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# New-designed measurement device for radon and thoron activity concentration based on gas direct detection

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#### ABSTRACT

A new-designed measurement device for radon and thoron activity concentration is developed based on gas direct measurement to support their in-situ calibration. It consists of a 2000 mm<sup>2</sup> Passivated Implanted Planar Silicon (PIPS) detector, a Multi-Channel Analyzer (MCA), a Micro Controller Unit (MCU), and a small electrostatic chamber with a volume of nearly 23 ml. The device records those alpha particles emitted from radon and thoron gas, and the detection efficiency and the crosstalk factor of  $^{218}$ Po/ $^{216}$ Po are determined by Monte Carlo simulation. Measurement results have been compared with AlphaGUARD DF2000 in pure radon and thoron environments, respectively. Results show that the measurement results of the devices and the reference monitor agree well with each other, with an average relative deviation of 0.48% for radon gas from about 3300 Bq/m<sup>3</sup> to 38 kBq/m<sup>3</sup> and -3.25% for thoron gas from about 25 kBq/m<sup>3</sup> to 70 kBq/m<sup>3</sup>. Uncertainty assessment has also been done, and a relative system uncertainty of radon is about 6.8%, while that of thoron is nearly 7.3%.

## 1. Introduction

As nuclides in the Uranium and Thorium decay series, radon ( $^{222}$ Rn) and thoron ( $^{220}$ Rn) occur everywhere on Earth. Indoor radon exposure has been epidemiologically proven to be the second cause of lung cancer after smoking (WHO, 2009). And as reported by UNSCEAR, the inner exposure of radon and thoron is one of the most important sources of natural radiation to humans (UNSCEAR 2000). Therefore, the accurate measurement of radon and thoron gas is of great importance, and the quality accuracy and quality control depend on the reliable metrological system of radon/thoron activity concentration.

Different methods have been developed for the metrological system of radon activity concentration, which can be divided into two types, namely with sources and without sources. The radon metrological method with sources (Collé et al., 1990; Linzmaier and Röttger 2013; Röttger et al., 2014; Mostafa et al. 2016, 2017; Mertes et al., 2020) is based on the <sup>226</sup>Ra activity reference and stable radon emission, and it was adopted in many radon chambers in early stage. Nevertheless, it relies both on the accuracy of <sup>226</sup>Ra activity determination and the stability of radon emanation. The radon metrological methods without sources include the radon primary standard system based on frozen radon source and defined solid angle method (Picolo 1996; Picolo et al.,

2000; Sabot et al. 2016a, 2020), and the multi-wire ionization chamber (MWIC) system (Busch et al., 2002; Linzmaier and Röttger 2014). Both methods can directly determine <sup>222</sup>Rn activity or radon activity concentration accurately combined with a precise volume measurement, and usually, a secondary standard is needed for radon reference transfer.

Due to the short half-life of <sup>220</sup>Rn (55.6 s), there are still difficulties in thoron activity concentration measurement, and studies did not show enough interest in the accurate measurement of thoron in the past. A few reference methods of thoron activity concentration are developed nowadays, including the <sup>232</sup>Th/<sup>228</sup>Th activity reference method (Möre et al., 1996; Qiu 2006; Röttger et al., 2010; Tang et al., 2012; Buompane et al., 2013; Wang et al., 2017; Rinaldi et al., 2022), the Lucas scintillation chamber method (Tokonami et al., 2002; Zhang et al., 2020; Sakoda et al., 2015), and the gas direct detection method (Sabot et al. 2015, 2016b; Ambrosino et al., 2020). Similar to radon, the  $^{232}$ Th/ $^{228}$ Th activity reference method is based on the stable emission of thoron gas, but is more difficult to realize due to the short half-life of  $^{220}\mathrm{Rn}$  as well as the temperature and humidity influence. The Lucas scintillation chamber method is based on direct measurement of thoron gas and its progeny. Nevertheless, the accuracy is limited by the distribution uniformity of thoron progeny, and it is hardly used as a reference (Zhao et al., 2012). To solve the problems of thoron reference standard, Sabot

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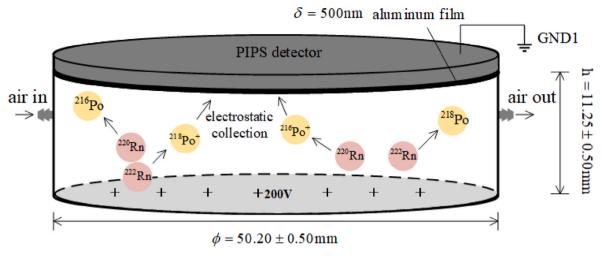


Fig. 1. The sketch map and working principle of the new-designed device.

