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A new-designed system for continuous measurement of radon in water

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ABSTRACT

On-line continuous monitoring of radon concentration in water is of great significance for its environmental application as a radioactive tracer, for example, as a potential precursor for earthquake forecast and volcanic eruption. To realize on-line continuous measurement on radon in complex water body, a compact measurement system mainly consisted of a simple degassing device and an electrostatic radon monitor is newly developed. The sensitivity of the measurement system is 73 ± 5 cph/(Bq/L), and the detection limit is 0.04 Bq/L with a 60-min cycle at 25 °C water temperature. Intercomparison measurements with RAD H2O were performed both in laboratory condition and in field, and consistent results within the error range were achieved. To test the developed measurement system, a continuous monitoring of radon concentration in water in the drainage tunnel of Mount Jinping was performed for 3 months. The arithmetic mean of radon concentration in water is 0.34 ± 0.09 Bq/L, varying in the range of 0.04–0.60 Bq/L during the period. Several rapid decreases of radon concentration in water were observed, which might be attributed to the increase of rainwater mixing in the drainage tunnel caused by heavy rainfall. The stability of long-term operation of the system enables it to be widely used in the field of radon in water as a tracer.

1. Introduction

Radon (^{222}Rn) is a natural radioactive noble gas produced by the alpha decay of ^{226}Ra in the uranium decay chain and widely exists in rocks and soils, and it has a considerable solubility in water or migrates as bubbles with water. Water contaminated with radon from soils and rocks is one contributor to indoor radon, while for drinking water, it also contributes to human internal exposure (UNSCEAR, 2006; NRC, 1999; Moreno et al., 2014). Due to its special characteristics of long-distance migration, radon in water is also an important tracer in related research fields, such as hydrology, oceanography and geology (Corbett et al., 1997; Schmidt and Schubert, 2007; Santos et al., 2008; Baskaran, 2016). Especially for the forecasting of geological activities, such as earthquakes and volcanic eruption which can affect radon concentration in groundwater, radon concentration anomaly is an effective potential precursor (Talwani et al., 1980; Teng, 1980; Igarashi et al., 1995; Morales et al., 2020). The long-term on-line continuous monitoring on radon concentration in water is of significance and is widely required.

Measurement methods and techniques of radon concentration in water can be quite different according to different purposes and requirement. Liquid scintillation counting (LSC) method, gamma spectrometry method and emanometric method are recommended by ISO (ISO 13164-1, 2013) and commonly used today. For LSC method, water sample is transferred to scintillation cocktail and then radon atoms are counted in liquid scintillation spectrometry (ISO 13164-4, 2013; Freyer et al., 1997; Salonen, 2010; Schubert et al., 2014; Saha et al., 2018). For gamma spectroscopy method, gamma-ray emitted by radon progeny ^{214}Bi and ^{214}Pb, which are in equilibrium with ^{222}Rn, is measured (ISO 13164-2, 2013; Sanchez et al., 1995). These two kinds of methods, measuring radon directly in water sample, are relatively simple in processing, and are not influenced by water temperature (Jobbagy, 2017). They are suitable for laboratory measurement but not quite suitable for field measurement since their measurement systems are usually too heavy to move. Furthermore, they cannot be used for continuous measurement.

For emanometric method, which is the mostly used method for field measurement on radon in water, the dissolved radon is degassed into the air, and the radon-in-air concentration is transferred and measured by a radon monitor. [ISO 13164-3, 2013] There are three ways commonly used for degassing process. One way is diffusion, in which radon diffuses

