

Chapter 2

Radon Measurement Techniques

2.1 Introduction

A number of techniques have been utilized for measuring the concentrations of radon (^{222}Rn) and its daughter products in the environment. The specific analytical method utilized depends primarily on the concentrations of ^{222}Rn in the medium and the precision needed. The ranges of radon concentrations in environmental samples are given in Fig. 2.1. 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. 2.2): (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 (details given later). 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.

There are three broad classes for time resolution when it comes to sampling and analysis (Fig. 2.2): (a) *grab-sample* technique involves measurement of ^{222}Rn in discrete sample of water (or air) collected over a very short time (compared to the mean-life of ^{222}Rn) at a single point. Examples include measurements of radon in groundwater, freshwater and marine waters, and air samples. Radon measurement equipment such as RAD7 can be used to measure ^{222}Rn in air at any time when it is used in ‘sniffer’ mode in which case radon is typically present with minimal ingrowth of its progeny and large number of measurements can be taken in a relatively short

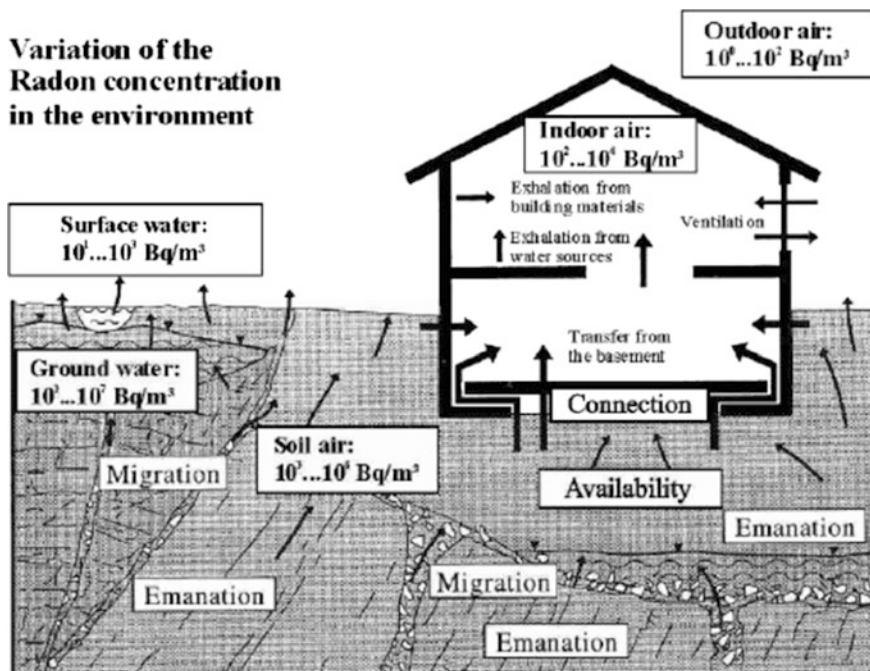


Fig. 2.1 Concentrations of radon in the environment (figure taken from <http://www.sarad.de/>)

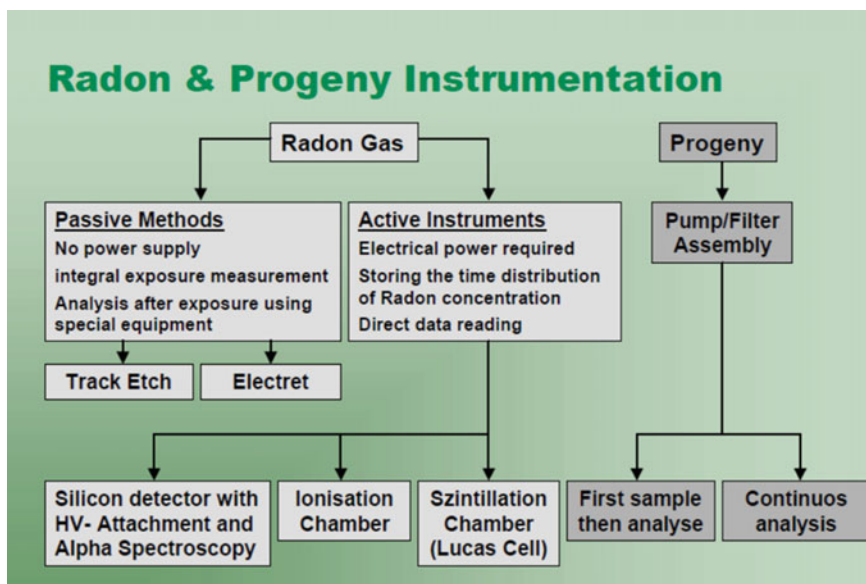


Fig. 2.2 Methods and Instruments to measure radon and its progeny (taken from <http://www.SARAD.de/>)

period of time; (b) *continuous* technique provide time-series concentrations of ^{222}Rn in samples (soil gas, air, water) and sampling and counting are done simultaneously. Recently developed instruments such as Smart Radon Duo (www.roopcreators.com) and Scintillation Radon Monitors (www.eeipl.in) can also be used for continuous monitoring of ^{222}Rn as well as ^{220}Rn in air, soil gas or water. Such data is quite valuable in the usage of ^{222}Rn as a tracer for predicting earthquakes (Chap. 10). Many of the continuous monitors are portable and nearly all of those are designed to detect alpha radiation (by ionization chamber, gross alpha counting or alpha spectrometry). It is more difficult to build a portable beta detector or gamma-ray spectrometer that has both low background and high sensitivity; and (c) *Integrating* technique provide the integrated concentration over a certain period of time. Such measurements are useful to determine monthly or annual average ^{222}Rn concentration of a specific building. The passive detectors, which are quite inexpensive, are examples of integrating techniques. Such devices are useful to assess the effectiveness of remedial techniques to alleviate indoor radon problems. Additionally, integrated results can also be determined from continuous long-term measurement records.

A review of the most commonly utilized ^{222}Rn measurement techniques are reviewed in this chapter. Furthermore, wherever applicable, the sensitivity and quality assurance/quality control (QA/QC) aspects of these techniques are also explored.

