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Study on an on-site radon-in-water measurement system based on degassing membrane

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ABSTRACT

Radon-in-water is an important tracer in hydrology, oceanography and geology, where a demand for fast response and long-term continuous measurement of radon-in-water concentration on-site is necessary. In this paper, an on-site radon-in-water measurement system based on a degassing membrane, an electrostatic radon monitor and a water temperature sensor is presented, and calibration experiment is carried out. The relative degassing efficiency of the degassing membrane was studied using self-made radon-in-water standard, which gives the result of (97.1 ± 7.3) %. The sensitivity of the measurement system is (3730 ± 190) cph/(Bq/L), and the lower limit of detection is 0.005 Bq/L, with 10 min cycle at 25 °C water temperature. The response time of the measurement system is about 26 min, which is independent of the water flowrate within the range of 0.5–2.5 L/min. Using this measurement system, field measurements were carried out on tap water and deep well water. And the measurement system has been successfully used in the continuous monitoring of radon-in-water concentration at a seismic station in Beijing.

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. Drinking water contaminated with radon from soils and rocks contributes to human internal exposure (UNSCEAR, 2000; Moreno et al., 2014). Radon-in-water is also an important tracer in many related research fields, such as hydrology, oceanography and geology (Corbett et al., 1997; Schmidt and Schubert, 2007; Santos et al., 2008). Especially for seismology, earthquakes and other similar geological activities like volcanic explosion will affect the concentration of radon in groundwater, which is usually measured for earthquake prediction (Talwani et al., 1980; Liu et al., 1984; Igarashi et al., 1995; Papastefanou, 2002). The time scale and range of those effects are usually not clear, so the fast response and long-term continuous measurement on radon-in-water concentration is required. Also, monitoring of radon-in-water concentration is needed for radiation and environmental protection.

Based on different purpose and requirement, the radon-in-water measurement methods and systems can be quite different. Most commonly used methods are liquid scintillation counting (LSC) method, gamma spectrometry method and emanometric method, which are recommended by ISO 13164 (2013).

In the LSC method, radon atoms are transferred from the water sample to the scintillation cocktail and then counted in liquid scintillation spectrometry (ISO 13164-4, 2013; Freyer et al., 1997; Salonen, 2010; Schubert et al., 2014; Saha et al., 2018). The sampling process of LSC method is relatively simple and the sampling volume is usually small (Pujol and Pérez-Zabaleta, 2017). The detection efficiency is very high. Measurement results of this method are not influenced by water temperature. But due to the transfer process of samples, the response time of this method is relatively long (Jobbágy et al., 2017). It is suitable for laboratory measurement but not quite suitable for continuous and on-site measurement of radon-in-water.

For gamma spectroscopy method, gamma-ray emitted by radon progeny ^{214}Bi and ^{214}Pb, which is equilibrium with ^{222}Rn, is measured to calculate radon-in-water concentration (ISO 13164-2, 2013; Sánchez et al., 1995). Scintillation spectroscopy or high purity germanium (HPGe) spectroscopy system are mostly used as gamma-ray detection system. This method is simple with no water temperature influence, and the measuring device is easy to setup. But the measurement results can also be influenced by several factors such as radon leakage during

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sample transfer, the ^{226}Ra content in water, sample density and homogeneity (Pujol and Pérez-Zabaleta, 2017; Jobbágy et al., 2017). Also the measurement system is usually too heavy for field measurement and this method cannot be used for continuous measurement.

