{222}Rn concentrations varied from 2.1 to 11.1 Bqm^{-3} , with an average value of 5.4 Bqm^{-3} and uncertainties in the range 23 %-47 %. As for ^{210}Pb , all measurements were higher than the detection limit, with activity on filter ranging from 0.3 to 19 Bq. The derived ^{210}Pb air activity concentrations ranged between 0.05 and 1.3 mBqm^{-3} , with an average value of 0.52 mBqm^{-3} and standard deviations ranging between 2 % and 16 %.

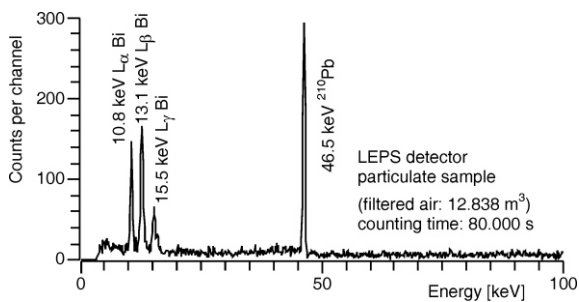


Figure 1. Gamma-ray spectrum of a particulate sample collected on filter and analyzed with LEPS detector

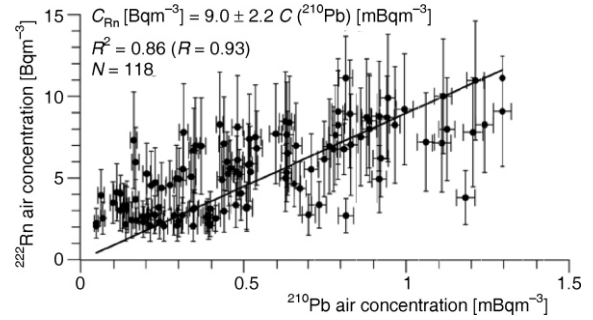


Figure 2. Correlation between the 14 h-average air concentrations of ^{222}Rn and ^{210}Pb

The correlation was verified by an interpolation of the experimental points with a straight line passing through the origin of the axes (fig. 2), resulting as

$$C_{\text{Rn}} [\text{Bqm}^{-3}] = 9.0 + 2.2 C (^{210}\text{Pb}) [\text{mBqm}^{-3}]$$

with $C(^{210}\text{Pb})$ the ^{210}Pb air concentration expressed in mBqm^{-3} . The high uncertainty value of the coefficient was largely related to the statistical precision of the measurements (reported in fig. 2), of ^{222}Rn concentration in particular, which low levels go up/down in value quite rapidly during the sampling time (night hours).

To confirm the validity of the experimental test, a comparison of the correlation coefficient with the value of outdoor air reported in [11] as function of EEC was carried out. To this goal, an experimental evaluation of F_{EQ} was performed in a 5-days test by using both the SILENA PRASSI ^{222}Rn monitor and SILENA Model 4S, which output in terms of WL can be converted to EEC since, at radioactive equilibrium between ^{222}Rn and its progeny, 1 WL corresponds to $\text{EEC} = 3.7 \cdot 10^3 \text{ Bqm}^{-3}$. Figure 3 reported the ^{222}Rn concentration, EEC and F_{EQ} values obtained during test days. The mean value of the equilibrium factor F_{EQ} was 0.65 ± 0.20 , *i. e.*, close to rounded value of 0.6 reported by UNSCEAR [27], and, considering also the large statistical fluctuation of the measurements, can be considered comparable with the Kojima [28] best estimate $F_{\text{EQ}} = 0.51 - 0.12$. Assuming F_{EQ} value of 0.65 for outdoor air, a relationship between ^{210}Pb concentration in air and EEC was obtained

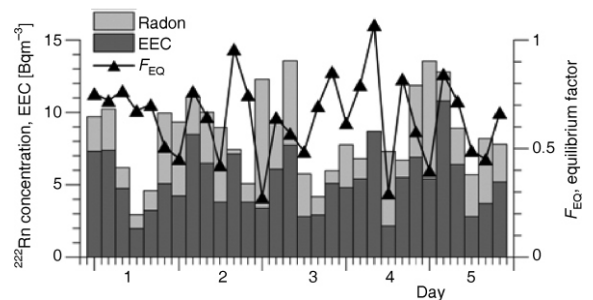


Figure 3. Values of air ^{222}Rn concentration and EEC measured near the sampling station, and corresponding behaviour of equilibrium factor, F_{EQ} . Mean value was $F_{\text{EQ}} = 0.65 \pm 0.20$

$$C(^{210}\text{Pb}) \approx 7.2 \cdot 10^{-5} \text{ EEC [Bqm}^{-3}\text{]}$$

with a coefficient very close to $7.5 \cdot 10^{-5}$ reported in [11], which confirms the consistency of the experimental approach, regardless of uncertainty extent.