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naturally from water surface into air for a long time to achieve the equilibrium between water and air (Galli et al., 2000; Plastino and Bella, 2001; Papastefanou, 2002). It is simple but cannot response to the quick change of radon concentration in water because of its relatively long response time. To solve this problem, the bubbling method is developed as the second way. In this method, the time to get to the equilibrium of radon-in-water and radon-in-air concentration is shortened by increasing the contact area between water and air with bubbling probe. Nowadays, this method is widely used as an accessory device for some commercial radon monitors (Burnett et al., 2001; Dulaiova et al., 2005; Dimova et al., 2009; Stieglitz et al., 2010; Durridge Co Inc, 2018a), and commercial degassing instruments such as RAD H2O(De Simone et al., 2015; Durridge Co Inc, 2018b), RAD AQUA (Durridge Co Inc, 2018c) and AquaKIT (Eenitron Ins, 2008) are developed. The third way is the degassing membrane method, which is developed along with the progress of materials science. In this method, the degassing process is achieved by using a degassing membrane. Degassing membrane has been used for radon-in-water measurement since 1996, and improved work has been presented in recent years (Schmidt et al., 2008; Hofmann et al., 2011; Pujol and Perez-Zabaleta, 2017; Lee et al., 2019). To make radon concentration in water a possible precursor for earthquake forecast, we also developed a continuous measurement device based on degassing membrane (Wang et al., 2020). However, after a long-term in-situ operation in real environment at a seismic station near Beijing, we found the limitation of the degassing membrane method which demands high water quality, and meanwhile we also noticed the complex change of groundwater quality.

To realize continuous measurement on radon concentration in water for a long-term observation, a compact system mainly consisted of a simple degassing device and an electrostatic radon monitor is newly developed by this study. Intercomparison measurements with RAD H2O were performed both in laboratory condition and in real site. The developed measurement system was successfully applied for a continuous monitoring of radon concentration in water in the drainage tunnel of Mount Jinping in Sichuan province for about 3 months, and we analyzed the results in detail at the last of this study.

2. Materials and methods

2.1. Measurement system and its principle

Fig. 1 is the schematic diagram of the new designed system for continuous measurement of radon in water. It is consisted of a simple degassing module and radon detector module. The below part of the cylindrical detector is inserted into the up part of the degassing cuboid, making the measurement system integrated and compact.

For degassing module, it is a brand newly designed 20 cm × 10 cm × 10 cm sealed stainless steel cuboid fully filled with small bubbling stones (main chemical components include SiO_2, Al_2O_3, Na_2O, CaO; 3D filter medium, Aqua-Clean, Zongli Tec.co. China),a kind of zeolite with irregular shapes and large effective surface areas, and their side length are less than 15 mm. Water current flows in and out through the bubbling stones in the bottom part of the cuboid naturally or by pumping. Radon, both in air bubbles and dissolved in water, can degas from water and diffuse into the air in the up part of the cuboid. The degassing capability, depending on the contact area and time of water and zeolite, is controlled by a certain constant flowrate maintaining the equilibrium of radon concentration between water body in the bottom part and air in the up part of the cuboid. In the up part of the cuboid, by passing through a thin waterproof membrane (Haichengshijie, China), radon diffuses into the detector chamber and is measured there. Water flowrate is controlled in the range of 0.3 L/min to 3 L/min considering the optimization of degassing efficiency and the responding time to the variation of radon concentration in water.

For radon detector module, the measurement techniques of a self-developed continuous monitor for radon in soil is adopted (Wang et al., 2021). Radon diffuses into an electrostatic chamber, whose volume is about 0.05 L. A 10 mm × 10 mm Si-PIN detector (S3209, Hamamatsu Co, Japan) is mounted inside. A high voltage of −200V is loaded on the detector and the positive ^{218}Po and ^{214}Po particles originating from radon decay could be collected on the surface of the detector, and the alpha spectrum obtained could be analyzed by a multi-channel analyzer. More information of humidity-response correction is given in detail in our previous work (Wang et al., 2021).