The emanometric method is the mostly used method for radon-in-water measurement. In this method, 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). Three ways are commonly used to achieve the degassing process. One way is diffusion, in which radon diffuses from water into air for a long time to achieve the balance between water and air. This method is simple, appearing as early as 1970s (Noguchi and Wakita, 1977; Cathey, 1978), and also used in other studies (Galli et al., 2000; Plastino and Bella, 2001). But the response time of this method is relatively long, and cannot response the quick change of radon-in-water concentration. To solve this problem, the bubbling method is developed as the second way. In this method, the balance time of radon-in-water and radon-in-air concentration is shortened by increasing the contact area between water and air with bubbling probe. This method has been widely used, and new measurement methods and device have been developed, such as the improved version of the ionization chamber, a series of devices based on bubbling chambers and RAD7 monitors (Burnett et al., 2001; Dulaiova et al., 2005; Dimova et al., 2009; Stieglitz et al., 2010; Lee et al., 2015; Durridge Co. Inc, 2018a), as well as commercial instruments like RAD H_2O (De Simone et al., 2015; Durridge Co. Inc, 2018b), RAD AQUA (Durridge Co. Inc, 2018c) and some other devices (Teng, 1980). Bubbling method has a shorter response time, and it is commonly used for discrete water sample measurement. Some of them can be used for continuous measurement, but the degassing efficiency is not clear. The third way is degassing membrane method. In this method, the degassing process is achieved by using a degassing membrane. Due to the high degassing efficiency of degassing membrane, this method is used in radon-in-water measurement since 1996, and improved work is presented in recent years (Schmidt et al., 2008; Lee et al., 2019). Due to the high degassing efficiency and the separated water-gas structure of the degassing membrane, it has the shortest response time and can be used for long-term continuous measurement of radon-in-water concentration.

In this paper, a new on-site measurement system for radon-in-water concentration is developed using a degassing membrane, an electrostatic radon monitor and a water temperature sensor. The calibration experiments and field measurements were also carried out in this paper. The details, the principle and the property of the system will be shown and some field measurement results will be given.

2. Materials and methods

2.1. Measurement system

Fig. 1 and Fig. 2 are schematic diagram and objective photo of radon-in-water measurement system, which mainly consists of two parts. The first part is degassing part using the degassing membrane. And the second part is measurement part, which is a radon monitor. These two parts are introduced as follow.

The part in the blue frame on the left side in Fig. 1 is the degassing part. Water can be pumped or flow through the central path of the Liqui-Cel™ Minimodule degassing module (Membrana GmbH, Germany), and radon atoms in the water sample are extracted into the side path of the degassing membrane. According to the product manual, the nominal water flowrate range of the degassing membrane is from 0.5 L/min to 2.5 L/min, and the product should be stably and well performed within the flowrate range. The water flowrate is controlled by LZB-10 liquid flowmeter (Senlod, China). Temperature of water is continuously measured and recorded by PT100 sensor (Dekong, China) with an accuracy of 0.1 °C.

The part in the yellow frame on the right side in Fig. 1 is the measurement part. This part is a self-developed radon monitor based on a

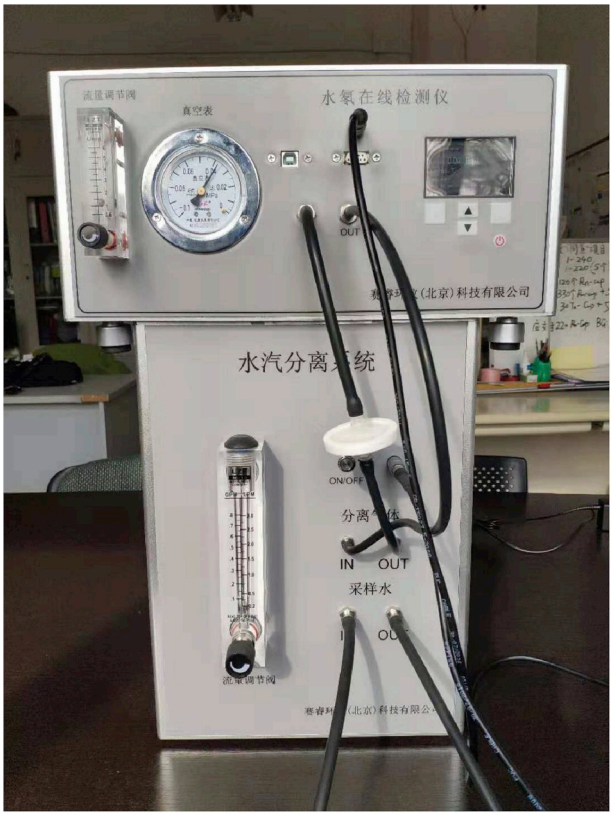

Fig. 2. Objective photo of the measurement system.

