Upgrade of a Highly Sensitive Monitor for Atmospheric Radon Measurement

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Abstract: Atmospheric radon is an ideal tracer that is widely used in atmospheric science. To meet the need for a continuous online measurement of atmospheric radon concentration, an upgraded radon monitor based on an electrostatic collection method was developed following Iida’s measurement system. Two major improvements have been realized. First, an 18 mm × 18 mm Si-PIN detector and a multi-channel analysis system were used to distinguish different alpha particles. Second, the P2O5 desiccant was replaced by a new membrane drying system, and the influence of humidity was corrected by a humidity correction coefficient. Calibration and comparison experiments were carried out in detail, and a one-year continuous measurement was also performed. Results showed that the measurement sensitivity was evaluated to be 24.3 cph/(Bq m⁻³), and the lower level detection limit was 0.2 Bq m⁻³ for a 1-h cycle at the absolute humidity of 0.34 g m⁻³. The annual average radon concentration of Beijing was 11.1 ± 4.0 Bq m⁻³, which changed from 2.8 Bq m⁻³ to 30.3 Bq m⁻³ between 15 October 2018 and 1 October 2019. The upgraded monitor’s high data acquisition rate and good performance indicate that it is suitable for long-term observation on atmospheric radon.

Keywords: atmospheric radon; activity concentration; continuous measurement; electrostatic collection; Si-PIN detector

1. Introduction

Radon (²²²Rn) is a naturally occurring gaseous radioactive decay product of the radium isotope ²²⁶Ra, which is present in all terrestrial materials. As radon is not only a naturally occurring radioactive element with a half-life of 3.824 days but also a noble gas, it is relatively chemically stable and, consequently, can be sensitively detected. These characteristics enable radon to be widely used as an ideal tracer in atmospheric studies, such as those on continental air mass diffusion, atmospheric transport model processes, turbulence and the stability of the lower atmosphere [1–9]. Considering the rather low-level concentration and the variation of atmospheric radon, continuous measurement with high sensitivity is essential for radon-related atmospheric studies.

A project entitled “Measurement of Radon as a Tracer of Air Pollutants in East Asia” was launched from 1985 until 2010, led by Nagoya University, Japan. During the project, measurements of the atmospheric radon concentrations at various locations in East Asia, including Japan, South Korea and China, have been carried out [10–12]. As a member of the project, our laboratory put an electrostatic radon monitor (ERM) into operation in 2002, and continuous measurements on atmospheric radon concentration were carried out in Beijing for more than 5 years [13,14], for 2 years in Qingdao [15] and 1 year in Hangzhou, China. This electrostatic radon monitor ERM, which was developed by T. Iida [16], consisted of a hemisphere electrostatic collection chamber with a volume of 16.8 L, an electrode ZnS(Ag) scintillator for alpha particle detection and a P2O5 desiccant for air drying. After many years of application, we found that the background had been slowly increasing due to the
210Po accumulation on the surface of the ZnS(Ag) scintillator, which, consequently, caused the increase in the lower level detection limit of the monitor. The frequent maintenance of the P2O5 air dryer was also quite a burden, especially in some remote areas or areas with high air humidity, such as coastal cities.

To meet the need for a continuous online measurement of atmospheric radon activity concentration with stable high-sensitivity and easy-performing maintenance, an upgraded radon monitor was newly developed. Calibration and comparison experiments as well as field measurements were carried out in this paper.

2. Materials and Methods

Considering the shortcomings of ERM, two major improvements were carried out. First, the ZnS(Ag) scintillator detection system was changed into a new measurement system with a Si-PIN photodiode detector and a multi-channel analysis system. Second, the P2O5 desiccant was replaced by a new air dryer module, and the influence of environmental humidity was corrected to meet the need for a long-term and stable operation.

2.1. Measurement Module

Compared with the ZnS(Ag) scintillation detector, the Si-PIN photodiode has been increasingly used for low-level radon concentration measurement in recent years [17–22] due to its higher resolution spectrum of alpha particles. With the Si-PIN detector, alpha particles from 218Po, 216Po and 210Po can be discriminated from each other easily, and the background for the regions of interest (ROIs) can be kept stable at a relatively low level without the influence of 210Po accumulation.

The hemispheric electrostatic collection chamber with a volume of 16.8 L was kept due to its electric field uniformity and high sensitivity, but a more compact structure was designed for the upgraded monitor, as shown in Figure 1.