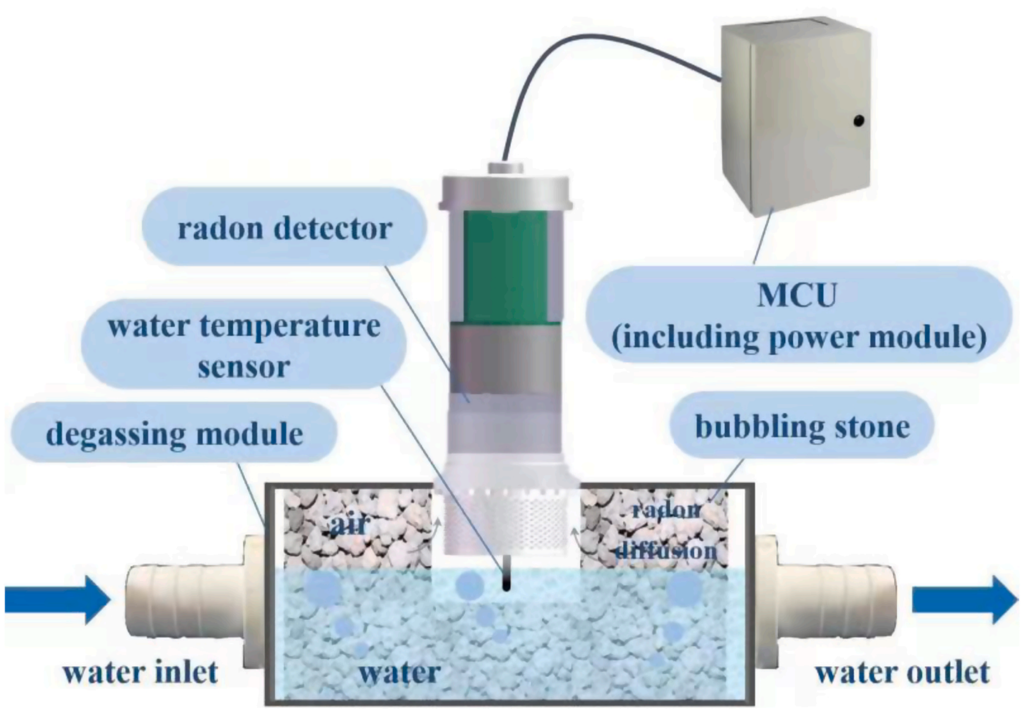

Fig. 1. Schematic diagram of the system for continuous measurement of radon in-water.

What radon detector measures directly is the radon concentration of the degassed air. For the calculation of radon concentration in water, a water temperature sensor (PT100; Dekong China) with an accuracy of 0.1 °C is installed under the radon detector module, see in Fig. 1., and water temperature is automatically measured and recorded by the main control unit (MCU). Radon concentration in water is automatically calculated based on radon concentration in degassing air and water temperature, and all measurement results are then uploaded to data center automatically and remotely.

On the basis of the principle of dissolution equilibrium, the relationship between radon concentration in water C_{water} and radon concentration in air C_{air} can be given as follows (Kertes, 1979):

$$C_{water} = \alpha(T) \cdot C_{air} \tag{1}$$

where $\alpha(T)$ is defined as partition coefficient, presenting the ratio of C_{water} (Bq/L) to C_{air} (Bq/L) at a certain water temperature, which only relies on temperature (T in °C) under the pressure of 1 atm, and is given in Eq. (2) (Weigel, 1978):

$$\alpha(T) = 0.105 + 0.405 \cdot e^{-0.0502 \cdot T} \tag{2}$$

Under an equilibrium condition, radon concentration in water, C_{water}, can be calculated by adopting formula (1) and (2), on the basis of the measurement results of both water temperature and radon concentration in air above water, C_{air}. The uncertainty of radon concentration in water can also be evaluated by error transfer formula on the uncertainty analysis of measurements on both water temperature and radon concentration in air.

2.2. Calibration and intercomparison experiments

2.2.1. Calibration

Calibration of the radon detector was performed in a temperature-humidity adjustable control box with an effective volume of 150 L (HS–250B, KOWINTEST, KW-TH, China). The relative humidity could be adjusted from 10% to 95% with an uncertainty of ±2%, and the temperature range is from −10 °C to 40 °C with an uncertainty of ±0.3 °C. The soil gas was used as radon source, which was pumped from 1 m depth in soil at a flow rate of 2.5 lpm. Radon concentration in the temperature-humidity adjustable control box was measured by an AlphaGUARD PQ2000 monitor (Saphymo, France), which can be traced back to the National Radon Standard of Metrological Institute of China, and the level of radon concentration in the box is also adjustable in a relative range (Mao et al., 2020). Radon concentration in water can be calculated based on formula (1) and (2) by adopting the above calibrated radon detector and the water temperature sensor mentioned.

2.2.2. Intercomparison experiments

To ensure the measurement accuracy of radon concentration in water, we took the widely used commercial instrument for measurement on radon in water, RAD H2O (Durride, USA) device, and a calibrated RAD7 monitor (Durridge, USA) as a reference instrument, and performed intercomparison experiments in both laboratory condition and in field condition respectively.

