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Observation and analysis of atmospheric radon in Qingdao, China

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Abstract
To investigate the levels and behaviours of the atmospheric radon concentration in Qingdao, a continuous measurement was carried out and recorded hourly over a three-year period from September 2006 to August 2009. Levels and variations were studied on the basis of 16 817 data points, and the trends of diurnal and seasonal variations were also analysed. The average concentration of atmospheric radon over the three years was 5.00 ± 3.01 Bq m^{-3}. The average diurnal pattern of radon concentration showed that the daily maximum appears in the early morning, and the daily minimum in the late afternoon, which is driven by the atmospheric stability. The annual pattern features a maximum around December and a minimum around June, which correlates with the origin of air mass brought by the monsoon.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Radon (^{222}Rn) is a naturally occurring gaseous radioactive decay product of the radium isotope ^{226}Ra, which is present in all terrestrial materials. As radon is the only naturally radioactive gas, it is necessary and important to study the levels and behaviours of atmospheric radon. At the same time, as a radioactive inert gas, radon is relatively chemically stable and can be sensitively detected. Besides, ^{222}Rn has a suitable half-life (3.825 days) to be used as a tracer for regional transport in the atmosphere, while the half-time of other isotopes of radon is too short. The study of radon concentration in the atmosphere can give information on the state of turbulence and the stability of the lower atmosphere and can also highlight the movements and origin of air masses. Studies on atmospheric radon as an environmental tracer gas have been reported in recent years (Iida *et al* 1996, Zahorowski *et al* 2005, Sesana *et al* 2003, Hirao *et al* 2008, Chan *et al* 2010).

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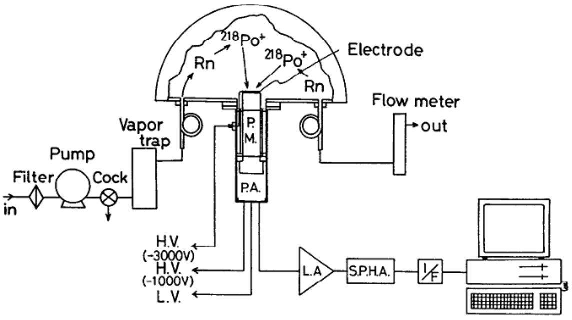

Figure 1. The electrostatic radon monitor (ERM).

In China, national surveys on natural background radiation have been performed, and many local investigations on atmospheric radon have been reported previously (Pan and Guo 1992, Cheng *et al* 2002); however, nearly all the measurements were carried out by grab sampling. Considering the variation of atmospheric radon concentrations, a continuous measurement is essential for the evaluation of atmospheric radon.

A project named Measurement of Radon as a Tracer of Air Pollutants in East Asia started several years ago, and measurements of the atmospheric radon concentrations at various locations in East Asia have already been carried out. As a member of the project, our laboratory put an electrostatic radon monitor (ERM) into use in Beijing in 2003, the results of which have been published (Zhang *et al* 2004, 2009), and another ERM in Qingdao since 2006. This paper reports the observed results in Qingdao.

2. Instrument and method

2.1. Observation site

Atmospheric radon concentrations have been continuously measured in Qingdao Institute of Marine Geology (36.43°N, 120.23°E). The instrument is placed about 15 m high from the ground, and there are no tall buildings in the close vicinity.

Qingdao city is located in the south of the Shandong Peninsula, surrounded by sea on three sides; therefore, the climate of Qingdao is typically dominated by the ocean monsoon. In summer, the maritime air stream keeps the region warm and humid, while in winter, the air mass from the Asian continent keeps the region cold and dry.

2.2. Continuous radon measurement instrument

Atmospheric radon concentrations were sampled and measured with the electrostatic radon monitor (ERM-B1), developed by the Iida group (illustrated in figure 1) (Iida *et al* 1996).

Outdoor air was pumped continuously into the monitor and filtered with a cellulose nitrate membrane filter (pore size 0.8 μm) to remove aerosols and atmospheric radon decay products, and then dried with P_2O_5 powder before entering a 16.8 l hemispheric chamber. Radon decays inside the chamber and positive ^{218}Po ions were collected electrostatically on the electrode of aluminised Mylar coated with a ZnS(Ag) scintillator. The scintillations due to alpha particles were detected by a photomultiplier tube. The scintillation pulse, after being amplified and processed, was then fed into a computer. Radon concentrations were calculated automatically from the accumulated alpha counts every hour. The flow rate was around 1 l min^{-1}, and the background of the monitor was 8.91 $\pm$ 2.81 counts per hour (cph), so the lower detector limit was calculated to be 0.48 Bq m^{-3} (with two standard deviations).