![Figure 1](image_url)

**Figure 1.** Schematic diagram and photo of the upgraded radon monitor.

In the upgraded measurement module, an 18 mm × 18 mm Si-PIN photodiode (S3204-09, HAMAMATSU PHOTONICS K.K., Hamamatsu City, Japan) was used, which was installed at the bottom center of the hemispheric chamber. Radon in the air is pumped into the electrostatic collection chamber, and inside the chamber, 222Rn decays into 218Po ions. Since most new-generated 218Po particles are positively charged [23], 218Po ions are then electrostatically collected on a negative electrode, the surface of a Si-Pin photodiode. Alpha particles emitted from 218Po and its progeny 214Po were detected by the photodiode detector. A spectroscopy amplifier, a multichannel analyzer (MCA) with 256 channels, and a microcontroller unit (MCU) were integrated into the electronic unit to record and
analyze the alpha spectrometry. Measurement data were recorded and uploaded to the data center automatically through a wired transmitter as well as a GPRS (General Packet Radio Service) unit.

A typical alpha spectrum of the upgraded measurement module is shown in Figure 2, and the peaks corresponding to $^{218}$Po (6.0 MeV) and $^{214}$Po (7.69 MeV) are clearly seen and could be analyzed by the MCU.

![Typical alpha spectrum of the upgraded measurement module.](image)

**Figure 2.** Typical alpha spectrum of the upgraded measurement module.

Assuming that during a measurement cycle, (1) radon concentration is constant, (2) the electrostatic collection efficiency is constant, and (3) the detection efficiency is constant, then the radon concentration and its uncertainty can be calculated as follows [24]:

$$C_{Rn} = CF(AH_0) \times \left( \frac{N_1 + N_2 - 0.56N_3}{T} - B \right) \times \eta(AH)$$

$$\sigma_{C_{Rn}} = \sqrt{\left( \frac{\sigma_{CF}^2}{CF} + \left( \frac{\sigma_{N_1}}{\eta} \right)^2 \right) \times C_{Rn}^2 + \left( \frac{\sigma_{N_1}^2 + \sigma_{N_2}^2 + 0.56 \sigma_{N_3}^2}{T^2} \right) \times \eta^2 \times CF^2}$$

where $C_{Rn}$ is radon activity concentration (Bq·m$^{-3}$); $N_1$ is the total counts in the ROIs of $^{218}$Po (6.0 MeV); and $N_2$ and $N_3$ are the counts in the ROIs of $^{214}$Po (7.69 MeV) and $^{212}$Po (8.78 MeV), respectively. The influence of $^{212}$Bi to $^{212}$Po is eliminated by the branch ratio 0.56. $B$ is the background in the ROIs of $^{218}$Po and $^{214}$Po (cph). $T$ is the measurement cycle (hour). $CF(AH_0)$ is the calibration factor (Bq·m$^{-3}$/cph) at the reference absolute humidity $AH_0$. $\eta(AH)$ is the humidity correction coefficient (non-dimensional), which is defined as the ratio of sensitivity at absolute humidity (AH) to that of at the reference absolute humidity ($AH_0$).

The lower level detection limit (LLD) of radon concentration for the confidence level of 95% could be calculated as follows [24,25]:

$$LLD_{C_{Rn}} \approx \frac{CF(AH_0) \times (2.72 + 4.65 \times \sqrt{M_B})}{T}$$

where $M_B$ is the total counts of background in the ROIs of $^{218}$Po and $^{214}$Po.

### 2.2. Air Dryer Module

Because the water vapor concentration of air inside the collection chamber greatly influences the measurement sensitivity and system stability of the electrostatic collection method [26,27], various designs of an air dryer system have been proposed to control or eliminate the influence of air humidity. For ERM, the dryer of $P_2O_5$ is adopted as a vapor trap to eliminate the influence. For UI’s radon monitor, an electric cooling dryer is adopted, and the influence of humidity is measured to control the influence [17]. For
Wada’s radon measuring system, a drying system that consists of an electric cooling unit and a commercial Nafion membrane dryer is used to control humidity [18].

In the upgraded monitor, a new air dryer module is proposed, considering the simplicity, drying efficiency and stable operation in a different environment, which is shown in Figure 1.

Without using a commercial Nafion membrane tube, a self-designed 25 cm × 25 cm plate dryer with Nafion membrane (Nafion N115, DuPont de Nemours Inc., Wilmington, DE, USA) was used to obtain a higher moisture removal effect. Air was sampled through a membrane filter (0.8 µm) into the bottom air channel at a flow rate of 2.5 L·min⁻¹, and water vapor in the gases could be removed when passing through the Nafion membrane system into the upper air channel by the pressure difference of water vapor between the up and down air channels, with pressure difference at nearly 0.4 bar. Dried air enters the collection chamber, where radon is measured. Subsequently, the radon exits through the upper air channel and carries out the water vapor simultaneously. The air dryer module is simple and stable and possesses high-drying efficiency, with only one pump driving the entire air system.