Fig. 2 is the schematic diagram of the intercomparison experiment system for radon concentration in water in laboratory condition. The experiment system is consisted of main measurement part (left side) and radon source circulating part of water sample (right side). 20 L purified water was transfused into a 30 L sealed stainless steel barrel, and water sample was made by a solid ^{226}Ra source circulating system.

Radon concentration in the water sample increased gradually when

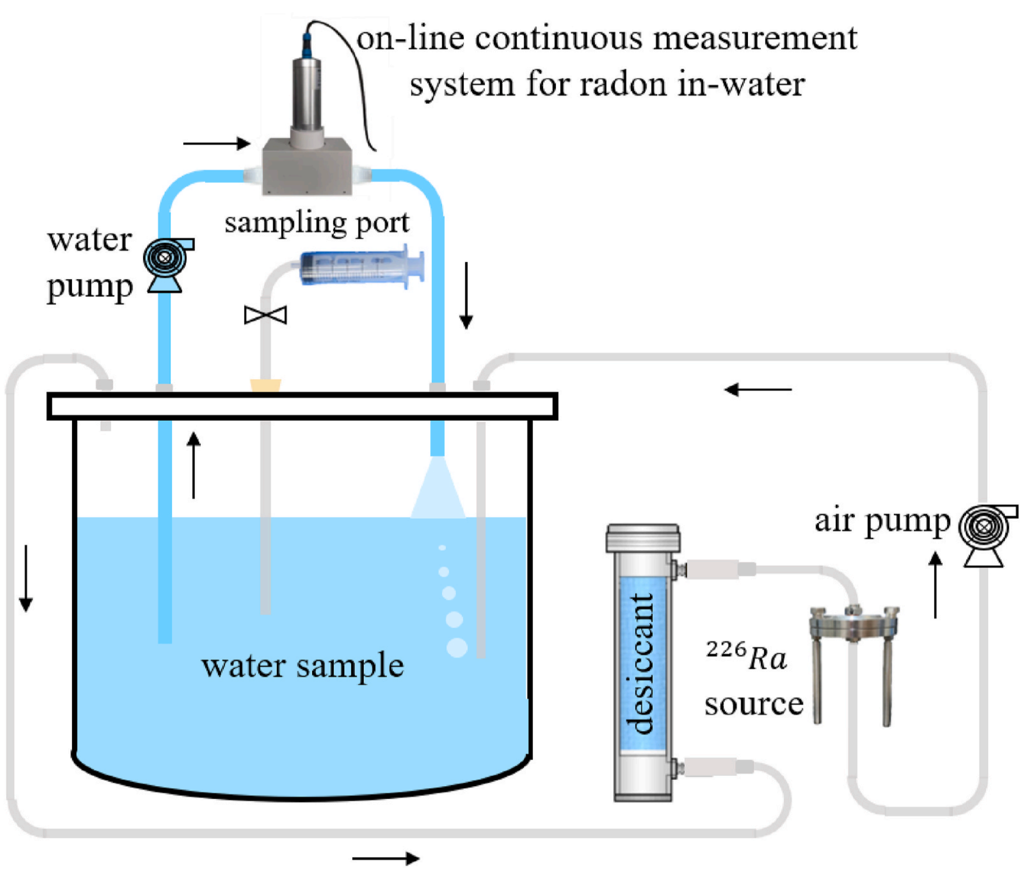

Fig. 2. Schematic diagram of intercomparison experiment system for radon in water in laboratory.

the air pump (Haixun, China) of the radon source circulating system started work. When the developed continuous measurement system (left side above) started to work simultaneously, the change of radon concentration in the water sample could be measured and recorded automatically with a measurement cycle of 60 min. At the same time, water samples with a volume of 40 ml (accessory of RAD H2O) were collected aperiodically from the sampling port on the lid of the barrel and then measured according to RAD H2O user operation manual (Durridge Co Inc, 2018b). For our comparison experiment, two 40-ml water samples were collected each time, and to get different levels of radon concentration in water, 8 times of water sampling were performed in total.