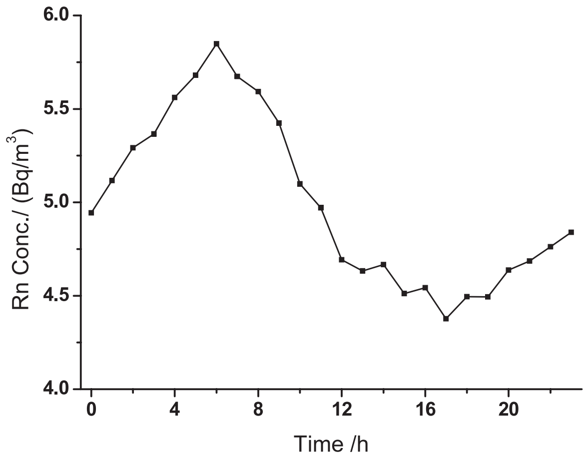

Figure 2. Diurnal variations of atmospheric radon in Qingdao.

The instrument was calibrated by the National Institute of Metrology. The calibration coefficient is 8.26×10^{-3} (Bq m^{-3})/cph.

3. Results and discussion

Atmospheric radon concentrations were continuously measured from September 2006 to August 2009, and recorded every hour. Some of the data were absent due to malfunctions; the effective data were 62.2% over the whole period. The missing data occurred in all seasons and could have little influence on the general trend.

3.1. General trend

Over the three-year period, 16 817 effective data points were obtained. The maximum atmospheric radon concentration was 28.35 Bq m^{-3}, and the minimum was 0 Bq m^{-3} (below the lower limit of detection). The average concentration during the three years from 2006 to 2009 in the Qingdao area was 5.00 ± 3.01 Bq m^{-3} (arithmetic mean), which was much lower than the average world concentration (10 Bq m^{-3}) (UNSCEAR 2000). This is because of the maritime monsoon climate of Qingdao, and the result is close to the other coastal monitoring sites in the world (Zahorowski *et al* 2005, Chan *et al* 2010). Note that the annual average concentration in Qingdao is lower than that in Hong Kong (about half of that in Hong Kong), which can be attributed to two factors: one is because in Hong Kong and adjacent areas the earth's crust is mainly made up of granite, which is abundant in uranium and radium, the other is related to the measuring height: the Qingdao data is measured about 15 m above the ground, while the Hong Kong detector is placed only 1.3 m above the ground, which is nearer to where radon is emanated.

3.2. Diurnal variation

The average concentrations at each hour of the day were calculated to evaluate diurnal variation; 24 data points on the average day were obtained to show the diurnal variation trend. The result is shown in figure 2. It can be observed that there was a minimum in the late afternoon (around 17:00) of each day followed by a maximum in the early hours of the morning (around 6:00), and the amplitude of variation was about 1.5 Bq m^{-3}.

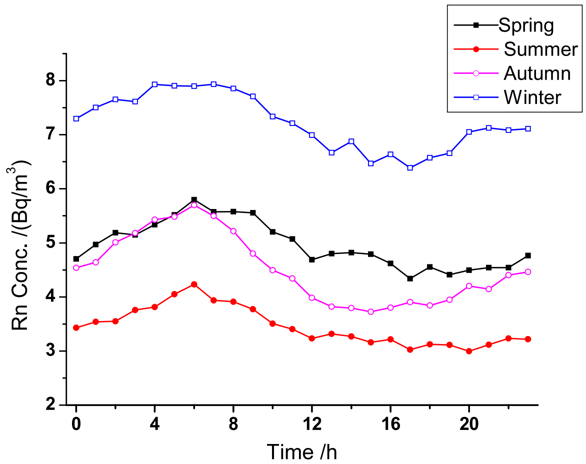

Figure 3. Diurnal variation of each season in Qingdao.

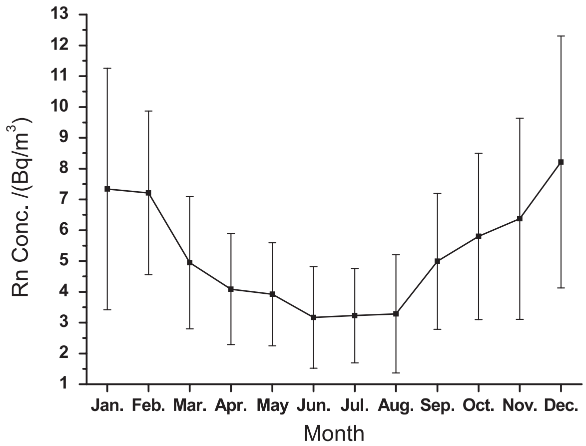

Figure 4. Monthly average concentration of atmospheric radon in Qingdao.