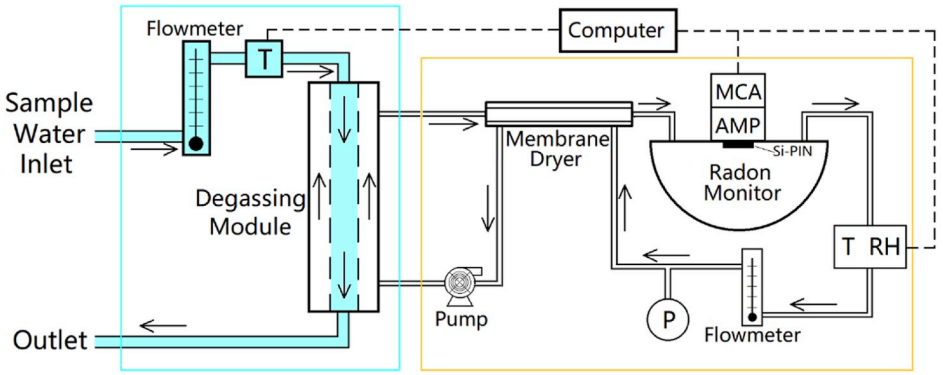

Fig. 1. Schematic diagram of the measurement system.

100 mm^2 Si-PIN detector (Otron Sensor, China), an electrostatic collection chamber with effective volume of 0.8 L, and a multi-channel analyzer. The Si-PIN detector is mounted at the center of the hemispherical collection chamber. The positive ^{218}Po particles, produced by decay of radon, are collected on the surface of the detector by electric field. α particles emitted by collected ^{218}Po will generate pulse signals on the detector, which are processed by the multi-channel analyzer to accumulate an α spectrum. Radon concentration is calculated by calibration factor and counting rate of ^{218}Po energy region. The air flowrates have no effect on the measurement results. However, considering the volume and ventilation rate of the chamber, and influence of negative pressure in the chamber, the air flowrate is optimized to be 1.0 L/min. The sensitivity of the radon monitor was calibrated in radon chamber of Peking University with a standard device AlphaGUARD PQ2000 (Saphymo, France), and uncertainty analysis was done. The lower limit of detection (LLD) of radon monitor was calculated using background measurement with high-purity N_2 and activated charcoal filter, by the recommended procedure in ISO 11929 (ISO 11929, 2019). To control the humidity, an MD070-24 membrane-based humidity control tube (Perma Pure, USA) is connected with the radon monitor, and relative humidity is controlled stable during radon-in-water measurement.

A control system collects all the measurement data such as radon concentration and water temperature. And it can control the air flowmeter, the water flowmeter and measurement cycle, which is developed by Sairatec company (Sairatec, China). The radon-in-water concentration is automatically calculated by radon-in-air concentration and water temperature in this control system. The measurement cycle can be set to different values from 1 min to 2 h.

2.2. Principle

According to the principle of dissolution equilibrium, the relationship between radon-in-water concentration C_{water} and radon-in-air concentration C_{air} is given as follow (Kertes, 1979).

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

where $\alpha(T)$ represents the partition coefficient which only depends on temperature (T in °C) under the pressure of 1 atm, and is given in Eq. (2) (Weigel, 1978).

$$\alpha(T) = 0.105 + 0.405 \cdot \exp(-0.0502 \cdot T) \quad (2)$$

when water sample flows through the degassing module, radon atoms dissolved in the water will partially pass through the degassing membrane and measured by radon monitor. The balance between radon-in-water and radon-in-air can be easily reached due to the high degassing efficiency of the degassing membrane. The relationship between radon-in-water concentration C_{water} in the central path and radon-in-air concentration C_{air} in the side path is shown as Eq. (3).

$$C_{water} = \rho \cdot \alpha(T) \cdot C_{air} \quad (3)$$

ρ is relative degassing efficiency, which is defined as the percentage of radon-in-water and radon-in-air equilibrium in Eq. (1). As the key parameter of the degassing module, it needs to be quantitatively determined. The water temperature is recorded by PT100 sensor and the radon-in-air concentration is measured by the radon monitor, then the radon-in-water concentration is calculated by Eq. (3). Uncertainty of radon-in-water concentration can also be evaluated by uncertainty analysis of degassing efficiency and radon-in-air concentration.