For intercomparison experiments in field measurement, we took tap water which was pumped from a well in a county near Beijing as water sample. The measurement system was connected with the tap water pipe, as shown in Fig. 3, water flowing in and out the degassing module. A 48-h continuous measurement with a measurement cycle of 60 min was carried out. For the intercomparison, 4 times of water sampling were done and measured with RAD7 monitor based on RAD H2O user manual during the period.

3. Results and discussions

3.1. Sensitivity and limit of detection of the measurement system

Table 1 gives the calibration results carried out at different absolute humidity conditions. The results indicate that the sensitivity of the radon detector has a negative correlation with the absolute humidity of the electrostatic measurement chamber, and their correlation curve can be fitted (Wang et al., 2021). On the assumption that the relative humidity of the air degassed from water is about 100%, and the temperatures of water and air are the same, 25 °C, its absolute humidity is then calculated to be 22.9 g/m^3. Furthermore, the measurement sensitivity for radon in water in this condition is evaluated to be 73 ± 5 cph/(Bq/L) based on the partition coefficient at 25 °C which is $\alpha(T) = 0.22$.

Table 1
Calibration results of sensitivity at different absolute humidity.

Absolute humidity (g/m^3)	2.1	3.8	13.6	14.9	19.3	25.6	34.6
Sensitivity (cph/(Bq/L)	45.5 ± 3.1	40.1 ± 2.3	26.7 ± 2.1	24.1 ± 1.9	18.8 ± 1.2	12.1 ± 1.4	9.6 ± 0.6

For background evaluation of the system for continuous measurements of radon in water, high purified Nitrogen, flowing through activated carbon, was pumped into the system, and the background measurement lasted for 72 h. The background of the system was 0.03 ± 0.02 cph, and the lower limit of detection (LLD) was then calculated to be 0.04 Bq/L (k = 2, 95% confidence level) with a 60-min measurement cycle and 25 °C water temperature.

3.2. Intercomparison experiment for radon in water in laboratory

The results of intercomparison experiment of the developed measurement system and RAD H2O instrument in laboratory condition are shown by Fig. 4, in which, abscissa is the result of measurement system, and ordinate is the result of RAD H2O, and the dotted line means y = x.

Fig. 4 suggests that results of the two different methods are in good agreement at different levels of radon concentration in water, all