2.2 Techniques for Measuring Radon

2.2.1 Grab Sampling Methods

2.2.1.1 Air Samples

Radon-222 measurements in air (both surface and upper atmospheric) are made either with direct measurement of radon (direct method) or its progeny (indirect method) (e.g., Papastefanou 2002). For direct measurements, grab samples have been collected either in flasks, stainless steel containers, in situ absorption of radon onto cold plates, or activated charcoal. The flasks or steel containers are brought to the laboratory and the radon is extracted onto activated charcoal column and subsequently transferred to Lucas cell (details given in Sect. 2.2.1.4). The cold plates can be heated inside a closed chamber and the released gas is filled in a Lucas cell for assay.

Indirect measurements are based on the collection and counting of radon progeny, but this method is based on the assumption of equilibrium between radon gas and its aerosol progeny. Such an assumption is considered to be valid for sites that are at a significant distance from the radon's terrestrial source and the weather conditions of the air masses are fairly calm en route to the sampling site. This method is likely to fail in the case of precipitating air masses (Chambers et al. 2014). Chambers et al. (2014) have summarized the radon data measured with

different techniques and resolution in the Antarctic over 5 decades (1958–2011; papers published in 1960–2013).

Although a number of sampling methods are available for surface air, the most common method for layers above planetary boundary layer involves collection of air samples using a manifold that is comprised of an inlet, fore-pump, main compressor, pressure monitoring sensors and relief valves, data system, and steel sample cylinder (Fig. 2.3). The sample cylinder has a water volume of 2.5 L and a pressure rating of 1800 psi, so that when it is fully pressurized it can hold 300 STP L (or 0.300 m³ at STP) of ambient air (details given in Kritz et al. 1998). Williams et al. (2011) used a 10-trap radon sampler installed in an underway pod of the instrumented motorized glider, and air samples were collected over 5 or 10-min at a flow rate of 20–30 L min⁻¹ at standard temperature and pressure. The

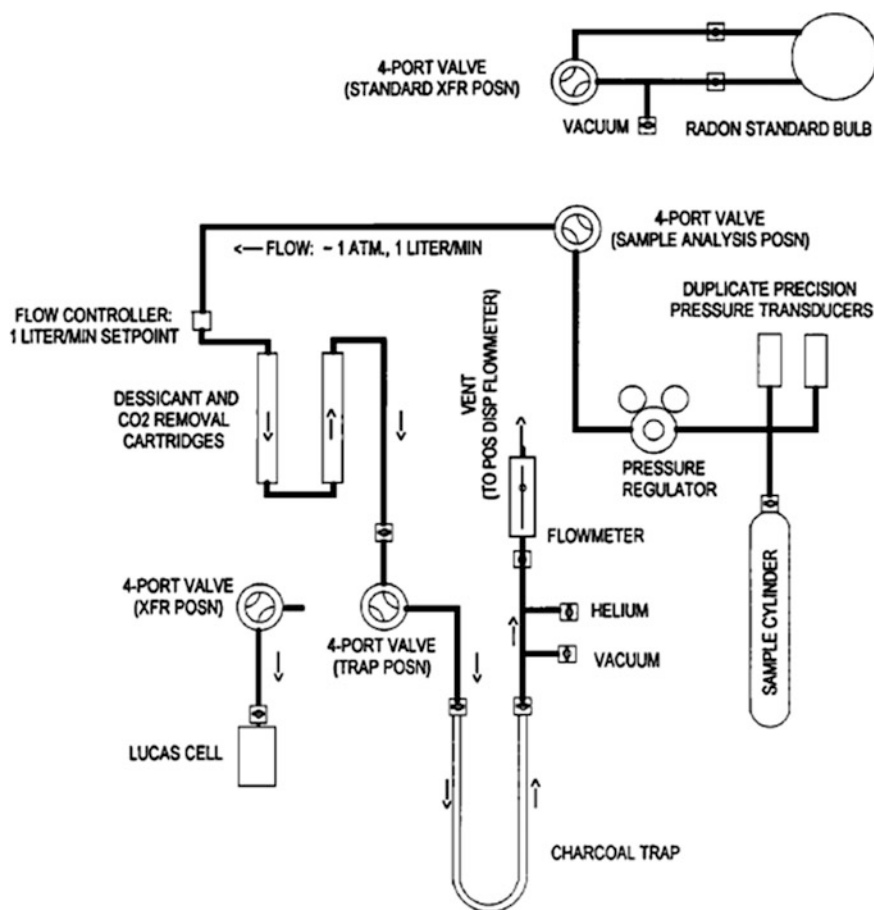


Fig. 2.3 Schematic diagram of ²²²Rn transfer and extraction system (figure from Kritz et al. 1998, with permission from American Geophysical Union)

filling procedure consisted of a short flush with both valves open, after which the exit valve was closed and the sample was collected. The model used by Kritz et al. (1998) had dimension of $\sim 4 \text{ m}^2$ of cabin floor space, weighing $\sim 500 \text{ kg}$ with $\sim 4.5 \text{ kW}$ of electrical power. The filled cylinders are brought to the laboratory where the Rn was stripped out by cold trapping. The cylinder is connected via $\sim 3 \text{ mm}$ stainless tubing to the trapping/transfer system. The air from the cylinder (with 2 ports, each one with its own solenoid valve) passes through a regulator, where its pressure is reduced to $\sim 1 \text{ atm}$ above ambient air through a flow controller where the flow is regulated at a constant 1 STP L/min . After passing through drierite (to remove water vapor) and ascarite (to remove CO_2) columns, the air flows through an activated charcoal column immersed in a dry ice—alcohol mixture at -78°C ,¹ where the radon contained in the air stream is trapped by the charcoal (Fig. 2.3). Note that the design of the trapping-transfer systems and the associated operating procedures and calibration procedures are similar to those described in Broecker and Peng (1971), Mathieu et al. (1988) and are often modified by different groups (e.g., Kritz et al. 1998).