Using this specially designed air dryer module, the humidity in the chamber could be limited to a small fluctuation range, and then the humidity response correction could be carried out in a narrow range, which would lead to higher stability and accuracy for the measurement system.

Furthermore, due to the air dryer module, most ²²⁰Rn could not enter the chamber. Only limited ²²⁰Rn, which entered the electrostatic chamber, could easily be discriminated from ²²²Rn, so the influence of ²²⁰Rn on ²²²Rn measurement could be ignored.

3. Results and Discussion

In order to confirm the performance of the upgraded radon monitor, detailed experiments were performed focusing on electrostatic-collecting high voltage, the effect of the air dryer module, and the dependence of sensitivity on humidity. The lower level detection limit (LLD) was also evaluated. A comparison experiment and a long-term continuous measurement in the field were also carried out.

3.1. Influence of Electrostatic-Collecting High Voltage

To determine the optimal electrostatic-collecting high voltage, an experiment was performed under a stable radon concentration (~2500 Bq·m⁻³) and a stable temperature and humidity (~23 °C, 40%RH) in a radon chamber in Peking University. The voltage was set and applied in order of 500 V, 1000 V, 1500 V, 2000 V, 2500 V, 2750 V, 3000 V, 3500 V, 4000 V and 5000 V. The measurement cycle was set to be 10 min. Only stable counts of ²¹⁸Po were used for comparison.

Figure 3 displays the result showing the relationship between the net counts of ²¹⁸Po and high voltage. The result suggests that the counts of ²¹⁸Po grow with the increase in high voltage and are nearly stable after 3000 V, which means that for the collection chamber of 16.8 L, 3000 V is near the saturated voltage.

Considering the distribution of electric field intensity, and the stability and reliability for the long-term operation of the electric field module, a higher voltage of 4600 V was applied in practice, which is slightly different to lida’s measurement system [16].
3.2. Effect of the Air Dryer Module

One major improvement of the upgraded radon monitor is using a simpler and more effective air dryer module to reduce air humidity, making it vary only within a limited range. To test the effect of the air dryer module, a field test was carried out in an extreme environment by the seaside for one month in Taizhou city, Zhejiang province (121.60° E, 29.08° N). The environmental temperature and relative humidity were recorded using a multi-parameter weather sensor (WXT530, Vaisala Oyj, Vantaa, Finland), and the humidity in the electrostatic chamber was recorded using an inner sensor (SHT31, Sensirion Co., Staefa, Switzerland). One month’s comparative results of humidity before and after the air dryer module are shown in Figure 4.

![Figure 4](image-url)

Figure 4. One month’s comparison result of humidity in environment and in the chamber.

It can be observed that the temperature and relative humidity of the location environment vary are in the ranges of 22.8 °C–37.0 °C and 55.0%RH–100%RH, respectively. Inside the collection chamber, the relative humidity of sampling air is greatly reduced, which is varied in a limited range from 6.6%RH to 15.9%RH. More than 80% water vapor
is removed by the dried air, and the dry effect is quite efficient, as expected in earlier calculations [28,29].

3.3. Dependence of Sensitivity on Humidity and the Low-Level Detection Limit

Although an efficient dry effect was achieved through the newly designed dryer module, the humidity inside the collection chamber still changes with time in real environments. As such, the dependence of measurement sensitivity on humidity was studied through experiment calibration, and the humidity influence was further corrected by adopting a humidity correction coefficient.

The upgraded radon monitor was calibrated in an adjustable temperature and humidity control box with a volume of nearly 1 m³ (GDS-010B, HuanWei Co., Shenzhen, China), where the soil radon pumped from a 2 m depth at a flow rate of 4 L·min⁻¹ was used as a radon source. The relative humidity could be adjusted from 10%RH to 95%RH with an uncertainty of ±2%RH, and the temperature range from −10 °C to 50 °C with uncertainty of ±0.3 °C. The radon concentration in the box was measured using a calibrated AlphaGUARD PQ2000 monitor (Saphymo, Montigny-le-Bretonneux, France), which can be traced back to the National Radon Standard of Metrological Institute of China.

Measurement sensitivity, which is defined as the ratio of the count rate of ROIs (²¹⁸Po and ²¹⁴Po) to the radon activity concentration, was calibrated at seven different levels of temperature and humidity, namely, 10 °C, 15%RH; 13 °C, 25%RH; 17 °C, 30%RH; 22 °C, 40%RH; 27 °C, 55%RH; 30 °C, 75%RH and 33 °C, 95%RH.

The relationship between measurement sensitivity and absolute humidity in the electrostatic collection chamber is shown in Figure 5.