An analysis of the causes of uncertainty has been made with reference to parameters for particulate sampling and measurements given in [29, 30]. Some of the parameters that may be affected by uncertainties were already taken into account by the methods used to determine ^{210}Pb and ^{222}Rn air activity. Among them, the reproducibility of source positioning and geometry was assured by adopting the „packet-sample” geometry and a Plexiglas box inside which was placed on the filter contained in a polyethylene bag. No correction for density and atomic composition was required since the filters, on average, had the same weight and composition of the calibration sample.

As regards instability of the gamma-ray spectrometric systems, a visual observation leads easily to identify the 46.5 keV photopeak position while the counting times have been chosen so to have uncertainties of no more than 20 % in the photopeak area values even with low activity. Correction for dead time, pile-up and accidental loss of count were performed by spectrometric analysis software. More attention deserves the filter efficiency, *i. e.*, the ratio between quantity of particulate effectively withheld by the filter and total particulate matter filtered. As the absolute value of ^{210}Pb activity depends on filter efficiency, a verification of the value of 75 % furnished by factory (Sofiltra Poelman) was needed. Measurements performed on samplings carried out with two filters, mounted one in front of the other, pointed out an efficiency value for ^{210}Pb of $76 \pm 5.5 \%$, with a very good agreement with the furnished value. The statistical precision for ^{210}Pb measurements is related to countings with 150,000 s lifetime for the first sample and 640,000 s lifetime for the second filter (count rate: $2.0 \pm 0.1 \cdot 10^{-3}$ counts per second).

As already mentioned, uncertainty on air volume sampled V was furnished by the turbine manufacturer as $\Delta V/V = 1 \%$, corresponding to what indicated by Makarewicz [30]. More importance had the variation of the flow rate during the same sampling period. It points out a quasi-linear time variation of air-flow rate, with a decrease of about 20 % in 14-hours suction time. As a relatively significant decrease in flow rate was noted in the remaining hours of the day, depending on quantity of particulate matter or weather conditions, it was thought not to extend the sampling for the entire day.

Finally, ^{210}Pb total activity does not provide exactly the value of the ^{210}Pb air concentration, as a fraction of activity can be attributed to short-lived ^{222}Rn daughters decay on filter. However, as stated in other works [10, 11, 31] and confirmed in some outdoor short-time (1 h) experimental samplings, contribution of short-lived ^{222}Rn daughters decay on filter to the value of ^{210}Pb activity can be considered in almost all

samples negligible, or at most, statistically not significant (2-8 %) with respect to the ^{210}Pb activity standard deviation. Therefore, ^{210}Pb air activity concentration measurement procedure described in this work can be considered reliable and used also in routine monitoring.

CONCLUSIONS

A preliminary experimental test to verify a correlation between outdoor ^{222}Rn concentration and ^{210}Pb activity collected on filter, was presented. The evaluation of ^{210}Pb air concentration through gamma-ray spectrometric measurement of air particulate collected on filter allows to establish a weak correlation with ^{222}Rn concentration in the same sampling time period. Lead-210 activity above MDA can be surely obtained by long time (14 h) samplings, and ^{210}Pb air activity concentration measurement procedure can be used also in routine monitoring of environmental radioactivity. Measurements of outdoor Radon concentration performed with a commercial ^{222}Rn monitor were affected by relative high uncertainties, so this result can be considered only as a preliminary test. Adopting a ^{222}Rn monitor with lower detection limit, uncertainties can be reduced, and this search can be improved obtaining a better correlation degree between outdoor ^{222}Rn air concentration and ^{210}Pb activity on filter. The evidence of correlation could confirm the hypothesis that both ^{222}Rn and ^{210}Pb are embedded in the same air masses and can be used as a tracer to study atmospheric aerosol transport.

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ЕКСПЕРИМЕНТАЛНО ОДРЕЂИВАЊЕ КОРЕЛАЦИЈЕ ИЗМЕЂУ КОНЦЕНТРАЦИЈЕ ^{222}Rn У СПОЉАШЊОЈ СРЕДИНИ И АКТИВНОСТИ ^{210}Pb У УЗОРКОВАНИМ ЧЕСТИЦАМА ВАЗДУХА

Намера овог рада је да се одреди корелација између концентрације ^{222}Rn у спољашњој средини и активности ^{210}Pb у честицама ваздуха узоркованим филтером. Постојање корелације би могло подржати хипотезу да су и ^{222}Rn и његов дугоживећи потомак ^{210}Pb уграђени у исте масе ваздуха и да подлежу истим феноменима транспорта ваздуха. Активност ^{210}Pb одређена је спектрометријом гама зрачења неколико дана после узорковања, док је концентрација ^{222}Rn мерена применом комерцијалног монитора. Експериментални тестови су као прелиминарни резултат показали постојање слабе корелације између концентрације ^{222}Rn и ^{210}Pb у ваздуху услед велике несигурности при мерењима концентрације ^{222}Rn у спољашњој средини.

Кључне речи: ^{222}Rn , ^{210}Pb , филтрирање ваздуха, ајмосферска честица, гама спектрометрија