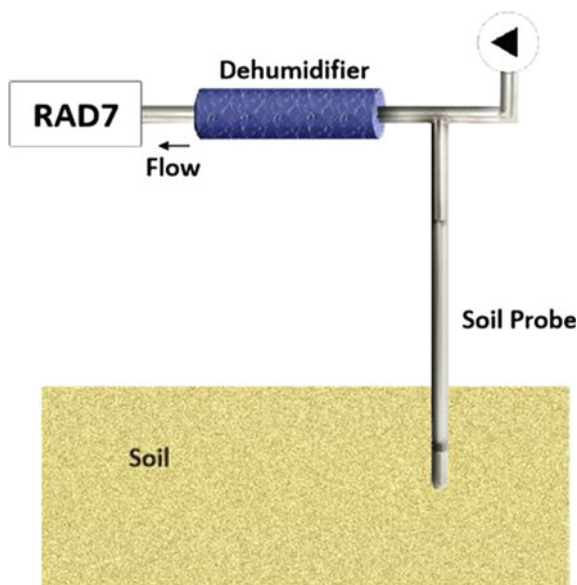
The charcoal column containing radon is then heated to 480°C (note: Kritz et al. (1998) reported heating to 300°C) and then, slowly back-flushed with helium to transfer the trapped radon into a Lucas cell for counting. Kritz et al. (1998) reported $\sim 8 \text{ h}$ to process eight samples in parallel. In the case of large volume air samples collected in stainless containers, the quantity of air passing through each trap is determined from the known volume and the beginning and ending pressures of each sample cylinder. The volume of air is also confirmed by passing air through a calibrated positive displacement flowmeter downstream of the trap. Kritz et al. (1998) reported radon trapping efficiency of $>99\%$ by placing two traps in series (trapping efficiency (%) = $[1 - \{^{222}\text{Rn trapped in the second trap}/^{222}\text{Rn trapped in the first trap}\}] * 100$). The fraction of ^{222}Rn transfer from the trap to the counting was reported to be $95 \pm 3\%$.

2.2.1.2 Soil Air Samples

Radon concentrations in soil gas is about 2–5 orders of magnitude higher than that in atmospheric air. One of the relatively easier ways of measuring radon in soil gas is using Soil Gas Probe coupled with RAD7 (Fig. 2.4, http://www.durrIDGE.com/products_soil_gas_probe.shtml). This soil probe can be operated anywhere above the water table and is used in conjunction with a drying unit (drierite column). The soil air can be sampled in the GRAB mode or Continuous Monitoring mode using the standard protocol. Continuous monitoring method is simpler and provides time resolution in the event of weather changes. RAD7 will pump air for the first 5 min of every half-hour cycle, and then for 1 min for every 5 min once the relative

¹The author's laboratory experience has shown that the dry ice alone or a mixture of salt and ice is sufficient to quantitatively trap radon in the activated charcoal column.

Fig. 2.4 Measurement of ^{222}Rn in soil gas using Soil Gas Probe coupled with RAD7 (http://www.durrIDGE.com/products_soil_gas_probe.shtml)



humidity drops below 10 %. A typical 2 h continuous run would amount to ~ 20 L of soil air and the volume of air passed through RAD7 will depend on the porosity and tortuosity of the soil.

2.2.1.3 Water Samples

Collection of grab water samples for ^{222}Rn measurements (or for any other dissolved gases) from rivers, lakes, oceans (including sea water at all depths) and underground is always challenging. A precaution that need to be exercised in the collection of any grab water samples is identical in that dissolved gases from the water should not escape during transfer from the water sample to the storage container. Aeration of the water sample during collection could result in a loss of radon, and thus the sampling procedures need to be designed to minimize this effect. Direct suction of water sample into a 20-L flint glass bottles have been used in the gas extraction procedures before (Broecker 1965). The bottle should be first evacuated and sealed off. A length of rubber tubing is usually attached to the exhaust side of the adaptor. The far end is weighted and lowered into the water to the desired depth in a lake (or river or sea). The exhaust valve is opened and the water is allowed to flow in until the desired amount is obtained. The container is then sealed, moved to the laboratory, and attached directly to the gas extraction system, thus collecting near surface waters with no chance of gas loss.

During GEOSECS (Geochemical Ocean Sections Study, 1968–1978), large volume (~ 20 L) water samples were collected and stored in gas-tight containers

and were analyzed in ships for ^{222}Rn . The protocol adopted during the GEOSECS cruises involved pumping helium through the water sample stored in a gas-tight container to drive all the Rn into an activated charcoal column inside a stainless steel trap that is kept at dry ice (Mathieu et al. 1988). Helium is then re-circulated through the sample trap using a diaphragm pump that comes with the Extraction Board (Mathieu et al. 1988). Water vapor and CO_2 are usually trapped by serially connected Drierite and Ascarite columns, respectively. Radon from the charcoal column is mobilized by heating and subsequently quantitatively transferring it into a Lucas counting cell. After transferring Rn, the cell is stored for ~ 3 h for all the daughter products of ^{222}Rn to reach secular equilibrium (note the long-lived daughter products of ^{222}Rn (before ^{210}Pb) is ^{214}Pb , with a half-life of 26.8 min; in about 2 h, $^{214}\text{Pb}/^{222}\text{Ra}$ activity ratio = 0.955; in about 3 h, the activity ratio becomes 0.99). The cell is then counted in an alpha scintillation counter (Mathieu et al. 1988). After the measurement of ^{222}Rn , the water sample can be stored for about 20 days and the concentration of in situ ^{226}Ra can be determined. For in situ ^{222}Rn measurements, appropriate correction for the decay and in-growth should be applied (Mathieu et al. 1988).

More recently, ^{222}Rn has been utilized widely as a tracer to quantify the submarine groundwater discharge in coastal areas. A multi-detector system that measures ^{222}Rn activities continuously in coastal waters has been developed (Fig. 2.5, Dulaiova et al. 2005). The system utilizes a constant stream of water that is delivered by a submersible pump to an air-water exchange system where radon in the water phase equilibrates with radon in a closed air loop. The air stream is then fed to three radon-in-air monitors connected in parallel to determine the activity of ^{222}Rn . The precision in this methodology is approximately $\pm 5\text{--}15\%$ for typical coastal seawater concentrations (Dulaiova et al. 2005).

