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RADON EXHALATION FROM SOIL AND ITS DEPENDENCE FROM ENVIRONMENTAL PARAMETERS

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An automatic measuring apparatus called exhalometer for measurement of the radon exhalation rate from soil is introduced. It consists of a pneumatic driven accumulation chamber with an open bottom, a PC-based control system, six Lucas cells for radon measurement and sensors for environmental parameters. It allows moving the accumulation chamber and hereby opening or closing it. The exhalation rate is determined through the increase of radon in the accumulation chamber. For studying exhalation and the affecting factors, the exhalometer was placed at an undisturbed meadow for the entire year of 2015. The daily radon exhalation rate ranges from 2.5 to 50.7 Bq m^{-2} h^{-1} with an average of 25.3 Bq m^{-2} h^{-1}. The exhalation rate shows daily and seasonal variations with its maximum in the afternoon and in spring. The dependence on several environmental parameters is discussed. The stable performance indicates the system's fitness for long-term measurements.

INTRODUCTION

Radon (^{222}Rn) is a radioactive gas and is ubiquitous in the environment, which contributes over half the total radiation exposure dose from natural sources[(1)]. It is the gaseous progeny of the radionuclide ^{226}Ra commonly found in soil and the exhalation from the soil is a major source of airborne radon. Due to its radioactivity and chemical inertness, radon can be used as a tracer for atmospheric transport study[(2, 3)] and is also applied to geological studies[(4)].

The exhalation of radon and the effects of environmental factors have received considerable attention in recent years[(5–9)]. However, most of these studies are based on random sampling and temporal measurement data, which are insufficient to clarify the relationship between exhalation and environmental factors. This is also the reason why some different studies come to contradictory conclusions about the influence of individual parameters on the exhalation. A long-term continuous measurement data is needed. In addition, among these studies, the accumulation method is prevalent for exhalation measurement. But the exhalation rate may be affected by the measuring device[(10)]. The use of an accumulation chamber prevents soil surface from precipitation, convection or wind induced effects, and thereby affects the conditions of the soil surface.

In this work, an automatic measuring apparatus called exhalometer was developed to solve these problems. The accumulation chamber is moved away from sampling site once the sampling is over, hereby the soil interference caused by the measuring system is minimized. Using this apparatus a long-term measurement was carried out. The variation of exhalation and its relation with environmental parameters are investigated.

MATERIALS AND METHODS

Experimental set-up

The experiment was conducted on an undisturbed meadow at the campus of HMGU in Munich–Neuherberg. The site and the outdoor installation of the exhalometer are shown in Figure 1.

The exhalometer consists of an accumulation chamber, a PC-based control system, six Lucas cells and sensors for environmental parameters. The accumulation chamber is a cylindrical hood attached to a pneumatically controlled arm. It is made of stainless steel and has a diameter of 40 cm and a height of 35 cm. Controlled by the computer program, the accumulation chamber can be lifted up and moved to another place off the sampling site. On the sampling site, a ring groove is embedded ~5 cm into the soil. During sampling the accumulation chamber is placed on this ring precisely and the groove is filled with water so as to isolate air in the hood from outside. The accumulation chamber is connected to Lucas cells through Viton tubing. The volume of each cell is 0.5 L.

Various sensors are placed around the sampling site in an attempt to record meteorological parameters (temperature, humidity, wind velocity, wind direction and precipitation) and soil parameters (soil temperature with depth of 15 , 20, 60 and 100 cm, and soil humidity).

Figure 1. Exhalometer set-up in operation.

Measurement cycle

The measurement process can be adjusted by setting the measurement parameters through a PC. In this study, the measurement cycle is set to be 4 h. At the beginning of a cycle, the hood is placed for collection on the ring by the pneumatically controlled arm. The six Lucas cells are sequentially filled with test gas from the enclosed hood. After a collection period of 1 h, the hood is lifted and moved aside. Soil and vegetation in the collection site then have 3 h to get back into balance with the ambient conditions. In this way, the interference of soil surface induced by the accumulation chamber is minimized, which makes long-term measurement feasible. Thereafter, the operation is repeated continuously. During the collection, six Lucas cells are sequentially filled with test gas (3 min filling time at 3 L/min) and then counting the alpha-decays caused by radon and its progenies for 1 h. Shortly after that, Lucas cells are flushed with purge gas and later on background counts are recorded.