2.3. Relative degassing efficiency and response time

To get the relative degassing efficiency of our degassing module, a special calibration system is designed and shown in Fig. 3, where a 20 L of radon-in-water standard sample was made by a plastic-sealed ^{226}Ra source with about 7 kBq activity, just as the works of Havelka (2009) and Kitto et al. (2010). The radon-in-water standard sample is contained in a stainless steel barrel with a volume of 30 L, which was put in an air-conditioned room with stable temperature 25 °C. The water sample barrel was placed in the air-conditioned laboratory for more than one month waiting for radon concentration to rise and reach a stable status.

The concentration of radon-in-water standard sample was determined by RAD H_2O (Durridge, USA) device, and the radon-in-air concentration degassed by the degassing module was measured by RAD7 monitor (Durridge, USA). All the devices are calibrated in the radon chamber of National Institute of Metrology of China, and can be traced to the national standard. Water samples with volume of 40 mL were collected from the sampling port on the lid of the barrel every 1 h and ten samples were measured. The water was simultaneously circulated through the degassing module by a water pump at 1 L/min flowrate, and the RAD7 monitor continuously measured the radon-in-air concentration C_{air} with 60 min cycle. Then according to the continuously measured water temperature, the relative degassing efficiency ρ is calculated using Eq. (3). Uncertainty of ρ is analyzed by propagation of errors. The errors of C_{air} and C_{water} are taken into account. The error of $\alpha(T)$ can be ignored due to the accurate measurement of water temperature.

For radon-in-water measurement, response time of the measurement system refers to the time from the start of measurement to the time when the measurement value rises to actual radon concentration in water sample. Measurement value can only be used after the response time. The response time of the measurement system was measured using the radon-in-water standard sample above. RAD7 monitor in Fig. 3 was replaced by the self-developed radon monitor, and the measurement cycle was set to be 2 min. Water flowrate should not affect the performance of degassing module within the range of 0.5–2.5 L/min, so 1.0 L/min and 2.0 L/min were chosen as two water flowrates to verify if there is any difference on response time. The air flowrate of radon monitor is stable at 1.0 L/min. Radon concentration curve over time was recorded as soon as the water sample began to flow through the degassing module. The response time of measurement system was determined using the rising time of measured radon-in-water concentration.

2.4. Field measurement

Field measurements of tap water in the laboratory and deep well water in the suburb area of Beijing were carried out for more than 6 h with 10 min cycle. After that, the measurement system was deployed at a

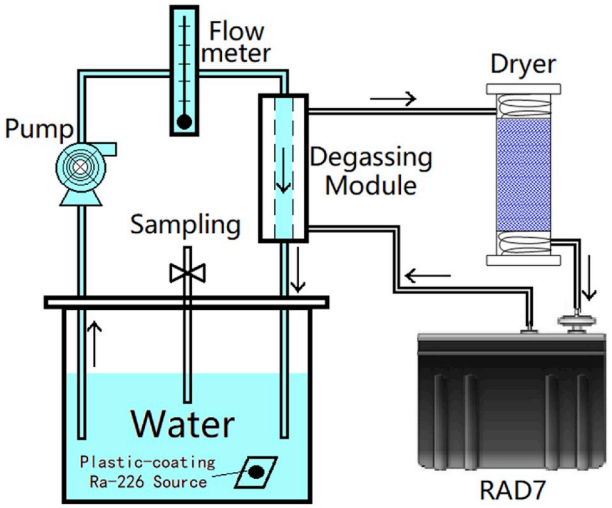

Fig. 3. Schematic graph of relative degassing efficiency determination device.

seismic observation station in Beijing (40.47°N, 115.94°E) for continuously monitoring radon-in-water concentration of water sample. Since the water samples to be tested were all discharged through faucet, it is easy to connect the measuring system to the faucet to achieve the measurement. The air flowrate and water flowrate were set to 1.0 L/min. Gas and liquid leakage inspection was done before the measurement to avoid radon leakage.