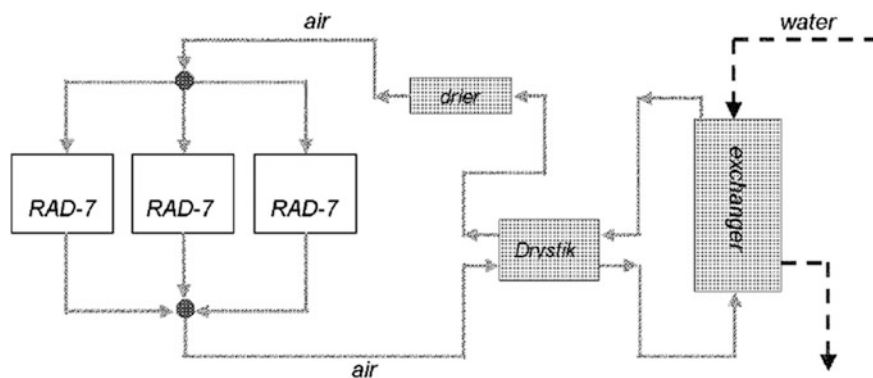


Fig. 2.5 A simplified sketch of an equilibrium three-stage radon measurement system. Lines shown represent the water pumped through the exchanger by a submersible pump (*dashed*) and a closed air loop (*solid*) that flows through three RAD-7 radon analyzers in parallel (Dulaiova et al. 2005)

2.2.1.4 Lucas Cell Method of Measurement

One of the most important devices to measure radon is the scintillation flask which was first introduced in 1950s by Henry F. Lucas and is commonly known as Lucas cell (Lucas 1957). The original cell has the shape of right circular cylinder with a hemispherical cap, with 5 cm diameter and a volume of 100 cm³. Typical efficiency is 70–80 %. In a new cell, the background count rate of ~ 0.1 count/min can be achieved. Commercially, different volumes of cells are available (e.g., 270, 160, 100 cm³). The inside is coated with silver-activated zinc sulfide (ZnS(Ag)) phosphor and one side (usually the bottom side) has transparent flat surface and constitutes a viewing window. The cell typically has two sampling ports, each fitted with a valve, and a photomultiplier tube is coupled to the transparent window of the cell. When an alpha particle produced within the cell strikes the phosphor, a flash of light is produced and is sensed by the photomultiplier tube which is optically coupled to the cell window and associated electronics (preamplifier and multiplexer) and is counted using a multichannel analyzer (MCA) (recorded as a count, not to be confused with the energy spectrum often used in alpha spectroscopy). Most often, the traditional method of gating amplifier/scalar arrangement has been widely used, but the use of multichannel analyzer advantages include: (i) ability to observe the number of counts above and below the ‘window’ of the counting region (similar to upper level of detection (ULD) and lower level of detection (LLD) in alpha-ray or gamma-ray spectroscopy); (ii) the overall shape of the distribution allows easy identification of spurious counts resulting from electrical noise or interference (Kritz et al. 1998); (iii) changes in the performance caused by voltage drifts or deterioration of the PMTs; and (iv) MCAs can be programmed to make a set number of individual counts during the counting period.

As mentioned earlier, after radon is filled in a Lucas cell and before it is assayed in a scintillation counter, it is usually stored for about 3 h for the daughter products ²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi and ²¹⁴Po to reach secular equilibrium with ²²²Rn (governed by the half-life the longest lived daughter, ²¹⁴Pb, with a half-life of 26.8 min). In this decay of ²²²Rn, three alphas (from the decay of ²²²Rn, ²¹⁸Po, ²¹⁴Po) and 2 betas (from the decay of ²¹⁴Pb and ²¹⁴Bi) along with a number of gamma-ray lines are produced (summarized in Porcelli and Baskaran 2011). The background of the Luca cell is expected to increase due to its repeated use from the decay of ²²²Rn daughter products during the time between filling and evacuation of the cell (after counting). In this interval, a finite amount of ²¹⁰Pb would have been produced from the decay of ²¹⁴Bi via ²¹⁴Po. With time, ²¹⁰Pb decay to ²¹⁰Po (alpha decay) via ²¹⁰Bi (beta emitter) takes place and thus periodic monitoring the background of each of the Lucas cell is essential.

2.2.1.5 Comparison of Grab Sample Analysis Versus RAD7-Based Method

While the grab sample analytical method described above for air and water samples is precise (typically $\sim \pm 5$ %) and straight forward, there are two main

disadvantages: (i) radon extraction/transfer method is labor-intensive and this method can only partially be simplified by automation and computer-controlled laboratory processing; and (ii) this grab sample method requires the extraction/transfer and counting facility to be in close proximity to minimize decay and in-growth time in charcoal between sampling and radon extraction. In addition, the throughput is poor. However, more recent methodology with Radon Monitor (RAD7) yields precision ($\sim \pm 10\text{--}15\%$) that is less than the previous quantitative extraction methods $\sim \pm 5\%$ (e.g., Mathieu et al. 1988). The advantages include (i) the ease of the analysis in the field, (ii) no need to apply decay/growth correction, (iii) no in-growth correction for the progeny of ^{222}Rn ; and (iv) a much higher throughput, making this method of analysis distinctly advantageous in the investigation of coastal oceanic processes. Details of this method are given in Sect. 2.2.

2.2.1.6 Calibration, Quality Control, Precision and Accuracy

The extraction/transfer analysis system used for small volume water samples (e.g., groundwater) is calibrated by flushing a known ^{222}Rn content (in equilibrium with ^{226}Ra) of sealed glass containers obtained from the National Institute of Standards or preparing ^{226}Ra standard using RGU-1 (IAEA) primary standard by dissolving a known amount of RGU-1 in which ^{238}U and all its daughter products all the way to ^{210}Po are in secular equilibrium with ^{238}U . Care must be exercised in dissolving the RGU-1 standard (e.g., drying the standard before weighing an aliquot for analysis to ensure the standard is free of water vapor, complete dissolution of the standard with ^{226}Ra -free mineral acids that include HF, HNO_3 and HCl, no loss of ^{226}Ra during digestion and handling/transfer of the solution, etc.). In the case of large-volume seawater (or surface water), 20 L of filtered seawater, with a known in situ ^{226}Ra concentration, is taken in a glass container (to prevent the loss of Rn through diffusion), spiked with a primary ^{226}Ra standard (or RGU-1 standard in solution form) and stored for >1 month. The sample is assayed for ^{222}Rn and the result obtained by this method is compared to the certified value.

Calibration of atmospheric air sampling device is more difficult. An atmospheric air sampling site at which radon concentration is simultaneously determined both with a calibrated device and an uncalibrated instrument. Intercalibration between laboratories that make routine airborne measurements would also be quite useful.