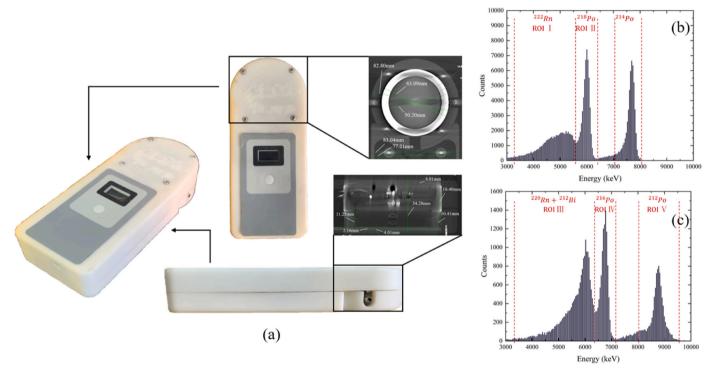


Fig. 2. The appearance picture and the CT photos of the DMD 1 chamber from the top and the side(a), and the measurement spectra in radon(b) and thoron(c) environment with ROIs marked.

et al. developed the gas direct detection method, which is based on directly recorded alpha spectra of thoron gas in a small chamber and is hardly influenced by humidity and the thoron progeny distribution. After that, Ambrosino et al. developed a similar direct measurement system based on the same technical route. Due to its significant advantages and wide applicability to both radon and thoron gas, this direct measurement method seems to be an excellent choice for in-situ calibration of online measurement instruments and could be used for radon/thoron standard transfer.

In recent years, a large number of online radon measurement systems have been installed in China due to the requirement of atmospheric radiation monitoring and the NORM effluent in-situ measurements, which leads to a great demand for field calibration of radon concentration (Zhang et al., 2020). To realize the field calibration of atmospheric radon monitors and the thoron activity concentration standard

transfer, a new-designed measurement device is developed with gas direct measurement in this study, and comparison experiments, as well as uncertainty analysis, were both carried out.

#### 2. Materials and methods

## 2.1. Measurement device and its working principle

The new-designed measurement device consists of a 2000 mm<sup>2</sup> Passivated Implanted Planar Silicon (PIPS) detector (AS2000E, HZDR-innovation GmbH, Germany), a Multi-Channel Analyzer (MCA), a Micro Controller Unit (MCU), and a small electrostatic chamber with a volume of nearly 23 ml. There is a 0.45 µm thick filter with a 25 mm diameter at its inlet to prevent the entry of those daughters, the detector at its top surface is covered by an aluminum film of about 500 nm thick,

and +200 V high voltage is applied to its bottom surface to form an electrostatic field. Two same measurement devices numbered Direct Measurement Device 1 (DMD 1) and DMD 2 are manufactured by 3D printing. The sketch map and working principle are shown in Fig. 1, and the appearance picture, the CT photos as well as the measurement spectra are shown in Fig. 2.

The detailed geometry size of the inner electrostatic chamber was determined by CT photography with a CT imaging equipment (X5000, North Star Imaging, USA) at the National Metrology Institute of China, with an accuracy of  $\pm 0.5$  mm. The inner chamber of DMD 1 is a small cylinder with a height of 11.25 mm and a diameter of 50.2 mm, and that of DMD 2 with a height of 11.94 mm and a diameter of 50.1 mm.