3. Results and discussion

3.1. Determination of relative degassing efficiency and response time

Radon-in-water concentration of the standard sample became stable and was over 10 Bq/L after more than one month of standing still. The measurement result of relative degassing efficiency is shown in Fig. 4, which gives the value of (97.1 ± 7.3) % and it is very close to 100%. It means the degassing module has excellent degassing ability. High degassing efficiency also indicates that the degassing module has a quick response to fluctuations of radon-in-water concentration.

The results of response time determination are shown in Fig. 5. The response time of the measurement system is about 26 min, and there is no difference between the two water flowrates. Actually, the response time is mainly contributed by the ventilation of measurement system and the response of ^{218}Po decay.

The sensitivity of the radon monitor section was measured to be (0.80 ± 0.04) cph/(Bq/m^3), and the uncertainty of sensitivity was mainly contributed by systematic error of standard device, and it is also contributed by other factors such as statistical uncertainty of counting and uncertainty of correction to standard condition, so the synthetic uncertainty gave a relative error of 5%. The background of the radon monitor is 0.75 cph, and the lower limit of detection (LLD) of the radon monitor section is calculated to be 23 Bq/m^3 (k = 2, 95% confidence level) with 10 min measurement cycle. Then according to the conversion relationship in Eq. (3), the sensitivity of the entire measurement system is converted to (3730 ± 190) cph/(Bq/L), and the lower limit of detection of the system is 0.005 Bq/L, with 10 min measurement cycle and 25 °C water temperature. The multichannel pulse resolution time is 1 μs, so the upper limit of detection of this measurement system can reach 1000 Bq/L, which far exceeds the actual radon-in-water concentration in the environment.

Comparing these parameters with other existing instruments, including commercial devices and research prototypes, the measurement system has reached a relatively high performance. The sensitivity of 3730 cph/(Bq/L) and lower detection limit of 0.005 Bq/L are better than most current mainstream commercial instruments such as RAD H_2O (Durridge Co. Inc, 2018b) and RAD AQUA (Durridge Co. Inc, 2018c). The response time less than 30 min is the highlight of the system, and it is faster than existing membrane-based systems (Schmidt et al., 2008; Hofmann et al., 2011; Durejka et al., 2019), which are generally more than 40 min. Plus, it achieves the complete separation of liquid and gas, which avoids the influence of liquid on the measurement. And the system does not require the use of a desiccant, which reduces a lot of trouble for the users and makes it suitable for long-term in-situ measurement.

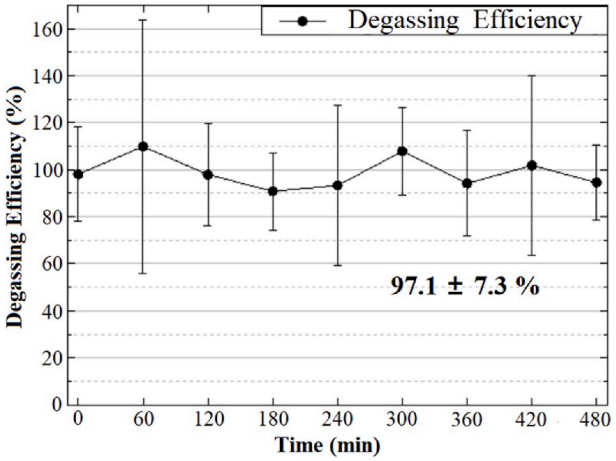

Fig. 4. Determination result of relative degassing efficiency.

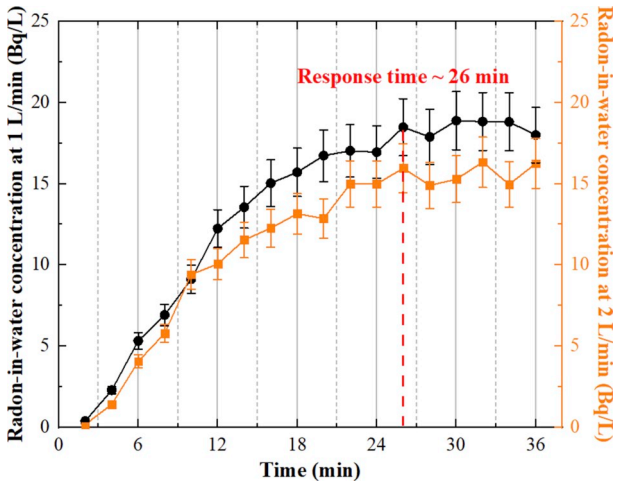

Fig. 5. Determination result of response time of the measurement system.

