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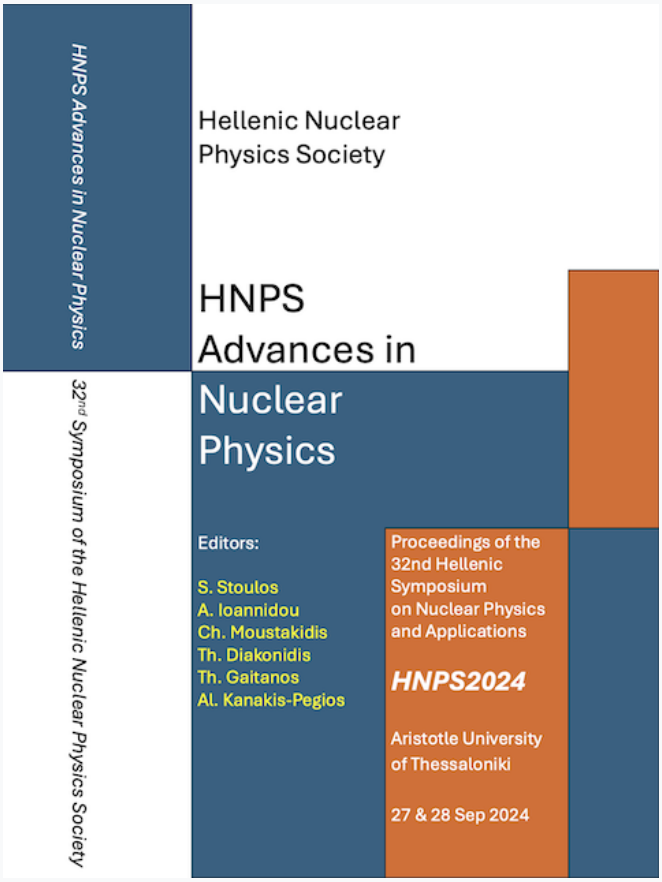

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A Review of Methodologies for Measuring Geogenic Rn Exhalation

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Abstract Radon (^{222}Rn) and thoron (^{220}Rn) are potential health hazards and therefore their concentration levels have been extensively monitored indoors across Europe resulting in indoor radon concentration maps [1]. Direct in situ measurements of geogenic radon and thoron exhalation are limited to a few locations worldwide while indoor measurements are more common. Potentially hazardous zones for long-term inhabitancy can be characterized, considering spatial geogenic radon exhalation data acquired before any construction activities. Furthermore, atmospheric simulations, earthquake prediction and identification of fractures within the lithosphere can be more accurate with the aid of radon exhalation data. In this review direct and indirect methodologies of measuring ^{222}Rn and ^{220}Rn exhalation are presented and compared.

Keywords Radon, Thoron, Exhalation, Geogenic

INTRODUCTION

Radon (^{222}Rn) is a noble gas, daughter radionuclide of radium (^{226}Ra) and part of the uranium (^{238}U) decay chain. Two additional radioisotopes of radon exist, namely thoron (^{220}Rn) and actinon (^{219}Rn) of the thorium (^{232}Th) and the actinium (^{235}U) decay chain respectively. Because of its short half-life (4.0 s) ^{219}Rn is neglected as a health hazard. Thoron used to be ignored in radon hazard case studies due to its short half-life (56 s), but its importance was recently recognized [2]. Although a smaller fraction of thoron, compared to radon, enters the lungs, its progeny ^{212}Pb has a longer half-life (10.6 h) and can be traced in breathable air in high concentrations [3]. In this review the sum of ^{220}Rn and ^{222}Rn concentration is referred to as Rn.

Rn is not only a health hazard, but also useful in atmospheric environmental research and in air mass tracking, where it can be used as an atmospheric tracer. Boundary layer development can be modelled by spatial distribution of radon gas [4]. When measuring fine particle pollution using beta-attenuation monitors (BAM) radon progenies are also important [5]. Seismic research uses radon exhalation for monitoring fault zones [6-9]. Attempts have been made to predict earthquakes by monitoring the changes of the exhalation rate of ^{222}Ra [10-11].

In this review, methodologies for measuring ^{220}Rn and ^{222}Rn exhalation from the soil are described. In the first part of this brief review the main radon exhalation methodologies will be presented. In the discussion, these methodologies are compared based on available publications and the corresponding datasheets. The advantages and disadvantages of the methodologies will be summarized in the conclusions.