As mentioned earlier, for every alpha particle produced from the decay of ^{222}Rn , there are two other daughters that produce alpha particles: ^{218}Po , with a half-life of 3.10 min and ^{214}Po , with an half-life of 164 μs (effective half-life ($\tau_{\text{eff}} = \tau_{\text{Po-218}} + \tau_{\text{Pb-214}} + \tau_{\text{Bi-214}}$) of 71.6 min). Since the normal procedure is to start counting the sample after two to three hours, it is assumed that ^{222}Rn is in secular equilibrium with ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po . The overall error (propagated error) in the determination of ^{222}Rn by this method arises from the errors associated with the following: (i) net counts (=total counts – background counts) for

a given counting cell; If N_t is the total counts n_t is error associated with the total count, and B_t is the total background count and b_t is error associated with the background count, then, the net count (N) and error associated with the net count (n) is given by:

$$N \pm n = (N_t - B_t) \pm (n_t^2 + b_t^2)^{1/2}$$

(ii) the efficiency (η_c) of the Lucas cell. The total variations in the amount of silver-activated zinc sulfide (ZnS(Ag)) coating can result in slight variations in the counting efficiency of the cell which could also change with time; (iii) counting efficiency of the scintillation counter (η_s) which is determined using a glass-sealed Lucas Cell with a known amount of ^{226}Ra in which ^{226}Ra and ^{222}Rn are in secular equilibrium; and (iv) volume of air used for study. We assume the transfer efficiency of ^{222}Rn from the sample container to the Lucas cell is 100 %. For a typical 1000 counts, the 1 standard deviation (1σ) on the net counts is ~ 3.2 % (assuming the counting error associated with the cell background is very small compared to the counting error associated with the gross count). Assuming that the 1σ error associated with the counting efficiency of the Lucas cell is 5 %, the 1σ error associated with the efficiency of the counting instrument is 2 %, and the 1σ error associated with the volume of air sample is 1.5 %, then, the propagated 1σ uncertainty of the overall measurement may be estimated by taking the square root of the sum of the squares of the individual elements entering into the calculation as follows: $[(3.2 \%)^2 + (5 \%)^2 + (2 \%)^2 + (1.5 \%)^2]^{1/2} = 6.4$ %. Note that in counting instruments, the most common practice is to report 1σ of the propagated error while in mass spectrometry, the standard practice is to report 2σ values. Williams et al. (2011) reported the following errors: variability associated with the counting efficiency of the cell is $\sim \pm 5$ %, accuracy of the radon calibrated source of ± 4 %; and the overall typical error with the measurement of ± 8 %. Presence of a small but variable amounts of ^{226}Ra in charcoal contributing to ^{222}Rn has been reported by Williams et al. (2011) and hence blank levels of the extraction/transfer system needs to be periodically monitored.

The activity of the ^{222}Rn in air sample (A , in disintegration per second (dps) per standard cubic meter, Bq m^{-3}) at the time of collection can be calculated as follows:

$$A(\text{Bq m}^{-3}) = N e^{\lambda t} / (3 t \eta_c \eta_s V * 60)$$

where t is the time elapsed between sampling and mid-counting (in minutes), λ is the ^{222}Rn decay constant ($1.2598 \times 10^{-4} \text{ min}^{-1}$), V is the volume of air samples (per standard m^3), 60 is to convert dpm to dps (or Bq) and the factor '3' is to account for the additional two alpha emissions that occur following each radon decay at secular equilibrium in the counting cell.

The air–water exchanger (Fig. 2.6) is simply a plastic cylinder that has water entering continuously via a nozzle that aspirates the water into fine droplets so that radon is emanated into a stream of air that is circulated through the exchanger, a drying system, then to the RAD7 for measurement, and then returned to the exchanger to begin another loop. After some time, the radon activity in the air reaches equilibrium with the radon in the water, the ratio at equilibrium being determined by the water temperature and salinity.

In order to increase throughput, several RAD7s can be arranged in parallel with one exchanger (Fig. 2.5). Using 3 RAD7s (Fig. 2.5), it has been shown that typical coastal water radon measurements can be made in 10-min intervals with a precision ranging from 5–15 % depending upon the radon activities present. Thus, a typical day of surveying in a coastal region can result in 50 or more measurements.

2.2.3 *Integration Radon Monitors*

The Integrated radon monitor method of radon measurement provides time integrated ^{222}Rn concentration for a period of sampling, typically few days to few months. The two requirements for this method to effectively work are: (i) the sampling technique must maintain an integrated record of each alpha particle impacting the measurement device; and (ii) the detector must maintain exposure information from the time of retrieval to the time of analysis in a laboratory.

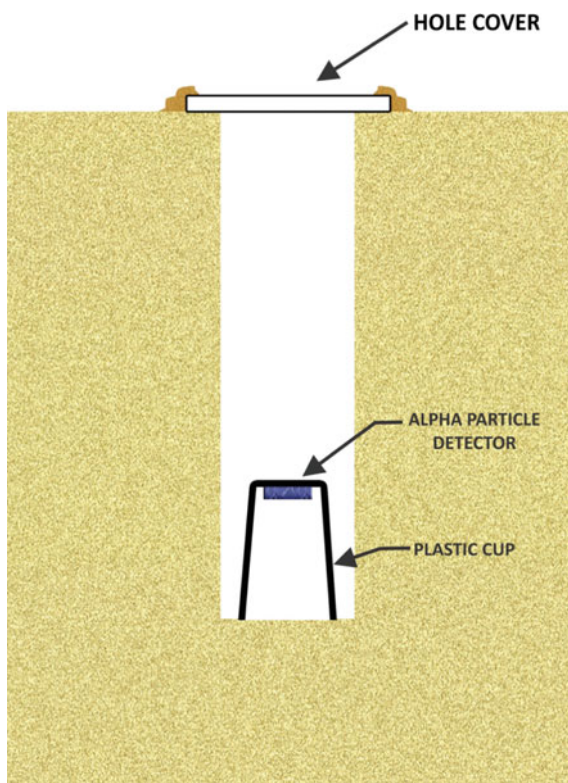
The solid-state nuclear track detectors (SSNTD) were recognized to hold the powerful potential of recording the primary radioactive signature of radon—alpha particle emission, and enabled the development of simple and quantitative methods of measuring the radon isotopes. Original discovery on the alpha particle tracks were seen in cellulosic materials such as cellulose nitrate and cellulose acetate butyrate. Subsequently, alpha particle tracks were also seen in polycarbonates such as bisphenol-A polycarbonate, i.e., Lexan and allyl diglycol carbonate, CR-39 (summarized in Fleischer 1988). Due to good ionization sensitivity and stability against various environmental factors, the CR-39 (polycarbonate material) has been used as the state-of-the-art track detector for environmental radon. More recently, LR-115 track detector-based twin-cup dosimeter has been developed and are used for indoor Rn-222 and Rn-220 measurements (Eappen and Mayya 2004).