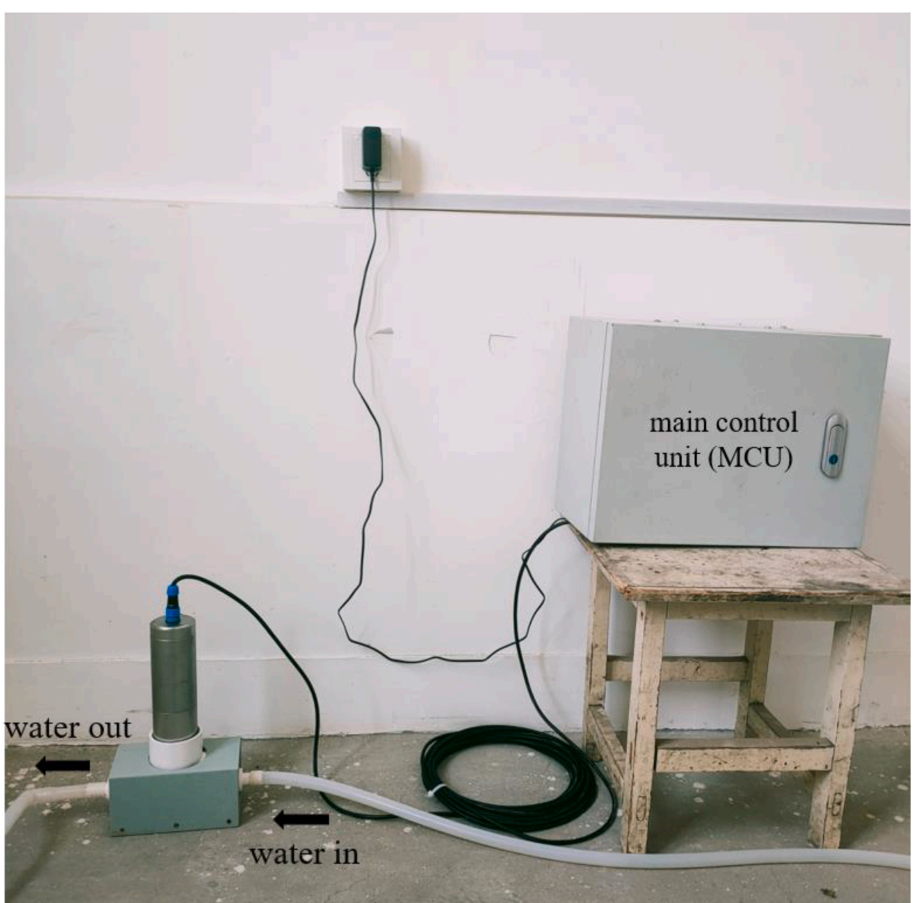

Fig. 3. Photograph of field measurement on radon in tap water.

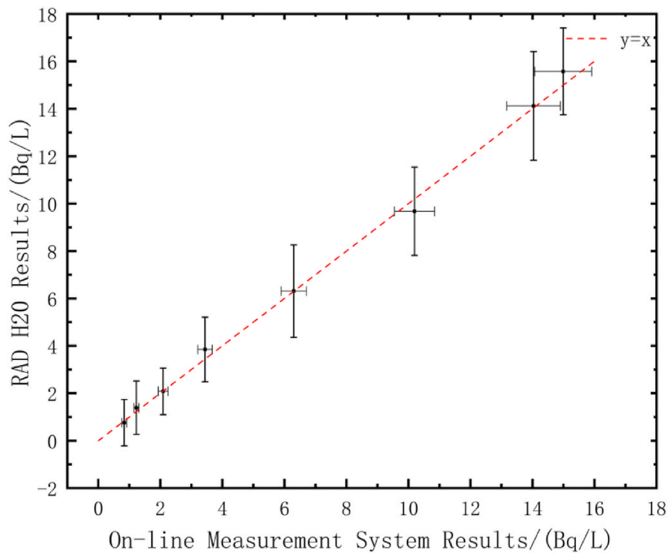

Fig. 4. Comparison Results of the continuous measurement system and RAD H2O in laboratory condition.

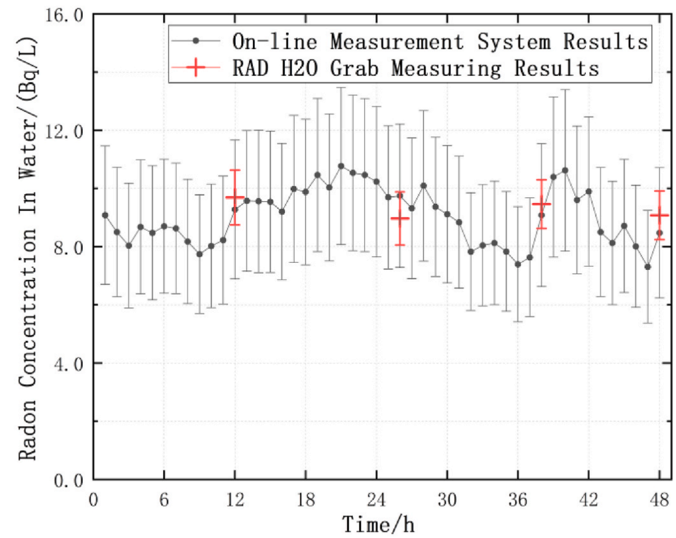

Fig. 5. Comparison results of the continuous measurement system and RAD H2O in field measurement.

deviations within the error range. The consistency of the 8 comparison experiments is 0.99 ± 0.22.

3.3. Intercomparison experiment for radon in water in field measurement

Fig. 5 shows the comparison results of the developed system for continuous measurement of radon in water and RAD H2O in a field measurement. The abscissa is monitoring time in hour (monitoring was preformed for 48 h in total). The ordinate is radon concentration in water. 4 grab sampling results of RAD H2O and results of continuous measurement with a 60-min cycle are indicated respectively.

The results of this continuous measurement indicate that radon concentration of the tap water varied in a certain range, (7.3–10.8) Bq/L, and the average concentration of the 48-hourobservation period is 9.1 ± 0.3 Bq/L. For the 4 times of grab sampling of RAD H2O, all deviations are within error range, and the average concentration of them is 9.3 ± 0.6 Bq/L.

