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An Overview of Instrumentation for Measuring Radon in Environmental Studies

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Abstract: A number of techniques have been used to measure the concentrations of (^{222}Rn) and their decay products in the environment. Three characteristics were used to describe the radon measurement techniques: (i) whether the technique measures ^{222}Rn or its daughter products; (ii) time resolution and (iii) radioactive detection of the type of emission either alpha, or beta particles or gamma radiation resulting from radioactive decay. Most common methods rely on detection of alpha particles. Sometimes a single alpha particle (e.g., ^{218}Po for ^{222}Rn) is detected to measure radon isotopes (as in air-in-monitors, RAD7, Durridge, USA) or by counting all three alpha particles produced in the decay of ^{222}Rn (i.e., ^{222}Rn, ^{218}Po, and ^{214}Po) using scintillation counters. Some methods are based on the detection of gamma-ray emitted radionuclides during radioactive decay of the progeny of ^{222}Rn (^{214}Bi, ^{214}Pb) and only a few methods utilize beta decays. The present work discusses the various methods available for radon measurements from different matrices causing radon release to the environment**,** wherever applicable, the sensitivity and quality assurance/quality control (QA/QC) aspects of these techniques are also explored.

Keywords: Radon measurements - Techniques- Environment.

1 Introduction

Three natural isotopes of radon namely, radon (Rn-222), thoron (Rn-220) and actinon (Rn-219) resulting from the radioactive decay of the uranium, thorium and the actinium series. ^{222}Rn consists of the decay of ^{226} Ra, the direct parent of the ^{238}U series, while its ^{220}Rn decomposes from ^{224}Ra, a member of the ^{232}Th series. Actinon produces 223Ra decay from the ^{235}U series, and is usually neglected because its presence is negligible in the atmosphere. Radon is spread as a gas element in the natural radioactive chain of the earth's crust [1]. Radon stems from rocks and soil and tends to concentrate in indoor places such as mines or underground homes. Other sources, including building materials and water extracted from wells, are less important in most circumstances [2]. The presence of radon in the free atmosphere was first noted by Elster and Geitael around 1901[3]. The main radiation dose received by the population comes from radon gas. Many studies in Europe and North America confirm that radon gas causes a large number of lung cancers in the general population is radon gas [4]. As the risk of developing lung cancer increases proportionately to increased exposure to radon gas. Because many people are exposed to low concentrations of radon, most lung cancers with radon are produced at low levels. Thus it is believed that radon is the second leading cause of lung cancer after smoking [5].

Such as the national reference level for radon, the maximum allowable concentration of radon in a residential dwelling, an important component of the national program. For homes with radon concentrations above these levels, corrective or required measures may be recommended. In establishing a reference level, different national factors such as radon distribution, the number of existing houses with high radon concentrations, the mean radon level and the prevalence of smoking should be considered. In the light of the latest scientific data, WHO proposes a reference level of 100 Bqm-3 to reduce the health risks of exposure to indoor radon [6]. Radon has many roles in

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ground water studies like (i) dating, (ii) ground water movements (iii) aquifer characteristics etc. It is pertinent that reliable and sensitive techniques are made available for radon studies [7].

Radon Measurements

Radon measurements are often discussed in terms of either a short-term or long-term test [8]. A short-term test for radon, using a Lucas scintillation cell, activated charcoal detector or another type of detector such as an electret ion chamber, can provide a first indication of the mean long-term radon concentration in a home. However, the daily and seasonal radon changes should be taken into consideration when conducting radon measurements in the short term.

Because high radon concentrations usually occur during periods when "houses are closed", short-term measurements during this period or season can over-estimate the average annual radon concentration. Alternatively, a short-term radon measurement conducted during a period when the home has increased ventilation (eg, open windows) can significantly reduce the average annual radon concentration. [9]. Most important active methods for radon measurements in air employ collection of the gas in scintillation flask / ionization chamber or suction of the gas through a two filter sampler. On the other hand, progeny levels are measured by collecting them on filter papers and counting for radioactivity at suitable intervals. Major difference in the methods of measurements is that in radon gas estimation using scintillation cell, the progeny in the sampled air is removed by filtration before the gas is collected in the cell. Active devices used by many countries included electronic fusion devices (EIDs) and continuous radon monitoring devices (CRMs). The passive devices do not require the use of electric power or the pump to operate in the preparation of samples, while the active devices require electricity and include the ability to draw radon concentrations and vibrations during the measurement period. For homes, ATDs are a common option for long-range radon measurement, often deployed for one year, while EICs are often used for short periods (eg, several days) to medium (eg weeks to months) [10].