Using the solid-state detector technique, radon concentrations have been measured in air, water and other earth material, with other applications spanning from biological dosimetry and home-ventilation design to prospecting for hydrocarbon and uranium resources, and to measurements of the respiration of the Earth, which can provide warnings of impending geological hazards such as earthquakes and volcanic eruption (Fleischer 1988). The technique is simple to use and relatively inexpensive. For the measurements of environmental radon levels, exposure of few weeks to a year are normally necessary to obtain desirable precision at relatively low analytical cost. Following the exposure, the film is etched in an alkaline solution (typically NaOH or KOH) to enhance the size of the tracks so that the track

density (number of tracks per unit area) can be easily determined by optical microscopy or by automated scanning and counting technology. Although this method is less reliable for rapidly obtaining information on indoor ^{222}Rn concentrations, it is well-suited for obtaining annual average concentrations.

A typical ^{222}Rn -only measuring device comprised of an enclosure that allows ^{222}Rn to enter through a permeable, anti- ^{220}Rn membrane is shown (Fig. 2.7). This enclosure excludes condensed alpha emitters and the membrane area and thickness are chosen so as to exclude the unwanted gaseous alpha emitters including ^{219}Rn (it is derived from $<1\%$ of uranium (^{235}U) and its half-life is 3.96 s) and ^{220}Rn , without significantly affecting the ^{222}Rn . Thus, only ^{222}Rn enters the enclosure and the three alpha particles emitted (^{222}Rn , ^{218}Po and ^{214}Po (note that the rate of production of ^{214}Po is determined by half-lives of ^{214}Pb and ^{214}Bi ; details in Chap. 1)). Some of the radon measurements made with the Terradex Track Etch system were designed to give integrated readings that minimize the effects of diurnal and barometric pressure. For sampling in soils, detector cups were buried at a depth of 40 cm (Fig. 2.7). The alpha tracks can be counted and it yields a highly reproducible unambiguous measure of ^{222}Rn (Fleischer et al. 1980).

Fig. 2.7 Alpha particle detector mounted on a plastic cup and this instrument is usually kept at 40 cm below ground



The unique advantages of solid-track nuclear track detectors include: (i) the energy of the alpha particles produced from the decay of ^{222}Rn and its progeny (alpha particles from the decay of ^{222}Rn , ^{218}Po and ^{214}Po) are sufficiently ionizing to produce etchable damage tracks in a variety of polymeric solids; and (ii) other lightly ionizing radiation such as β - and γ -rays from other natural radioactivity and from cosmic-rays are not recorded.

2.2.4 Continuous Monitors

The passive integrating-type detectors are frequently deployed for indoor radon surveys. The on-line radon monitors are commonly used for special radon diagnostic measurements in residential environments, in particular build-up in confined spaces, as well as in quality assurance programs. Real-time data not only help in deciding control measures to minimize radon exposure in the environment, but also are useful along active fault zones where radon concentrations may change significantly and rapidly.

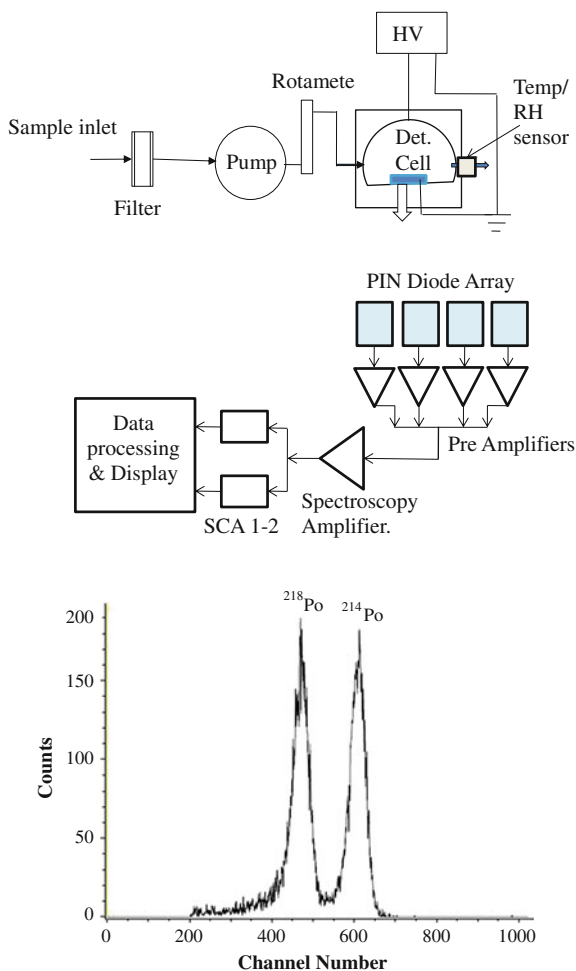
A large amount of efforts have gone into the development of fast responding, highly sensitive radon monitors. The most common ones available in the market are: (i) RAD7 (DurrIDGE, Boston, USA (<http://durrIDGE.com>); (ii) Radon Scout Plus (SARAD, GmbH, Dresden, Germany, (<http://www.sarad.de>)); (iii) RTM 2200 (SARAD, Dresden, Germany, (<http://www.sarad.de>) and (iv) CRM (BARC, Mumbai, India; Ashokkumar et al. 2014). Conventional detectors used in the development of radon monitors include pulse ionization chambers, scintillation detectors coupled with photomultiplier tubes and silicon PIN diodes. Of these, the silicon PIN diode detectors are found to be better in terms of low bias (<100 V), intrinsic alpha detection efficiency (approaching near 100 % for alpha particles), energy resolution, its simplicity, robustness and its cost (Ashokkumar et al. 2014). A comparison of the sensitivity of these four commonly used silicon-detector-based radon monitors are shown in Table 2.1. The CRM developed at the Bhabha Atomic Research Center appears to have higher sensitivity (=counts per hour, CPH/radon