![Figure 5. Calibration results of the relationship between sensitivity and absolute humidity.](image)

It can be observed that the measurement sensitivity declined from 24.3 cph/(Bq·m⁻³) to 11.2 cph/(Bq·m⁻³) with the increase in absolute humidity in the chamber from 0.34 g·m⁻³ to 9.47 g·m⁻³. It exhibited an obvious negative exponent relationship. The fitting results of the measurement data are also shown in Figure 5 with a correlation coefficient R² of 0.989, following a previous equation [24].

We chose 0.34 g·m⁻³ as the reference absolute humidity A_H₀ such that the CF(A_H₀) is 0.041 Bq·m⁻³/cph and the humidity correction coefficient η(AH) could easily be calculated from a fitting equation. The background count rate M_B was measured using a put monitor in a highly purified nitrogen gas environment for more than 72 h, which gave the results of 0.3 ± 0.2 cph. Then, the LLD at the 95% confidence level was evaluated to be 0.2 Bq·m⁻³ for a 60 min cycle using Equation (3).
3.4. Comparison with Reference Device in Radon Chamber

After humidity response correction, a comparison experiment was carried out in the radon chamber, where the radon concentration could be adjusted from nearly 100 Bq·m$^{-3}$ to 1000 Bq·m$^{-3}$, and the relative humidity could be adjusted from 30%RH to 80%RH with the temperature stable at 22 °C. The upgraded radon monitor was compared with an AlphaGUARD PQ2000 monitor. Comparison results with the relative humidity are shown in Figure 6.

![Figure 6](image-url)

**Figure 6.** Results of comparison between the upgraded radon monitor and AlphaGUARD at different levels of radon concentrations and relative humidity.

The results show that, under different radon concentrations and relative humidity, the two radon monitors agree with an average relative deviation of less than ±3.7%, although two monitors have different response times when the radon concentration changes greatly. These results indicate that the humidity response correction works well and could eliminate the humidity influence.

3.5. Long Term Continuous Measurement

In order to verify the stability of the upgraded monitor, a one-year continuous measurement of atmospheric radon concentration was carried out by setting the upgraded radon monitor at a three-storied building platform, located in the campus of Peking University, Beijing (39.59° N, 116.19° E). The unattended field observation was performed between 15 October 2018 and 1 October 2019. The hourly measurement results are shown in Figure 7, with a data acquisition rate of more than 99.9%.

The high data acquisition rate suggests that the upgraded monitor worked stably and performed well during the one-year observation period. Radon concentration varied with time in a large range from 2.8 Bq·m$^{-3}$ to 30.3 Bq·m$^{-3}$. The annual average radon concentration during the measurement time is 11.1 ± 4.0 Bq·m$^{-3}$, which is nearly at the same level of both the previous 5-year result of 12.1 ± 4.9 Bq·m$^{-3}$ in Beijing [14] and the average value of global outdoor radon concentration of 10 Bq·m$^{-3}$ [30].
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![Image of Figure 6 showing comparison results between the upgraded radon monitor and AlphaGUARD at different levels of radon concentrations and relative humidity.]

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![Image of Figure 7 showing hourly measurement result of atmospheric radon concentration in Beijing.]

4. Conclusions

Radon, as a radioactive ideal tracer gas, is widely used in atmospheric science today. Due to its variation and low level, continuous high-sensitivity measurement is required for atmospheric study application, and an electrostatic collection method is most appropriate. In this paper, an upgraded radon monitor was newly developed after many years of previous application with ERM, and two major improvements on the detection module and dryer module were achieved. Calibration and comparison experiments were performed in detail in different experimental conditions of humidity and radon concentration. As a field test of environmental suitability, a one-year continuous measurement on atmospheric radon concentration was carried out and reported in this paper.

The measurement sensitivity of the upgraded monitor was evaluated to be 24.3 cph/(Bq·m\(^{-3}\)), and the lower level detection limit was 0.2 Bq·m\(^{-3}\) for a 1-h cycle at the absolute humidity of 0.34 g·m\(^{-3}\). This sensitivity is higher compared to that of Wada’s system (19.6 cph/(Bq·m\(^{-3}\))) and Iida’s system (17.8 cph/(Bq·m\(^{-3}\))), both of which are at the same chamber volume. The lower level detection limit is comparable with that of Wada’s system (0.2 Bq·m\(^{-3}\)), which makes the upgraded monitor satisfy the need of atmospheric radon measurement in a terrestrial environment. However, for coastal places or islands where the radon concentration is sometimes lower than 0.1 Bq·m\(^{-3}\), a more sensitive measurement system might be needed, or a longer measurement cycle might be required for an upgraded monitor.

Due to the simple structure and stable performance of the air dryer system, the upgraded monitor can meet the need of in situ online measurements of atmospheric radon for the long term with little maintenance, and this is an obvious advantage compared with previous radon monitors.

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