Table 1: shows radon gas measurement devices and their characteristics [9].

Detector Type	Passive / Active	Uncertainty[a] [%]	Sampling Period	Cost
Alpha-Track Detector (ATD)	Passive	10-25	1-12 Months	Low
Activated Charcoal	Passive	10-30	2-7days	Low
Electret Ion Chamb	Passive	8-15	5days-1year	medium
Electronic Integrating Device (EID)	Active	~25	2days-year (s)	medium
Continuous Radon Monitor (CRM)	Active	~10	1hour-year (s)	high

[a]Uncertainty expressed for optimal exposure durations and for exposures ~ 200 Bq/m^3

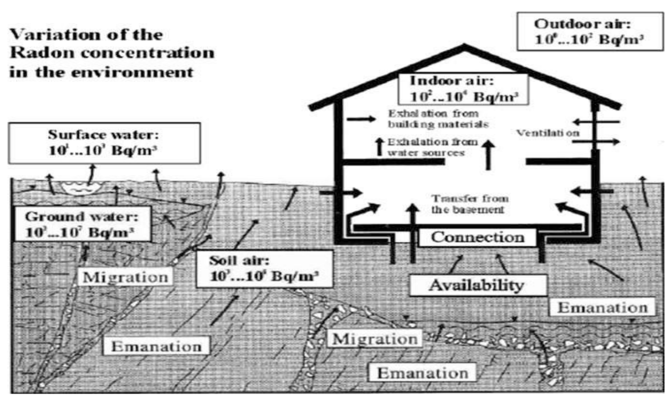

Fig. 1: Concentrations of radon in the environment)(figure taken from http://www.sarad.de/).

Radon Gas Detectors

The ranges of radon concentrations in environmental samples are given in Fig. 1. Radon concentrations in surface waters including ocean waters are much lower compared to subsurface groundwater and the methodology of sampling and/or analytical methods often differ. Radon concentrations in atmospheric air also are generally low compared to soil air. Radon measurement techniques are classified based on three characteristics (Fig. 2).

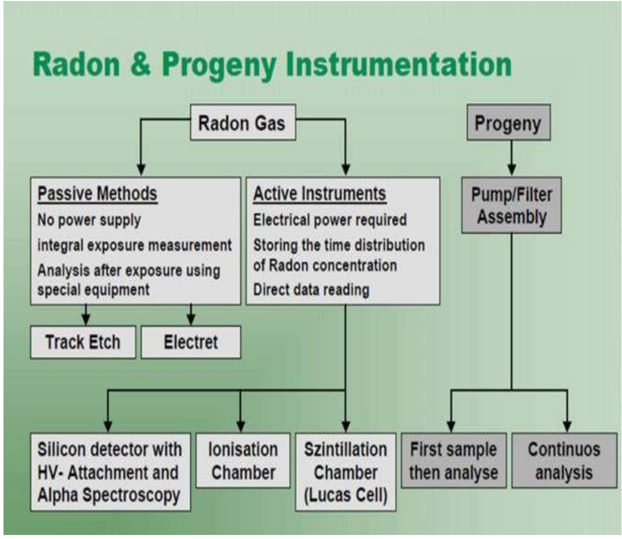

Fig. 2: Methods and Instruments to measure radon and its progeny (taken from http://www. SARAD.de/)

Alpha-Track Detectors

Filtered Alpha Track Detection

ATD is a small piece of specially produced plastic substrate inside a filter-covered diffusion chamber that excludes the entry of radon decay products as shown in Figure 3. Plastics are generally polylyl diglyl carbonate (PADC or CR-39), cellulose nitrate (LR-115) or polycarbonate (Makrofol). When alpha particles are generated by radon or radon decay products close to the detection material, they can collide with detection materials, producing microscopic areas of damage called latent alpha pathways. The chemical or chemical pattern of the plastic detector extends the size of the alpha pathways, making them observable through optical microscopy so that they can be counted either manually or by an automatic counting device. A minimum detectable concentration (MDC) of 30 Bqm-3, calculated by methods discussed elsewhere for a 1 month exposure is generally achievable for ATDs [9-10].