Table 2.1 Comparison of sensitivity of various silicon-based radon monitors (from Ashokkumar et al. 2014)

Instrument	Radiation detector	Mode of operation	Sensitivity (CPH/Bq m ⁻³)
RAD7, DurrIDGE ¹	Si-PIPS	Sniff	0.32
		Normal	0.65
Radon Scout, SARAD ²	Si detector	Diffusion	0.12
RTM2200, SARAD ²	Si detector	²¹⁴ Po-excluded	0.19
		²¹⁴ Po-included	0.42
CRM, BARC ³	Si PIN	Fast	0.91
	Diode array	Sensitive	1.76

¹www.durrIDGE.com; ²www.SARAD.de; ³Ashokkumar et al. (2014)

Fig. 2.8 *Top*: Diagram of the continuous radon monitor developed at the Bhabha Atomic Research Center; *Bottom*: Typical alpha spectrum acquired during calibration (courtesy of Ashokkumar, 2016; details given in Ashokkumar et al. 2014)



concentration, Bq m^{-3} , CPH/Bq m^{-3} ; Ashokkumar et al. 2014). The CRM instrument (Fig. 2.8 top) seems to be less dependent on the humidity of the air compared to other existing systems. It has been shown earlier that the electro-deposition efficiency of radon progeny depends strongly on the relative humidity (RH) of the sampled air. The minimum detection level, C_{MDL} [$=2 \times$ standard deviation of background counts/(sensitivity factor, CPH/Bq m^{-3}) \times counting time] of 0.80 Bq m^{-3} for 1 h counting appears to be significantly lower than other instruments (in the fast mode with a 15 min cycle time, CMDL is 3.1 Bq m^{-3} (Ashokkumar et al. 2014)). Furthermore, the volume of the hemispherical metal collection chamber (1 L) is substantially larger than the other systems (0.150–0.750 L).

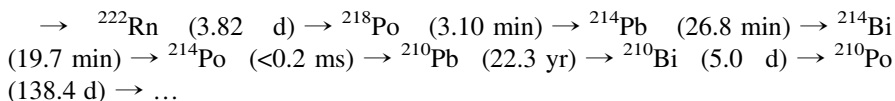
Scintillation cell-based continuous radon monitors (Smart Radon Duo and Scintillation Radon Monitors) have been developed by the Bhabha Atomic Research Center in Mumbai (Bombay, India). These are based on a software that takes into

account the ingrowth of progeny in a given counting interval without making equilibrium assumptions. An advantage of this technique is that its performance is not affected by relative humidity and hence does not require silica-gel or any other dehumidifying agents. The performance of this instrument has been satisfactorily compared against Alphaguard and RAD7 and has been extensively used for radon and thoron measurements in India (Kumar et al. 2013; Chauhan et al. 2014).

In addition to these air monitors discussed above, continuous radon concentrations in groundwaters are monitored by means of an electronic automatic probe, known as Clipperton (CNRD-URA 1767, Universite Montpellier II, F-34094, Montpellier, France). It is based on the detection of alpha-particles using 1 cm² area solid-state electronic sensor. The Clipperton probe allows the measurement of radon activity levels in a range between 100 and 1,000 kBq m⁻³, with an accuracy of $\sim \pm 7.5\%$ (Monnin and Seidel 2002). In addition to the Rn sensor, probes for other geochemical, geophysical and hydrological parameters (such as CO₂, CH₄, N₂, Eh, pH, DO, electrical conductivity, temperature, water level fluctuations, microseismicity) can also be simultaneously deployed which are useful for earthquake research (Streil et al. 1997).

2.2.5 Analysis of ²²²Rn Using Its Progeny

The decay of ²²²Rn results in the formation of a series of short-lived daughters before ²¹⁰Pb:



While radon is a noble gas, all of its daughter products are metals (although Po is sometimes classified as metalloid because of its position in the periodic table). As mentioned earlier, the longest-lived daughter product between ²²²Rn and ²¹⁰Pb in the decay chain (Chap. 1) is ²¹⁴Pb, and hence in about 3 h, we expect the activities of: ²²²Rn = ²¹⁸Po = ²¹⁴Pb = ²¹⁴Bi = ²¹⁴Po. Polonium, Pb and Bi are highly particle-reactive and hence upon formation quickly and irreversibly attach to submicron-sized aerosol particles of the ambient air. In an isolated air mass when there are no major preferential removal of Pb-Po-Bi-laden aerosol particles from the air, ²²²Rn concentrations can be determined from the measurements of any of the daughter products.

The alpha particles from the decay of ²¹⁸Po (6.002 MeV), ²¹⁴Po (7.687 MeV) or ²²²Rn (5.489 MeV), or the beta particles from the decay of ²¹⁴Pb ($\beta_{\text{max}} = 1.023$ MeV) and ²¹⁴Bi ($\beta_{\text{max}} = 3.272$ MeV) using Geiger-Muller counter or the gamma-emissions associated with the decay of ²¹⁴Bi (0.609 MeV ($\sim 100\%$), 1.120 MeV (1.120 MeV (15.0 %), 1.764 MeV, 15.9 %)), and ²¹⁴Pb (0.3159 MeV (35.9 %), 0.2952 MeV (18.5 %); (0.2420 MeV (7.5 %))), using gamma spectrometry (non-destructive) can be utilized to assess the activities of ²²²Rn.

2.2.5.1 Determination of ^{222}Rn Using Its Progeny in a Solid Surface-Barrier Detector

The alpha-emitting progeny of ^{222}Rn can be quantitatively determined using alpha spectrometry. A known volume of air is drawn through a cylindrical system through two in-line filters. The first filter retains the particulate matter and radon progeny so that only radon passes through the first filter. As the filtered air from the first filter moves to the 2nd filter, ingrowth of radon progeny occurs. This radon progeny is collected on a second filter which faces the surface barrier detector. An alpha-spectrum of ^{214}Po and ^{218}Po are shown in Fig. 2.8 bottom.