In the flowing working mode, radon/thoron is sampled into the inner chamber by pumping and decays into a series of radon/thoron progeny which are enriched on the detector due to electrostatic collection. The radon/thoron gas and its short-lived progeny emit alpha particles with

$$C_{\text{Tn}} = \frac{N_{\text{Tn}} - N_{^{216}\text{Po}} \bullet \mu_{^{216}\text{Po}} - N_{^{212}\text{Po}} \bullet r_{212}}{V \bullet T \bullet \eta_{\text{To}}},$$
(2)

where N<sub>Tn</sub>, N<sub>216Po</sub> and N<sub>212Po</sub> are alpha counts from thoron in ROI III during T (s), from <sup>216</sup>Po in ROI IV, and from <sup>212</sup>Po in ROI V, respectively, and  $\mu_{216Po}$  and  $\eta_{Tn}$  represent the crosstalk factor of <sup>216</sup>Po to ROI III (dimensionless) and the detection efficiency of thoron (dimensionless), separately.  $r_{212}$  is the ratio of <sup>212</sup>Bi activity and <sup>212</sup>Po activity at the secular equilibrium (dimensionless), which is defined to subtract the interference of <sup>212</sup>Bi from ROI III and can be calculated as follows (Bé et al., 2004):

$$r_{212} = A(212Bi) / A(212Po) = 0.5625.$$
 (3

The uncertainties of  $C_{\rm Rn}$  and  $C_{\rm Tn}$  can be given by the following formulae:

$$u(C_{Rn}) \approx \frac{\sqrt{u^{2}(N_{Rn}) + \mu_{218p_{0}}^{2} \bullet u^{2}(N_{218p_{0}}) + N_{218p_{0}}^{2} \bullet u^{2}(\mu_{218p_{0}}) + C_{Rn}^{2} \bullet T^{2} \bullet [u^{2}(V) \bullet \eta_{Rn}^{2} + u^{2}(\eta_{Rn}) \bullet V^{2}]}}{V \bullet T \bullet \eta_{Pn}}$$
(4)

$$u(C_{\text{Tn}}) \approx \frac{\sqrt{u^{2}(N_{\text{Tn}}) + \mu_{^{216}P_{0}}^{2} \bullet u^{2}(N_{^{216}P_{0}}) + r_{^{212}}^{2} \bullet u^{2}(N_{^{212}P_{0}}) + N_{^{216}P_{0}}^{2} \bullet u^{2}(\mu_{^{216}P_{0}}) + C_{\text{Tn}}^{2} \bullet T^{2} \bullet [u^{2}(V) \bullet \eta_{\text{Tn}}^{2} + u^{2}(\eta_{\text{Tn}}) \bullet V^{2}]}}{V \bullet T \bullet \eta_{\text{Tn}}},$$
(5)

different energy, which could be easily detected by the PIPS detector, and the obtained alpha spectrum is analyzed and recorded by the MCA as well as the MCU. The small chamber is designed to reduce the influence of thoron gas distribution, and the electrostatic collection of the progeny on the detector is aimed to improve the quality of alpha spectra. For activity calculation, only alpha particles from gas are used due to the uniform distribution of gas just like Sabot's idea. However, as shown in Fig. 2(b)(c), the crosstalk interference of the radon/thoron progeny to the ROI of radon/thoron gas is inevitable due to the peak broadening from the detector dead layer and electronic noise. Therefore, use a crosstalk factor to describe the interference of nuclide A to nuclide B ROI, which is defined as the ratio of counts resulting from A in the B ROI to counts in the A ROI. Moreover, not all alpha particles emitted by radon/thoron gas can be recorded in ROI I/III, so the detection efficiency is introduced, which is the ratio of alpha counts caused by radon/ thoron in ROI I/III to alpha particles emitted by radon/thoron. Besides, this device cannot discriminate between alpha particles emitted by radon and thoron gas, so it is designed only for pure radon or pure thoron environments.

Take that the inner volume of the measurement chamber is V (m³), alpha counts from radon gas in ROI I during the measurement time of T (s) are  $N_{Rn}$ , and alpha counts from  $^{218}Po$  in ROI II during T (s) are  $N_{^{218}Po}$ , and ignore the crosstalk of  $^{214}Po$  in consideration of the big energy difference between alpha particles from  $^{214}Po$  and radon. The radon activity concentration ( $C_{Rn}$ , Bq/m³) can then be calculated using the following formula:

$$C_{\rm Rn} = \frac{N_{\rm Rn} - N_{^{218}\rm P_0} \bullet \mu_{^{218}\rm P_0}}{V \bullet T \bullet \eta_{\rm Rn}}.$$
 (1)

In formula (1),  $\mu_{^{218}Po}$  is the crosstalk factor of  $^{218}Po$  to ROI I (dimensionless), and  $\eta_{Rn}$  is the detection efficiency of radon (dimensionless).