Evaluation algorithm

As soon as the hood sits on the groove ring above the sampling area, radon exhaled from soil starts to accumulate inside. This will contribute to an increase of the radioactive counts. The evolution of radon in the accumulation chamber can be described as follows[11]:

$$\frac{\mathrm{d}C(t)}{\mathrm{d}t} = \frac{EA}{V} - \lambda_{\mathrm{eff}} C(t) \tag{1}$$

where, C (Bq m^{-3}) is the radon concentration in the chamber; E (Bq m^{-2} h^{-1}) is the exhalation rate; A (m^2) is the area of the chamber bottom; V (m^3) is the volume of the chamber; and λ_{eff} (h^{-1}) is an effective decay constant of radon, which contains decay constant, back diffusion coefficient and leakage coefficient. The solution of equation (1) is as follows:

$$C(t) = \frac{EA}{V\lambda_{\mathrm{eff}}}(1 - \exp(-\lambda_{\mathrm{eff}} t)) + C_0 \exp(-\lambda_{\mathrm{eff}} t) \tag{2}$$

In this study, a short accumulation time (1 h) was adopted as suggested by Petropoulos *et al.*[12], thereby the radon concentration in the chamber can keep a low level and the back diffusion effect is negligible. Leakage of this system was assumed to be insignificant because of the short sampling period[13]. Taylor expand equation (2), it can be rewritten as follows:

$$\mathrm{C(t)} = \frac{\mathrm{EA}}{V\lambda_{\mathrm{eff}}}(\lambda_{\mathrm{eff}}\mathrm{t} - O((-\lambda_{\mathrm{eff}}\mathrm{t})^2)) + \mathrm{C}_0(1 - \lambda_{\mathrm{eff}}\mathrm{t} + O((-\lambda_{\mathrm{eff}}\mathrm{t})^2)) \tag{3}$$

$O((-\lambda_{\mathrm{eff}}\mathrm{t})^2)$ is the Taylor expansion remainder term, which is in the same order of $(-\lambda_{\mathrm{eff}}\mathrm{t})^2$. When the accumulation time is short, which means $\lambda_{\mathrm{eff}}\mathrm{t} \ll 1$, the remainder term $O((-\lambda_{\mathrm{eff}}\mathrm{t})^2)$ can be omitted. Thus equation (3) can be written as follows:

$$\mathrm{C(t)} = \frac{\mathrm{EA}}{\mathrm{V}}\mathrm{t} + \mathrm{C}_0(1 - \lambda_{\mathrm{eff}}\mathrm{t}) \tag{4}$$

Besides considering the low initial radon concentration, which means $\mathrm{EA/V} \gg \mathrm{C}_0\lambda_{\mathrm{eff}}$, finally equation (4) can be written as follows:

$$C(t) = C_0 + \frac{EA}{V}t \tag{5}$$

The six Lucas cells measure the radon concentration at six time points. Through the increase of the radon concentration with time, the radon exhalation rate, E, can be deduced.

In the data analysis, two kinds of abnormal data were screened out:

- Negative radon concentration: when the background count was higher than the total count, the net counts and also the measured concentration would be negative. If this situation happened only to the first Lucas cell, the first radon concentration would be sorted out. The remaining radon concentrations are still valid for linear fitting. At the very beginning of the measurement cycle the radon concentration is very small. Then it is possible that the background count is larger than the total count due to statistical fluctuation. If the situation happened to other Lucas cells, the whole measurement cycle was rejected. Prolongation of the sampling period (which will cause other problems) or improving the sensitivity of the Lucas cells might reduce the probability of the occurrence of negative results.

- Negative exhalation rate: at short accumulation times, the back diffusion effect is negligible. The concentration of the actual Lucas cell should be higher than that of previous Lucas cell. Then it is unlikely to obtain a negative exhalation rate. In this case, the entire measure cycle data was rejected. This happens when the accumulation chamber sits on a wrong place during sampling time or the groove ring runs out of water thereby the chamber is not airtight.

Calibration

Calibration of Lucas cell was performed with a secondary standard (PTB calibrated AlphaGuard, Saphymo Inc., Germany). A uraninite source and the AlphaGuard were put into a stainless steel container. Then the container was closed for more than 3 weeks to achieve an equilibrium state. The radon concentration was ~1.6 kBq m^{-3}. Later the container was connected to the experimental set-up via tubes as in the final installation. The air in the container was drawn into the Lucas cells and then sent back to the container for 3 min at 3 L/min by a pump as in the ordinary experimental condition.

Comparing the radon concentration measured by AlphaGuard and the count measured by Lucas cells, the efficiency of Lucas cells can be expressed as follows:

$$\varepsilon_{\mathrm{i}} = \frac{N_{i\ \mathrm{net}}}{\alpha(t) * V_{\mathrm{L}} * C_{\mathrm{AlphaGuard}} * t} \quad (6)$$

where ε_{i} (i = 1, 2, 3, 4, 5, 6) is the efficiency of the six Lucas cells respectively; $N_{i\ \mathrm{net}}$ is net count of each of the six Lucas cells; V_{L} is the volume of Lucas cell; $C_{\mathrm{AlphaGuard}}$ is the radon concentration measured by the AlphaGuard and t is the counting time. $\alpha(t)$ is the number of alpha emitters present in the cell per Becquerel of radon during a certain counting period.