Unfiltered Track Detection

The unfiltered alpha track detectors operate on the similar idea as the alpha track detector work. Here is only one difference that, it has no filter to detach radon progeny and other alpha particle ejectors. Presently, it has been recommended by EPA that such instruments should not be utilized, when the equilibrium fraction is smaller than 0.35 or higher than 0.60 without modifying its calibration factor.

Activated Charcoal Adsorption (ACD) Detectors

ACDs are passive devices deployed for 1-7 days to measure indoor radon. The principle of detection is radon adsorption on the active sites of the activated carbon. After sampling, the detector is sealed and the radon decay products equilibrate with the collected radon. After a 3 hour waiting period, the collectors can be directly gamma counted, or analytically prepared for liquid scintillation counting techniques. In the gamma counting method, the charcoal canisters or bags contain 25-90 g of activated carbon. In the alpha counting method, 20 ml liquid scintillation vials containing 2-3 g of activated carbon are used. The canisters can be open or equipped with a spread barrier to extend the measurement period to 7 days. Because the response of ACD devices is affected by humidity, they must be calibrated under different levels of humidity. Devices must be calibrated over exposure periods and temperatures likely to be encountered in the field. The method only provides a good estimate of the mean radon concentration on exposure time if changes in the radon concentration are small. The use of the propagation barrier reduces the effects of drafts and high humidity. Since radon decomposes with a half-life of 3.8 days, the detectors must be returned for analysis

Fig. 3: Twin cup dosimeter with ATD

Continuous Radon Monitoring

This method includes devices that can record radon time measurements continuously. The air either diffuses or pumps into a counting chamber. The counting chamber is in particular a scintillation cell or ionization chamber. Scintillation counts are processed by electronics and radon concentrations are stored for predetermined periods in the device memory or are transferred directly to the printer.

Electret Ion Chambers (EIC)

In this method radon gas, but not decay products, enters the chamber by passive diffusion through a filtered inlet. Radiation emitted by radon and its decay products formed inside the chamber ionizes the air within the chamber volume. The negative ions are collected by the positive electret located at the bottom of the chamber. This in turn is related to the radon concentration. The electret discharge in volts is measured using a non-contact battery-operated electret reader. The long-term EICs measure radon over 3 to 12 months at a concentration of 150 Bqm^{-3}[13, 14].

Electret Ion Chamber: Long-Term

In this technique there is an electret (electrostatically charged disk detector) inside a small ion chamber (container). During the measurement period, radon is spread through a filter-covered opening in the chamber, where the ionization caused by the decay of the radon and its offspring reduces the electrical voltage on the electrit. EL detectors can be deployed for 1 to 12 months. Since the ion chambers are true integrators, the EL type can be displayed at shorter intervals to sufficiently high levels of radon.

Electret Ion Chamber: Short Term

An electrostatically charged disk detector (electret) is placed in a dwarf container (ion chamber). During the measurement period, radon diffuses via a filter-covered opening in the chamber, where the ionization caused by the decay of radon and its decay products declines the voltage on the electret. ES detectors can be deployed for 2 – 15 day as electret-ion chambers are true integrating detectors, the ES type may be exposed for prolong time intervals if radon concentrations are adequately stubby.

Grab Radon/Activated Charcoal

For this method a skillful instructor is required to sample radon through a pump or a fan to drag air via a cartridge occupied with activated charcoal. It may take about 15 minutes to 1 hour time period for sampling. Afterwards, the cartridge is put in the sealed container and brought to a laboratory where analysis is almost the similar as in the case of LS techniques.

Grab Radon/Scintillation Cell

An experienced and skillful technician hauls air throughout a filter to remove radon progeny into a scintillation cell. To examine an air specimen, the window end of the cell is positioned in a photomultiplier tube to count the scintillations (Bright light flashes) induced when alpha particles from radon decay strikes the zinc sulfide coating on the inside of the cell. An evaluation is done to convert the counts to radon levels. It may take about an hour to accomplish this test.