Another simple method involves collecting the +ively charged decay products onto a metal surface maintained at a negative potential placed inside a metallic chamber. The positively charged ^{218}Po collected on the metal surface is then counted using surface-barrier detector.

Advantages of a surface-barrier detector includes very low background (<0.1 count per hour in the regions of interest), high energy resolution and ruggedness, ability to spectrally separate the signals of progeny of ^{222}Rn and ^{220}Rn (they have different alpha energies). However, the alpha spectrometer requires electrical power and associated equipment, vacuum pump and the cost of the instrument is relatively high.

2.2.5.2 Determination of ^{222}Rn Using Electret

Electrets are passive, lightweight, integrating and relatively inexpensive detectors (Kotrappa et al. 1992). It is electrically charged Teflon disc, serving both as a source of an electric field and sensor. When an alpha particle decays in the detector chamber, ionization of the air takes place leading to a decrease in the total charge on the electret. The decrease in the total charge results in the voltage drop (charge, Q = capacitance, C^* potential difference, V) over the measurement period and is used to quantify the ^{222}Rn concentration.

2.2.5.3 Determination of ^{222}Rn Using Beta Counter

Radon concentrations can be determined by measuring the beta activity of ^{214}Pb and ^{214}Bi collected on filter papers, assuming secular equilibrium between ^{222}Rn and its progeny in the air. The beta activity can be assayed with plastic scintillators mounted on photomultiplier tubes or the filter paper can be directly counted in a beta counter (a Geiger-Muller counter), with appropriate use of absorber film (such as Mylar, with suitable mass density). The assumption of secular equilibrium between ^{222}Rn and its daughter products has been observed in the atmosphere over the ocean even after rain and during fog, although uncertainties of up to 15 % have been reported (i.e., $^{214}\text{Pb}/^{222}\text{Rn} = ^{214}\text{Bi}/^{222}\text{Rn} = 1.00 \pm 0.15$ (Larson 1978). However, due to short-term atmospheric turbulence and other short-term events, it

may be necessary to check the secular equilibrium between radon and its progeny periodically due to differences in the geochemical properties of radon and its progeny.

2.2.5.4 Radon Measurements Using Gamma-Ray Spectrometry

Atmospheric ^{222}Rn concentrations can be measured by measuring ^{214}Pb and ^{214}Bi by gamma spectrometry (e.g., Duenas et al. 1994). 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 .

2.2.5.5 Radon Measurements Based on Direct Progeny Monitoring Technique

Direct radon (and thoron) progeny sensors have been developed (Mishra et al. 2009a, b; Mayya et al. 2012) to measure directly time integrated radon progeny concentrations indoor. The direct radon progeny sensor (DRPS) and direct thoron progeny sensor (DTPS) are absorbers mounted on solid-state nuclear track detector on which alpha particles emitted from the progeny atoms deposit on the surface during deployment in the field. The absorber offers the right energy window to detect either ^{214}Po or ^{212}Po alpha emission and virtually cuts-off alpha contributions from other air-based emissions. DRPS has a $37\text{ }\mu\text{m}$ absorber to detect mainly the alpha particles emitted from ^{214}Po (7.69 MeV) formed from the eventual decay of ^{218}Po , ^{214}Pb and ^{214}Bi atoms deposited on it (Mishra et al. 2009a, b).

2.2.6 Indoor Radon Measurements

There are three inexpensive and widely used methods to measure indoor ^{222}Rn concentrations (charcoal canisters, electrets, and alpha-track detectors), all of these methods require long-term (48 h to 3 months) exposure durations. Following the exposure of activated charcoal, the quantity of radon adsorbed can be analyzed, either by gamma-ray spectrometry (via ^{214}Pb and ^{214}Bi) or desorption followed by scintillation counting. Due to the short half-life of ^{222}Rn , 3.82 days, the exposure period is useful for only about a week following exposure and the sampler has to be returned to the laboratory for prompt analysis. This method could complement the etched-track detector in survey work. However, none of these methods seem suitable for measurement of radon concentrations in soil gas below buildings because the measurement devices are relatively large and cannot easily be placed below a building foundation for the required sampling period. In addition, the high

moisture levels in soil gas interfere with the sorption of radon onto charcoal canisters, and the gaseous diffusion constant for radon in the vadose zone is unknown. As a result, grab samples are the most effective means to measure radon concentration in soil gas. Track etch detectors placed inside two cups one of which cuts-off thoron completely is most popularly used for indoor radon measurements (Eappen and Mayya 2004).

2.2.7 Identification of Entry Points of Radon Inside Buildings Using RAD7

The soil gas released from Earth surface contains ^{222}Rn and ^{220}Rn and they start to decay. Thoron has half-life 55.6 s, and so soil gas that has been out of the soil for 5 min will have lost $\sim 97\%$ of thoron. Assuming that the soil gas moves about 20 cm/min, by the time it has moved about 1 m, most of the thoron ($>97\%$) has decayed. It is reported that at one half meter away from the hotspot thoron, it is barely detectable. If the air is sampled near an entry point in the building and thoron is detected, then, the Rn entry hot spot is <1 m from the point of sampling. The first daughter of thoron is ^{216}Po (half-life = 150 ms) and so we need to have the instrument that can measure instantaneously and RAD7 that is currently available in the market has been demonstrated to be useful for this purpose. A fast monitoring of thoron level is to set RAD7 by listening to the beeps, with the tone set to Geiger mode, and monitoring the status display which will show if the beep is in the B (thoron) window. The sample location corresponding to the highest count rate of thoron is the entry point of radon.

2.2.8 Summary and Future Direction

A large number of methods have been developed for ^{222}Rn (and a few methods for thoron) for a number of sample matrices and for different applications. The precision/accuracy of measurements depends on the applications. Indoor radiation dose assessment requires generally lower precision compared to grab samples from a marine environment where it is used as a tracer for gas-exchange studies. The spatial and temporal variability of radon in subsurface media (soil air and groundwater) limits the desired precision. However, in oceanic and limnological studies where radon is used as a tracer, higher precision is desired. The throughput for such a study is low and enhancing throughput with higher precision is needed. Overall, there are sufficient methodologies and instruments exist in recent literature which overall meets the research needs.

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