RADON EXHALATION METHDOLOGIES

A series of methods for measuring radon exhalation are available. Direct methods measure the actual radon gas emitted from the soil. Direct measurement techniques are further categorized into active and passive. In passive techniques the detector is exposed to radon without the need for electric power consumption. Active methods, also known as continuous, dynamic measurements, involve the

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use of an electric power source to detect instantly the α-particles emitted by the radioactive decay. The detectors involved in these methods are: i) ionization chambers (IC) ii) semi-conductor detectors (SCD), also known as solid state detectors (SSD), and iii) scintillation cells (SC). Indirect methods use proxies like ^{226}Ra concentration in soil or the ambient γ dose equivalent rate (ADER) to assess ^{222}Rn activity (Figure 1).

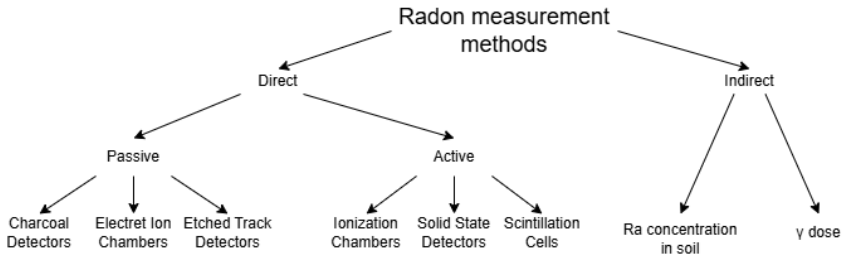

Figure 1. *Flow chart of the different radon measurement methods*

Based on the duration of the measurement, methodologies can be categorized further into i) grab sample methods, where the measurement's duration spans from seconds to minutes. ii) short-term average measurements which last for days, like the charcoal adsorption method [6] iii) continuous methods measure over a period of minutes or hours repeated indefinitely as desired. This group includes IC, SCD, and SC [12] iv) long-term average measurements last for weeks or months. In this case etched track (ETD) detectors and electret are used [13]. According to the way radon is entering the assembly, two methodologies can be distinguished: Diffusion and flow-through. In the first case, the gas diffuses freely into the probe (Figure 2a). In the second case, the gas is pumped into the probe using an electric pump (Figure 2b). Charcoal adsorption [6], etched track and electret detectors use diffusion, IC, SCD, and SC can use both.

Activated Charcoal is used in direct passive measurement methods as an adsorption medium. It is placed as close as possible to the surface being investigated. After the exposure, the sample is left for 3 hours and the activities of radon progenies ^{214}Pb and ^{214}Bi are measured via a beta-radiometer based discharge counter. From this the activity of ^{222}Rn is determined [6].

Solid State Nuclear tracks Detectors (SSNTDs), also known as Etched Track Detectors, are manufactured from polymers sensitive to charged particles and of linear energy transfer (LET) greater than or equal to 5 keV/μm [14]. Alpha particles generated by the decay of radon and its progeny, irritate the plastics and produce damage tracks. The damage done by the α-particles is revealed by etching the plastic film with an acid solution of NaOH or KOH and supplemented with ethanol. The tracks are then counted with a microscope by vision or by track counting software.

The ion detector of the Electret Ion Chamber (EIC) is the electret (electrically charged Teflon disc). Electrets have a permanent surface charge of several keV. They are the electrostatic equivalent of permanent magnets [13] which are mounted in the interior of an chamber able to conduct electricity. The charged progeny of radon collected by the electret reduces its charge which corresponds to the total ionization during the measurement. Radon concentration in air is calculated using specific calibration factors and the measurement time from the reduction in charge which is obtained by a battery-operated electret reader in the laboratory.

In direct active methods, α-particles emitted from the decay of ^{222}Rn or ^{220}Rn are counted by using IC and SC. With SCD, Rn concentration is deduced by its progeny ^{218}Po and ^{212}Po, for ^{222}Rn and ^{220}Rn respectively. In ICs, a pulse-counting, open-air ionization chamber (sensitive to the kinetic energy of α-particles emitted in the chamber) is used. These devices collect the charges created by the stopping power of α-particles in the gas-filled detector. The concentration of the α-emitter in the chamber is directly proportional to the amount of electric current generated in the ionization chamber [15].