Three-Day Integrating Evacuated Scintillation Cell

In its experimental structure, the scintillation cell is fitted with a negative pressure gauge and a restricted valve. Prior to diffusion, the scintillation cell is evacuated. In the sample location, the skilled worker notices negative pressure readings and opens the valve. The flow across the valve is very slow so that it takes more than a sample period for 3 days to fill the scintillation cell. At the end of the sample time period, the valve is closed, the negative pressure gauge is read down, and the call is re-dialed to the laboratory

Electronic Integrating Devices (EID)

Most EIDs use a solid state silicon detector within the propagation chamber to calculate alpha particles emitted from radon decay products. Because of the small dimensions of the propagation chamber, long integration times (more than two days) are often necessary to read statistically stable at moderate radon concentrations. Higher sensitivities can be achieved by applying high voltage to the assembly of electrolytic decay products with direct contact with the detector. High humidity may affect the measurement. MDC of 20 Bqm^{-3} is typical for a 7 day exposure period. For several popular EIDs, the ability to routinely calibrate these detectors is lacking.

Continuous Radon Monitors (CRM)

There are several types of commercially available CRMs using various types of sensors including scintillation cells, current or pulse ionization chambers, and solid state silicon detectors. CRMs either collect air for analysis using a small pump or by allowing air to diffuse into a sensor chamber. All CRMs have electrical circuitry that provide a summary report, and often a time-resolved recording, which allows the calculation of the integrated radon concentration for specified periods. The different types have their specific advantages. For example, when using solid-state silicon detectors, alpha spectrometry is possible allowing discrimination between radon and thoron [15].

Fig.4: CRM using scintillation cell

Fig.5: CRM based on electrostatic collection

Radon in Water Mmeasurements Devices

The presence of radon gas in groundwater is mostly due to the disintegration of radium (^{226}Ra) found in rocks and soils. Radon can also be generated in water distribution systems with high radium concentrations from radium adsorbed iron pipe scales [16]. Radon exposure may occur from sources of waterborne radon, either from ingestion or inhalation of radon emitted from water. The risk of cancer caused by the release of waterborne radon (bathing, dishwashing, etc.) is much greater than the risk of drinking radon water [17]. A commonly used estimate of the radon-water transfer coefficient for homes in North America is 1.0×10^{-4} [18]. In most parts of the world, radon in indoor air is released from water-based sources much less than radon emitted from ground sources down the house. There are several established ways to collect and measure radon in water [16]. Radon measurements in water include direct gamma [19], electret ion cells [13], and gas transfer via membranes [20]. The counting of liquid luminescence and radon emission-measurement techniques is the most common method for measuring radon concentrations in water [21] and will be discussed in detail.

Liquid Scintillation Counting

Liquid scintillation counting (LSC) is the most sensitive and widely used method to measure radon in water. The popularity of liquid scintillation for radon analysis is due to several factors including the excellent accuracy and precision of the method, the low level of detection, the limited need for sample preparation. The ability to rapidly measure a large number of samples, and the ability of the counter to change samples while unattended. The LSC technique quantifies the activity of radon and decay products from the rate of photons emitted from the scintillation fluid [16, 21]. Limitations of the LSC technique include the initial start-up cost to purchase the counter and the need to perform the analyses in a laboratory. a delay of about 3 hours to establish radioactive equilibrium between radon and its decay products. Depending on the counting time, a detection limit below 1 BqL^{-1} can be achieved. Besides EIC, two other techniques for measuring radon in water are direct gamma counting and gas transfer by membranes [18].

Fig.6: Lucas scintillation cell

For measuring the relatively low concentrations, it is recommended to use a large collection volume and then concentrate the sampler into a smaller volume for counting. Pre-concentration can be achieved by passing air through the active charcoal trap immersed in liquid nitrogen. At this temperature, radon is kept in the coal. This coal is heated in a chamber resulting in the release of trapped radon. This is then collected from the closed chamber using the scintillation cell or ionization chamber and the concentration is estimated using the method described above.