3.4. A field continuous measurement on radon concentration in drainage tunnel water of Mount Jinping

Environmental stability of long-term operation system is highly required for radon monitoring in water. For testing in practice, we installed the measurement system at a drainage tunnel of Mount Jinping, Sichuan Province, where the Chinese underground laboratory for rare-event related physical experiments is located. The water body of the drainage channel is an underground river formed by rainwater, water seepage inside the mountain and spring water. The width of the river is about 3 m, the water temperature is constant at about 10 °C, and the water flow is abundant. The continuous measurement on radon concentration in water was carried out from Aug.6 to Nov. 24, 2020, with a 60-min cycle. A total of 2613 valid data are obtained, and the effective data acquisition rate is about 100%. Fig. 6 shows the results.

The results suggest that radon concentration in the drainage water was not high in general, varying from 0.04 Bq/L to 0.60 Bq/L, and that the average concentration of the observation period is 0.34 ± 0.09 Bq/L. They also suggest that ^{226}Ra content in the rock of Mount Jinping is not high. However, it can be seen from the figure that several rapid decreases of radon concentration in water occurred during the observation period. According to local metrological record, rainfall occurred at corresponding time. It is speculated that these phenomena were due to the increase of rainwater mixing in the drainage tunnel caused by heavy

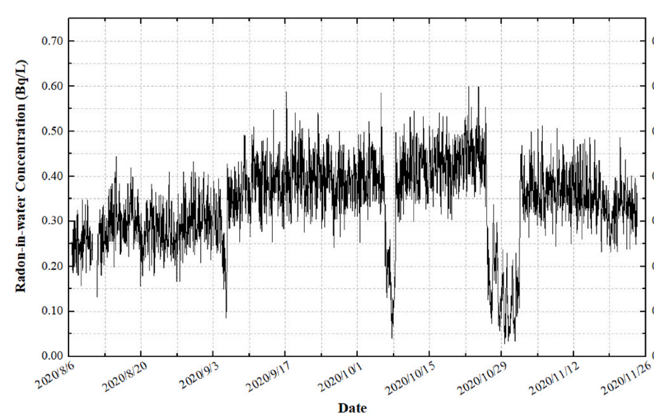

Fig. 6. Radon concentration in water of a drainage tunnel of Mount Jinping (Aug.6-Nov. 24, 2020).

rainfall.

The performance of the 3 months field measurement suggests the stability of long-term operation of the system. Nevertheless, to maintain a consistent degassing capability in practice, periodical replacement of bubble stones should be considered according to water quality, and a replacement at least every 6 months is suggested.

4. Conclusion

To meet the requirement of on-line monitoring of radon concentration in complex water quality, this study developed a new system for continuous measurement of radon in water consisted of a simple degassing module and a radon detector module. The sensitivity of the measurement system is 73 ± 5 cph/(Bq/L)(water-air equilibrium at 25 °C), and the detection limit is 0.04 Bq/L (60 min cycle). Through intercomparison measurements and a three-month field application, it is proved that the measurement system can accurately and reliably response to the temporal variation of radon concentration in water. Furthermore, the system also shows the stability of long-term operation and easy maintenance, which are the most required characteristics for making radon in water an effective tracer in the areas of earthquake and volcano prediction.

It should be noticed that the calculation of radon concentration in water is based on water-air equilibrium as the measurement principle mentioned above shows, which means accurate results can be achieved for radon dissolved in water. However, in practice, especially in the field such as hot spring and volcanic vent, radon not only dissolves in water but also exists in the form of bubbles, and bubbles can be transported over long distance due to the gradient of water temperature and pressure. In this case, it is hard to get to the equilibrium between air (radon in gas phase) and water (radon in liquid phase). Therefore, attention should be paid to the accuracy and practical significance of measurement results.

CRediT authorship contribution statement

Chunyu He: Writing – review & editing, Writing – original draft, Formal analysis, Data curation. **Zhi Zeng:** Funding acquisition. **Lei Zhang:** Supervision, Methodology, Investigation, Conceptualization. **Yunxiang Wang:** Formal analysis, Data curation. **Qiuju Guo:** Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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