Commercially available probes are i) the older AlphaGUARD (AG) P-series (Genitron, Saphymo) and the newer D Series (Bertin) which are equipped with a 0.56 L IC and ii) the ATMOS (Radonova) which has a 2.2 L IC. The AG P-series counts thoron together with radon [15], but segregation is not possible by its electronics. If the electronics of an IC are set to reject the α-particle (^{214}Po) with the highest energy (7.7 Mev), which is the last in the chain of the short-lived decay products of ^{222}Rn, fast response times can be obtained [13]. The AG P-series can measure ^{220}Rn only if the following setups are used: i) Flow through method with delay time [16], and ii) Flow through method with delay volume. These setups are described in [17]. The AG PQ PRO RnTn and the AG D2000 can measure ^{220}Rn by using two measurements. The first measurement is achieved by using flow-through to get the gas into the probe, while the second measurement is achieved by diffusion. Using these different measurements, ^{220}Rn can be assessed.

SCD are used by the RAD7 probe (Durringe) and RPP-U (Piketronic). The RAD7 has a 0.7 L container. The SCD is a solid state-Ion-planted, planar, Silicon alpha detector. When a ^{222}Rn nucleus decays, the positively charged ^{218}Po is accelerated by the high-voltage (HV) of 2.4 to2.4 kV, producing a pulse proportional to the energy of the α-particle distinguishing between the 5.49 MeV of radon and 6.29 MeV of thoron. The RPP-U works on the same principle but cannot distinguish radon from thoron.

In SC, flashes are emitted when radiation, like α-particles, interacts with the walls of the container which is coated with a scintillating material like ZnS(Ag). These flashes are converted to an electric pulse by a photomultiplier. This probe counts the pulses and calculates the concentration. The Pylon EB-7 is such a commercially available device.

The probes mentioned using IC and SCD detectors can measure Rn gas flux using the following two setups: i) Diffusion Accumulator ii) Flow-through.

An accumulator chamber, i.e. a box open at one side, is placed upside down on the ground after vegetation is removed. The accumulator is sealed by wet soil. The radon probe is placed inside the box and set to diffusion mode. Measurements are recorded in 10-minutes intervals. Nine consecutive measurements are sufficient to calculate the slope of the best fitting line. The exhalation is calculated by equation (1),

$$F \approx \frac{V}{A}\frac{C(t)}{t} \quad (1)$$

where F is the exhalation rate ($Bq{\cdot}m^{-2}{\cdot}h^{-1}$), V is the net air-volume of the accumulator (m^3), A is the surface covered by the accumulator box (m^2) and C(t)/t is the slope of the best fitting line [18].

In the flow-through method the accumulator is placed in the same manner. However, in this case, the air in the chamber is continuously removed at a constant rate and the radon concentration in the exhaust stream is measured [3]. Exhalation is calculated with the same methodology applied as in the accumulation method.

By using the two methods mentioned above, the total Rn concentration is measured. To measure only the ^{222}Rn concentration (excluding ^{220}Rn), the flow through method with delay time (Figure 2c) is used. The gas is collected from the accumulator and is pumped into a canister where it stays for 10 times the half-life of ^{220}Rn, i.e. 56 s, to eliminate its concentration in the measured gas. Then the gas is pumped into the pulse-counting, open-air ionization chamber to measure the ^{222}Rn concentration [4]. Exhalation is calculated by applying the same methodology as described in the accumulator method. To measure both ^{220}Rn and ^{222}Rn, the flow method with delay volume is applied (Figure 2d). Two radon probes are used (pulse-counting, open-air ionization chambers). The gas is collected from the accumulator and pumped into the first detector where the sum of both ^{220}Rn and ^{222}Rn concentration is measured. Then the gas is pumped into a canister where it stays for 10 min. Thus, the second detector

measures only ^{222}Rn concentration. By subtraction of the two concentrations the ^{220}Rn can be calculated [5].

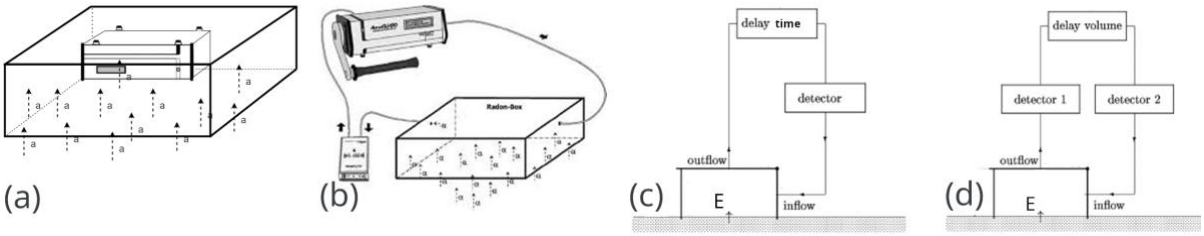

Figure 2. *Direct measurement methods: a) diffusion b) flow through c) flow through with delay time d) flow through with delay volume*