Similarly, for thoron activity concentration ( $C_{Tn}$ , Bq/m<sup>3</sup>), the calculation formula is as follows:

where u(i) represents the uncertainty of i. For gas direct measurement, the detection efficiency  $\eta$  is determined by simulation, whose uncertainty u( $\eta$ ) is quite small with a large sampling number. Ignoring the measurement time uncertainty, the main sources of the uncertainty of activity concentration shown in formula (4)(5) are the uncertainty of the chamber volume u(V), the uncertainty of crosstalk factor u( $\mu$ ), and the statistical uncertainty u(N).

# 2.2. Simulation of detection efficiency and crosstalk factor

For absolute measurement, both the detection efficiency and cross-talk factor should be determined by Monte Carlo simulation. During simulation, random sampling of initial positions is carried out to satisfy the uniform assumption of radon/thoron gas distribution in the measurement chamber, and the emission directions are also randomly sampled to satisfy the isotropy of alpha particle emission.

After their energy attenuation in the air, the aluminum film, and the detector dead layer, part of the alpha particles finally reaches the sensitive region of the detector and deposits energy through ionization interaction et al. Only those alpha particles with energy higher than 3 MeV are counted due to the ROI setting. Therefore, radon/thoron detection efficiency can be given by simulation.

The simulation of radon/thoron progeny is similar to those two gases, but the energy of alpha particles and the initial sampling position are different due to the assumption that nearly all  $^{218}\text{Po}/^{216}\text{Po}$  particles are collected to the top surface uniformly. After being emitted by  $^{218}\text{Po}/^{216}\text{Po}$ , part of the alpha particles deposits its energy into the detector after the attenuation in the aluminum film and the dead layer of the PIPS detector. Thus, an energy spectrum of  $^{218}\text{Po}/^{216}\text{Po}$  can be obtained with an assumption of equal dead layer thickness. Because the thickness is hardly known and varies with the detector bias, the actual thickness of the dead layer is given by comparing the simulated energy spectrum and the measured energy peak of  $^{218}\text{Po}/^{216}\text{Po}$  in this paper. Then the crosstalk factor of  $^{218}\text{Po}/^{216}\text{Po}$  can be given by the ratio of

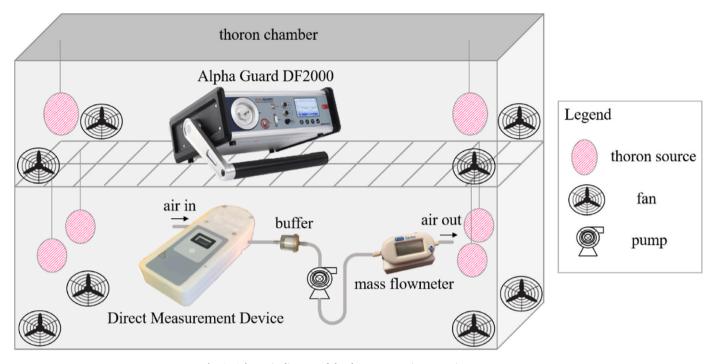


Fig. 3. Schematic diagram of the thoron comparison experiment system.

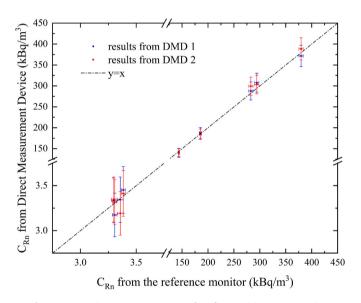


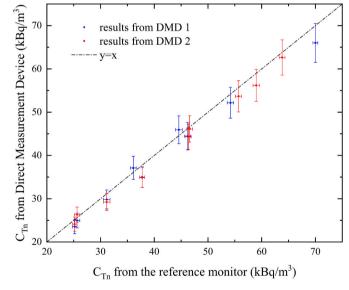
Fig. 4. Comparison measurement results of Rn activity concentration.

counts of ROI I to ROI II or ROI III to ROI IV in the final simulated spectrum of  $^{218}\text{Po}/^{216}\text{Po}$ .