Measurement of Radon/Thoron in Water and Air by Durridge RAD7

The Durridge RAD7 is a precisely versatile instrument for radon measurements. The RAD7 is a rugged and long-lasting piece of equipment. The RAD7 is a sophisticated quantifying device widely used in laboratories and research work all over the world, by radon testers and home inspectors, in mines and deserts, on the ocean and up volcanoes, at extremes of temperature (Figure7).

The RAD7 is also the simplest computer-driven electronic detector to use, with pre-arranged setups for common practices. It's made to resist day-to-day use in the field. A rugged, handsome case encloses the detector, which is self-contained and self-sufficient. The RAD7 arrives complete, along with a built-in air pump, rechargeable batteries, and a wireless infrared printer. If required, the detector may collect data and reserve it for later printing or downloading to a PC. [22-25].

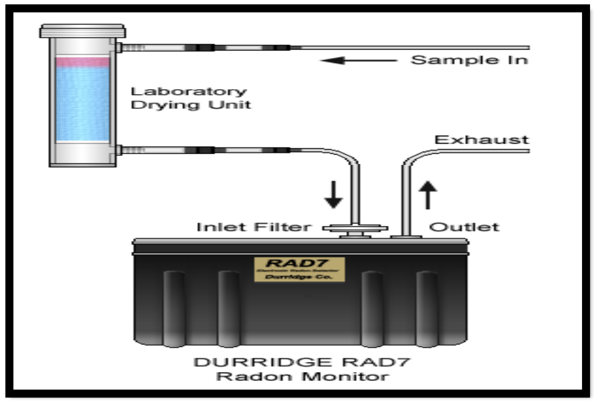

Fig. 7: Block diagram of RAD 7

Alpha GUARD

The Alpha GUARD PQ2000 PRO is the center piece of a compact portable measuring system for the continuous determination of the radon- and radon progeny concentration in air as well as selected climatic parameters. The Alpha GUARD uses the proven principle of the pulse ionization chamber (Alpha GUARD 2012). The measurements can performed using radon detector (ALPHA GUARD) connected through the air pump to a specific kit of glass vessels Aqua KIT, shown in Fig .8. It was adjusted to continuous flow of 0.5 L/min. Each water sample was submitted to measurement during the time interval of about 60 minutes. Detected 222Rn activity levels were analyzed by the computer using the software DataEXPERT by GENITRON Instruments. Obtained data were processed taking into account the volume of water sample and its temperature, atmospheric pressure and the total volume of the air in the vessels [26].

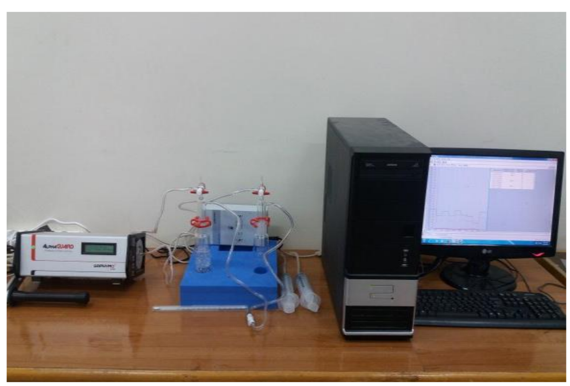

Fig.8: Detector Alpha GUARD (Genitron Instruments) in measurements of radon activity concentration in water.

Radon Measurements Using Gamma-Ray Spectrometry

Atmospheric ^{222}Rn concentrations can be measured by measuring ^{214}Pb and ^{214}Bi by gamma spectrometry. This particular method may be appropriate where there is temperature inversion leading to very little or no vertical movement of air masses. Also, no major wet precipitation event should have taken place within 2 h of sampling, as wet precipitation events will most likely remove ^{218}Po, ^{214}Pb, ^{214}Bi, but not ^{222}Rn [27].

Conclusions

A large number of methods for 222Rn have been developed (and a few methods of thoron) for a number of sample matrices and for different applications. The accuracy of the measurements depends on the applications. The spatial and temporal variation of radon in the middle of the ocean (soil and groundwater air) limits the accuracy required. Overall, there is enough.

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