The diurnal cycle has been well described and generally accepted (Sesana *et al* 2003, Chan *et al* 2010). It is attributed to the change of the mixing depth, which is driven by the atmospheric stability. The nocturnal accumulation is due to a low mixing layer caused by a temperature inversion at ground level. When the sun rises in the morning and heats the ground, the inversion is destroyed and remixing of radon takes place in increasingly higher layers with a consequent decrease in concentration. The minimum concentration is normally reached in the late afternoon, in correspondence with the maximum extension of the mixing layer.

Diurnal variation of each season was also observed, which is shown in figure 3. It appears that the diurnal variation pattern of each season was quite variable, and the variational ranges were quite different in different seasons.

3.3. Seasonal variation

The average concentration for each month in the three-year period, with standard deviation, is shown in figure 4. It seems that in Qingdao the maximum of atmospheric radon concentration appeared in December (8.21 ± 4.09 Bq m^{-3}), and the minimum appeared in June (3.17 ± 1.65 Bq m^{-3}).

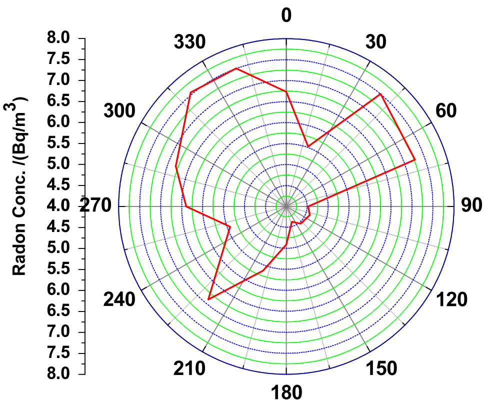

Figure 5. The mean radon concentrations from different wind directions in Qingdao.

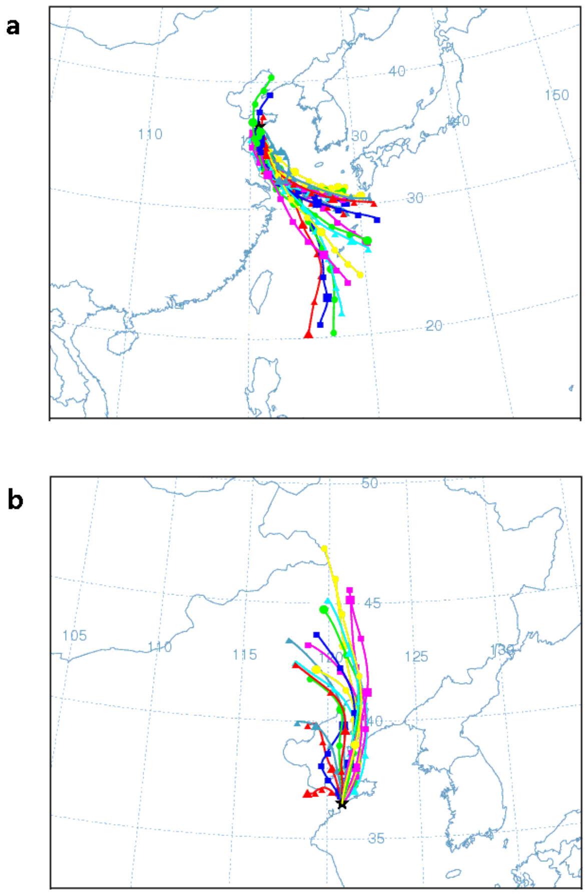

Figure 6. 24 h backwards trajectories every 6 h in six successive days typical of June (a) and December (b).

This seasonal variation correlates with the winter/summer Asian monsoon: air mass fetch is predominantly land fetch in winter, yielding high radon concentrations, and oceanic fetch in summer, yielding low radon concentrations. Figure 5 shows that the mean radon concentration varies with the wind direction in the three-year period, from which it can be obviously seen that the radon concentration correlates closely with the wind direction.

With the help of the HYSPLIT model developed by NOAA, the typical backwards trajectories of air mass are made in June (figure 6(a)) and December (figure 6(b)), respectively: in both months the trajectories are traced back 24 h every 6 h in successive six days. It can be clearly observed that in June the air mass mainly travels over the Yellow Sea, while in December the air mass is dominated by continental origin.

4. Conclusion

A continuous measurement of the atmospheric radon concentration in the Qingdao area was carried out from 2006 to 2009. The three-year average concentration was 5.00 ± 3.01 Bq m^{-3}, which is typical for coastal regions. The diurnal variation tends to be at a maximum in the early morning and at a minimum in the late afternoon, which is closely related to the atmospheric stability. With respect to seasonal variation, monthly average concentrations vary between a maximum in winter and a minimum in summer, which correlates with the winter/summer Asian monsoon.

Further analysis of the correlation between radon concentration and important meteorological conditions is necessary for a better understanding of the levels and variation of atmospheric radon concentrations.

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