Indirect methods calculate radon gas concentration by i) measuring ^{226}Ra concentration in soil from its progenies or ii) by the terrestrial γ-radiation measured. In the first method, soil samples are collected from the site, dried for 8 hours in an oven and then pulverized. Then the sample is placed into an airtight petri dish. These samples must be kept for 30 days before γ-spectroscopy, to allow ^{220}Rn to reach secular equilibrium with ^{226}Ra contained in the dish. With the use of a high purity germanium detector (HPGe), the photopeaks of ^{214}Pb and ^{214}Bi yield their concentrations, which equal the concentration of ^{222}Rn captivated in the dish, which in turn is in secular equilibrium with ^{226}Ra. With this method, the total Gamma Dose Rate (GDR), $D_{tot,}$ by natural radioactivity is acquired.

In order to calculate the ^{226}Ra contribution (D_{Ra}) to the dose (D_{tot}), the empirical equation **Error! Reference source not found.**) [12] is used.

$$D_{Ra} = 0.24 \times {D_{tot}}^{1.01} \quad (2)$$

The ^{226}Ra activity is then derived using the hypothesis that ^{226}Ra activity is closely related to ^{238}U activity [19].This hypothesis is not often valid because of the delusive radium, compared to uranium. Exhalation is obtained by using the radium activity with the use equation (3) [12].

$$F = A_{Ra}\, \lambda_{Rn}\, f\, \rho\, (1-\varepsilon) \left[\frac{D_e}{\lambda_{Rn}}\right]^{\frac{1}{2}} \quad (3)$$

where F is the radon flux (Bq m^{-2} s^{-1}), A_{Ra} the ^{226}Ra activity in (Bq kg^{-1}), λ_{Rn} is the decay constant of ^{222}Rn, f is the emanation material coefficient, ρ is the soil density, D_e is the bulk material diffusion coefficient, which is influenced by soil characteristic and conditions (temperature and relative humidity), and ε is the material porosity when dry. Typical geological parameters, for soil characteristics are f (0.23), ρ (1.5•10^3 kg m^{-3}), D_ε (2•10^{-7} m^2 s^{-1}) and ε (0.25) [20].

In the second indirect method, the terrestrial gamma radiation method is used. Gamma probes measure the terrestrial γ radiation, which is used as a proxy, to calculate the geogenic ^{222}Rn. The progenies ^{214}Pb and ^{214}Bi of radon emit γ-radiation which contribute to the total γ-radiation derived by cosmic rays, natural radioactivity, anthropogenic radiation, e.g. Cs^{137} and the inherent background of the probe. By measuring the terrestrial γ radiation and subtracting the non-geogenic and natural occurring U, Th, and ^{40}K dose radiation fraction, the radon gas and its progenies can be deduced [4]. Thoron can be measured by direct passive probes by subtracting the measurement acquired after letting the gas rest for 10 minutes from the initial measured concentration of the gas. After this time ^{220}Rn has almost completely decayed. For direct passive methods two probes can be used, one measures the initial gas mixture, while the second measures the thoron-free gas.

DISCUSSION

Indirect methods (Ra from soil and γ spectroscopy) are easier to apply for prospecting larger areas but are not as accurate as direct methods. The calculation of radon exhalation with the use of ^{226}Ra in

soil samples includes many variables which are difficult to obtain (soil parameters) and involve time costly laboratory work. Direct passive methods (Electret Ion Chambers, Etched Track Detectors, and Charcoal Adsorption) are cheap alternatives which provide accurate measurements but also need laboratory work which slow down the final results. Comparison of direct passive (CR-39) and active method (IC, AlphaGuard) showed similar measurements 28.7±4 Bq/m^3 for CR-39 and 34.6±3 Bq/m^3 for AlphaGUARD) [21]. Direct active methods provide quick, easy and accurate in situ measurement for Rn. Older IC probes require complicated setups for ^{220}Rn and ^{222}Rn segregation, like the flow through with delay time [16] and flow through with delay volume methods [12]. The newer AG D2000 IC probe models have built-in capabilities with delay-time procedures. SCD probes can segregate the α-particle energies derived by ^{220}Rn and ^{222}Rn but have smaller concentration measurement range (4 Bq/m^3 -750 kBq/m^3) compared to IC probes (2 Bq/m^3 – 2 MBq/m^3) (Table 1). The comparison of the coefficient of variance (SV) for IC and SCD using measurement data obtained by published work [19] shows that the SCD (RAD 7) can measure radon exhalation with a CV 4.34%. The CV of the IC (AG PR2000) is 8.55%. The RPP-U measures radon exhalation with a CV 11.16%.