Geant4 software is used in this paper, ten groups of 10000 samples were sampled for each kind of nuclide to determine the detection efficiency and the crosstalk factor, and the arithmetic mean and deviation of the ten calculation results were given as the final results. For DMD 1, the simulation results of  $\eta_{\rm Rn}$  and  $\eta_{\rm Tn}$  are  $0.3051 \pm 0.0067$  and  $0.3198 \pm 0.0050$ , separately; the corresponding results are  $0.2988 \pm 0.0066$  and  $0.3168 \pm 0.0049$  for DMD 2. The simulation results of  $\mu_{\rm 218}_{\rm Po}$  and  $\mu_{\rm 216}_{\rm Po}$  for DMD 1 are  $0.3528 \pm 0.0226$  and  $0.2392 \pm 0.0192$ , and those for DMD 2 are  $0.3356 \pm 0.0215$  and  $0.2234 \pm 0.0179$ .

## 2.3. Comparison experiment in pure radon/thoron environment

To verify the accuracy of the new-designed measurement device,



 $\textbf{Fig. 5.} \ \ \textbf{Comparison measurement results of Tn activity concentration.}$ 

comparison experiments were carried out in a radon/thoron chamber using an AlphaGUARD DF2000 (Saphymo, Germany) monitor as a reference, whose measurement results can be traced back to the national standard of China.

The measurement cycle of DMD 1 and DMD 2 in both comparison experiments is set to be 60 min. The radon comparison experiments were carried out in the big radon chamber of the Chinese National Metrology Institute (Liang et al.,2015). A total of nine comparisons were launched at five different activity concentration levels, four comparisons at 3300  $\rm Bq/m^3$ , two at 28 kBq/m³, and one at 14 kBq/m³, 19 kBq/m³ and 38 kBq/m³, separately. During the comparisons, the temperature and humidity are stably controlled at 21 °C and 28 %RH. At the radon concentration of 3300 Bq/m³, each comparison measurement lasted for at least 40 h, while at the radon concentration higher than 10 kBq/m³ for at least 5 h.

 Table 1

 Results of uncertainty assessment of direct measurement devices.

Device Number	DMD 1	DMD 2
V (ml)	$22.29\pm1.433$	$23.56 \pm 1.450$
$\eta_{ m Rn}$	$0.3051 \pm 0.0067$	$0.2988 \pm 0.0066$
$\eta_{ m Tn}$	$0.3198 \pm 0.0050$	$0.3168 \pm 0.0049$
$\mu_{^{218}Po}$	$0.3528 \pm 0.0226$	$0.3356 \pm 0.0215$
$\mu_{^{216}Po}$	$0.2392 \pm 0.0192$	$0.2234 \pm 0.0179$
$u_{rs}(C_{Rn})^{\mathbf{a}}$	6.75%	6.76%
$u_{rs}(C_{Tn})^{\mathbf{a}}$	7.30%	7.26%

<sup>&</sup>lt;sup>a</sup>  $u_{rs}(C_{Rn}/C_{Tn})$  is the relative system uncertainty of  $C_{Rn}/C_{Tn}$  without statistical uncertainty.

The thoron comparison experiment was carried out in the thoron chamber of Peking University (He et al., 2023) as shown in Fig. 3, where the thoron concentration was adjusted by changing the number of lantern mantles. A diaphragm pump (Lianhezhongwei technology, China) with a flowrate of 1.5 L/min was adopted for sampling, and the flowrate was recorded by a reference mass flowmeter (TSI4046, USA). According to the calculation of flowrate and chamber volume, the influence of thoron decay during sampling is less than 1% and thus is negligible (He et al., 2023). A total of nine comparison measurements were also launched at nine concentration levels from about 25 kBq/m³ to 70 kBq/m³ in the environment of 38 °C and 50 %RH. Each measurement lasted for at least 10 h to lower the statistical uncertainty. Furthermore, as it is difficult to control the long-term stability of thoron gas concentration, the thoron concentrations were not the same when DMD 1 and DMD 2 were compared with the reference monitor.