3.2. Field measurement

Field measurement results of tap water in the laboratory and deep well water are shown in Fig. 6. Relative humidity in the collection chamber was stably controlled at (20 ± 3) %, so it cannot affect the radon measurements. There is big difference between radon-in-water concentrations of tap water and well water, which were measured to be (0.04 ± 0.01) Bq/L and (11.87 ± 0.30) Bq/L, respectively. The radon-in-water concentrations changed very little within the uncertainty range in 6 h measurement. The results show that the measurement system can be used for field measurement with a large range of radon-in-water concentration.

Fig. 7 is the on-site monitoring result at the seismic station for more than three days. It can be seen that the variation of concentration of water sample had a wide range both in amplitude and time scale. The measurement system is able to identify those abnormal variations due to its high and stable relative degassing efficiency and short response time, which supports the continuous measurement and quick response on radon-in-water concentration. These properties are of great importance

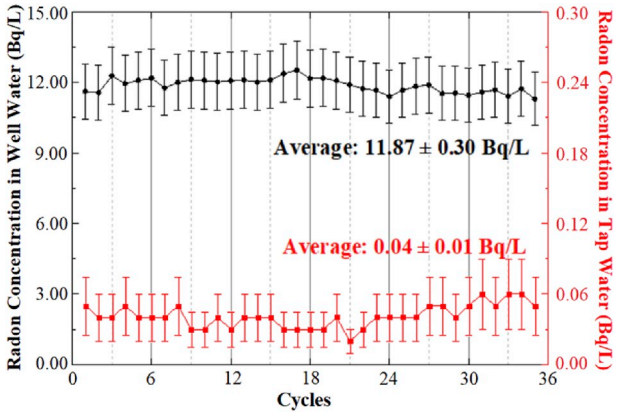

Fig. 6. Field measurement results of laboratory tap water and deep well water.

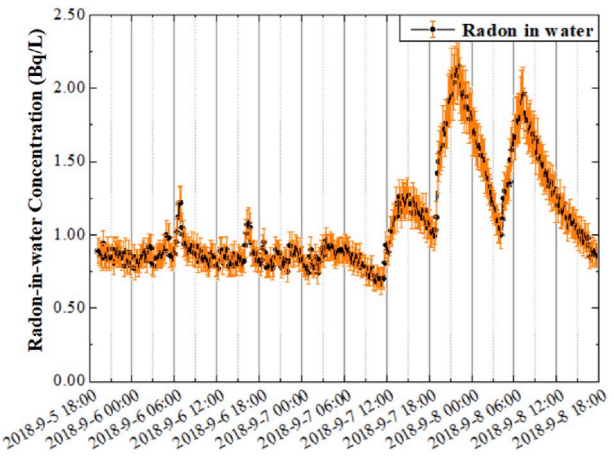

Fig. 7. On-site monitoring result of radon-in-water concentration at a seismic station for 3 days.

in radon-in-water monitoring for research on tracer techniques of radon-in-water, especially for earthquake prediction and other relative research. The physical mechanism behind those large-range abnormal variations of radon-in-water concentration is not clear yet. More data accumulation is necessary, and it need to be further studied.

4. Conclusions

In this study, an on-site radon-in-water measurement system has been developed. The sensitivity of the measurement system is (3730 ± 190) cph/(Bq/L), and the lower limit of detection is 0.005 Bq/L, with 10 min cycle and 25 °C water temperature. The response time of the system is about 26 min. Due to the high and stable relative degassing efficiency and high sensitivity, the measurement system can achieve the fast responsive and continuous measurement of radon-in-water concentration without the influence of environmental parameters such as air humidity, water temperature and air pressure. Field measurement results proved that the system is suitable for continuous measurement in situ. More data is still being collected at the seismic observation station in Beijing, and it is hoped to be helpful for earthquake prediction and other related research.

Declaration of competing interests

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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