Table 1. *Comparison of Continuous Radon Probes. PIC: Pulse Ionization Chambers, SCD: Solid State Detectors, SDPD: Solid State Photodiode Detector, SC: Scintillation Cell, α: a-spectrography, ^{218}Po: Polonium 218 (data acquired from the manufacture's datasheets) († former Genitron, Saphymo, *former Tesla. +former SunNuclear)*

Manufacturer	Probe	Model	Detector	Measuring Method	Sensitivity	Measuring Range min (Bq/m³)	Measuring Range max (KBq/m³)	Chamber size (Liter)	HV (Volt)	Thoron
Bertin†	AlphaGUARD	PQ2000PRO	PIC	^{222}Rn	1 cpm at 20 Bq/m3	2	2000	0,56	750	No
Bertin†	AlphaGUARD	PQ2000PRO RnTn	PIC	^{222}Rn	1 cpm at 20 Bq/m3	2	2000	0,56	750	Yes
Bertin	AlphaGUARD	D2000	PIC	^{222}Rn	1 cpm at 20 Bq/m3	2	2000	0,56	750	No
Bertin	AlphaGUARD	DF2000	PIC	^{222}Rn	1 cpm at 20 Bq/m3	2	2000	0,56	750	Yes
Radonova	ATMOS	NA	PIC	^{222}Rn	20 cpm at 1000 Bq/m³	1	100	2,2	NA	NA
Durringe	RAD	7	SCD	^{218}Po	16.7 cpm/(Bq/m³)	4	750	0,7	2100-2400	Yes
Piketronic*	RPP	U	SCD	^{218}Po	0.25 cpm/(Bq/m³)	20	100	0.76	NA	No
SunRadon+	NA	1027	SSPD	^{222}Rn	2.5 cph/pCi/l	3.7	370	NA	NA	No
Pylon	AB7	600A	SC	^{222}Rn	0.37 cpm/(Bq/m³)	27.4	NA	0,272	NA	Yes
Pylon	AB7	610A	SC	^{222}Rn	0.021 cpm/(Bq/m³)	48.1	NA	0,154	NA	Yes

In 2010 an inter-comparison of direct and indirect methods to determine radon flux was conducted. The compared radon measurement methods were i) active direct methods: α-spectrometry by using an IC (AG probe) and a Solid State Photodiode Detector (SSPD) (model 1027 Sun Nuclear probe), ii) passive direct methods used were Electret Ion Chambers (E-PERM) and charcoal adsorption, and iii) indirect methods consisted of the natural occurring γ-dose (RSS-112 and the GammaTRACER), and iv) radium concentration obtained from various databases like the EURDEP [22] and by in situ collected samples [12]. Results showed a good agreement among the direct method (continuous and integrated with CV=10-23%. The indirect methods were between 20-40% of the values obtained by direct methods. By comparing three radon probes using two different detection technologies, namely IC (AlphaGUARD, ATMOS) and SCD (RAD7) in a spa environment for 24 h the time series showed similar temporal variability with minor outliers from the IC probes. The source of these outliers have not yet been explained [23].

CONCLUSIONS

Direct methods produce more reliable measurements than indirect methods for measuring Rn exhalation [12]. Direct passive methods (CA, EIC, and ETD) are more reliable than indirect methods but demand more time in the laboratory to obtain final results. Direct active measurement methods combine accuracy, ease of use, portability and time efficiency. Exhalation results from the SCDs (RAD7) probe present lower CV (4.34%) than probes using ICs AG, which present CV of 8.55%. SCD monitors have lower measurement range, (4 Bq/m^3 -750 kBq/m^3) compared to IC probes (2 Bq/m^3 – 2 MBq/m^3). Furthermore, SCDs can distinguish radon from thoron by their α-particle energies. This does

not apply for the PRR-U, which uses an SCD (CV=11.16%) and no built-in ^{220}Rn measurement ability). ICs and direct passive methods need special setups which allow thoron to decay before radon is measured. In newer IC probes this setup is built in the instrument.

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