#### 3. Results and discussion

#### 3.1. Comparison measurement results

The comparison results of radon activity concentration from DMD 1, DMD 2 and the reference monitor are shown in Fig. 4, where the measurement values and uncertainties of direct measurement devices and the reference monitor are marked and the dotted line representing y=x is also shown

Comparison results show that the radon activity concentrations from the two direct measurement devices agree very well with the reference monitor from about 3300 Bq/m $^3$  to 38 kBq/m $^3$ . However, because of the lower sensitivity of direct measurement devices, their measurement uncertainties seem quite larger. The relative deviations of DMD 1 with the reference monitor range from -4.01% to 4.65% with an average deviation of 0.19%, and the relative deviations of DMD 2 with the reference monitor vary from -4.92% to 5.90% with an average of 0.77%.

Fig. 5 shows the results of thoron comparison measurements with the reference monitor, in which measurement results and uncertainties of direct measurement devices and the reference are marked and the dotted line represents y=x.

On the whole, the thoron measurement results of the devices and the reference monitor are consistent with each other from nearly  $25~\mathrm{kBq/m^3}$  to  $70~\mathrm{kBq/m^3}$ . The relative deviations of DMD 1 with the reference monitor are from -7.28% to 3.06% with an average of -3.09%, while those of DMD 2 with the reference range from -7.56% to 2.72% with an average of -3.41%. Also, due to their smaller chambers and lower sensitivities, the thoron measurement uncertainties of DMD 1 and DMD 2 are larger than the reference monitor. However, with good consistency with the reference and the ability to make accurate measurements over a wide range of concentrations, these direct measurement devices are qualified for field calibration and reference transfer.

#### 3.2. Uncertainty assessment

According to formula (4)(5), the main parameters affecting the uncertainty of  $C_{\rm Rn}$ /  $C_{\rm Tn}$  are the uncertainty of the chamber volume, the uncertainty of crosstalk factor, and the statistical uncertainty. Since the statistical uncertainty is different in different measurement periods and can be greatly reduced by extending the measurement period, this study only considers the system uncertainty of measurements other than the statistical uncertainty as the evaluation and comparison parameter of uncertainty.

The uncertainty of V is mainly determined by the accuracy of the CT imaging equipment, which is  $\pm 0.5$  mm. The uncertainty of the crosstalk factor and detection efficiency is determined by multiple sampling and Monte Carlo simulation. Uncertainty assessment results of DMD 1 and DMD 2 are given in Table 1. The average relative system uncertainty of radon measurement for DMD 1 and DMD 2 is about 6.75%, and that of thoron measurement is nearly 7.28%. As shown in Table 1, there is little difference between the uncertainties of the two devices, and the relative system uncertainty of  $C_{\rm Tn}$  is slightly greater than that of  $C_{\rm Rn}$  due to the larger relative uncertainty of  $\mu_{\rm 2^{16}Po}$  than  $\mu_{\rm 2^{18}Po}$ . Besides, the most important source of  $C_{\rm Rn}/C_{\rm Tn}$  system uncertainty found in the assessment is the uncertainty of V, accounting for nearly 89% of the system uncertainty of  $C_{\rm Rn}$  and nearly 95% of the system uncertainty of  $C_{\rm Tn}$  for both devices.

#### 4. Conclusion

To realize in-situ calibration of radon instruments and the precise measurement of thoron gas, a new measurement device of radon/thoron activity concentration based on gas direct detection is developed.

The detection efficiency and the crosstalk factor are determined by simulation, and comparison experiments were carried out in a pure radon/thoron environment for verification. Results show that the direct measurement device and the reference monitor agree well with each other with an average relative deviation of 0.48% for radon from about  $3300~{\rm Bq/m^3}$  to  $38~{\rm kBq/m^3}$  and an average relative deviation of -3.25% for thoron from nearly 25  ${\rm kBq/m^3}$  to 70  ${\rm kBq/m^3}$ . The relative system uncertainty of radon activity concentration is about 6.8%, and that of thoron is nearly 7.3%. The most important source of the system uncertainty is the uncertainty of inner chamber volume, which could be lowered by using a more precise dimension measurement method.

Actually, for gas direct measurement, the sensitivity is limited due to the small volume of the inner chamber but could be enlarged by using bigger detectors and cumulative measurement for a long time in consideration of the high stability of the measurement system, which might be the future development direction. However, this new-designed device is already a good choice for field calibration and reference transfer of radon and thoron activity concentration.

## CRediT authorship contribution statement

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

#### Data availability

The authors are unable or have chosen not to specify which data has been used.

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