RESULTS AND DISCUSSION

Efficiency calibration

After connecting the calibration container to the Lucas cells, there is a decrease of the radon concentration for a few days but will eventually reach an equilibrium state. Only data during this period was chosen for efficiency calibration. The results are summarized in Table 1.

The final radon concentration was around 1.6 kBq m^{-3} with small fluctuations. Under such radioactive level, as Table 1 shows, the radioactive counts were around 4500 for 1-h measurement. Overall, 22 sections of experimental data during the equilibrium period were used for the calibration and the standard deviation was <3%.

Daily variation

Some studies have reported that airborne radon concentration is subjected to daily variation: high concentration in the early morning and low concentration in the afternoon[14, 15]. As mentioned earlier, soil exhalation is the most important source of airborne radon. It is expected to find some similar daily variation pattern of radon exhalation from the soil. However, the exhalation rate showed a contrary tendency compared to airborne radon concentration. As illustrated in Figure 2, exhalation rate is lowest in the early morning, then increases slowly and reaches maximum value in the afternoon and then decreases during the night. During nights the temperature is low, air and also radon accumulate near the ground level and form a thermal inversion layer. Thus, radon concentration above the soil surface increases during the night. On one side, this will reduce the radon gradient between soil and air. Thereby, the diffusion of radon from soil to atmosphere is suppressed. On the other side, the thermal inversion layer near the ground reduces air convection between soil and atmosphere, which also suppress radon escape

Table 1. Efficiency calibration of the Lucas cells.

Cell no.	Count rate (h^{-1})	Rn conc. (Bq m^{-3})	Efficiency	Standard deviation
1	4643	1599	0.76	0.026
2	4676	1599	0.77	0.028
3	4644	1599	0.76	0.029
4	4490	1599	0.74	0.020
5	4583	1599	0.75	0.023
6	4506	1599	0.74	0.023

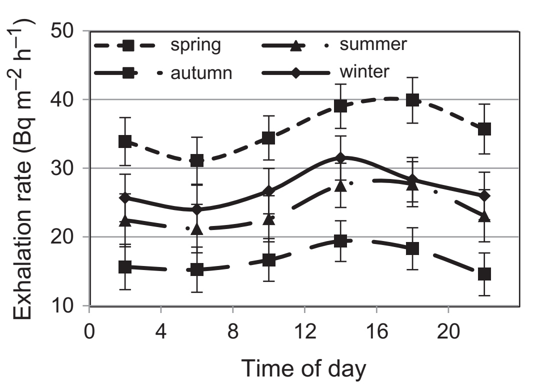

Figure 2. Daily variation of radon exhalation rate. Time is adjusted to GMT + 1.

from soil to atmosphere. After sun rising, temperature rises and air turbulence enhances. Air and radon near ground rise up to upper atmospheres. Airborne radon near ground decreases and radon gradient increases, which promotes radon flux.

Seasonal variation

In general, the exhalometer works well. Figure 3 shows the daily average and monthly average radon exhalation rate of 2015. Some data were missing due to maintenance break. In total, only 29 days data was missing. As shown in Figure 3, even though the data varied from day to day dramatically, it is apparent that radon exhalation rate in summer and autumn is lower than that in winter and spring. The daily exhalation rate varied from 2.5 to 50.7 Bq m^{-2} h^{-1} with an average of 25.3 Bq m^{-2} h^{-1}. The highest and lowest monthly exhalation rates were 37.5 Bq m^{-2} h^{-1} in March and 14.5 Bq m^{-2} h^{-1} in October, respectively.

Correlation with environmental parameters

As aforementioned, environmental parameters such as air temperature, soil temperature, air humidity and precipitation were recorded.

Data suggest that air humidity had a weak negative correlation with exhalation rate. The correlation coefficient was −0.24.

The overall correlation coefficient between exhalation and air temperature is only −0.04, which means it is nearly uncorrelated. But soil temperature showed a moderate negative correlation coefficient, and the temperature of deeper soil layers, which is smoother and more stable, has an even stronger correlation. The correlation between soil temperature at the 100 cm depth and exhalation rate can be as high as −0.35.

Some other authors have pointed out that the temperature has a positive effect on the exhalation process[6, 16]. The exhalation is also influenced by other environmental parameters, e.g. the soil humidity. Therefore, it is interesting to test for the effect of temperature at stable soil humidity, instead of the overall correlation.

The measurements at 9% soil relative humidity were selected. The results show that the correlation coefficient between exhalation rate and air temperature and soil temperature of 20, 60, 100 cm depth are 0.46, 0.28, 0.03 and −0.08, respectively. It can be explained by a faster diffusion of radon atoms at higher temperature. The influence of the upper soil layer which warms up first seems to be more important.

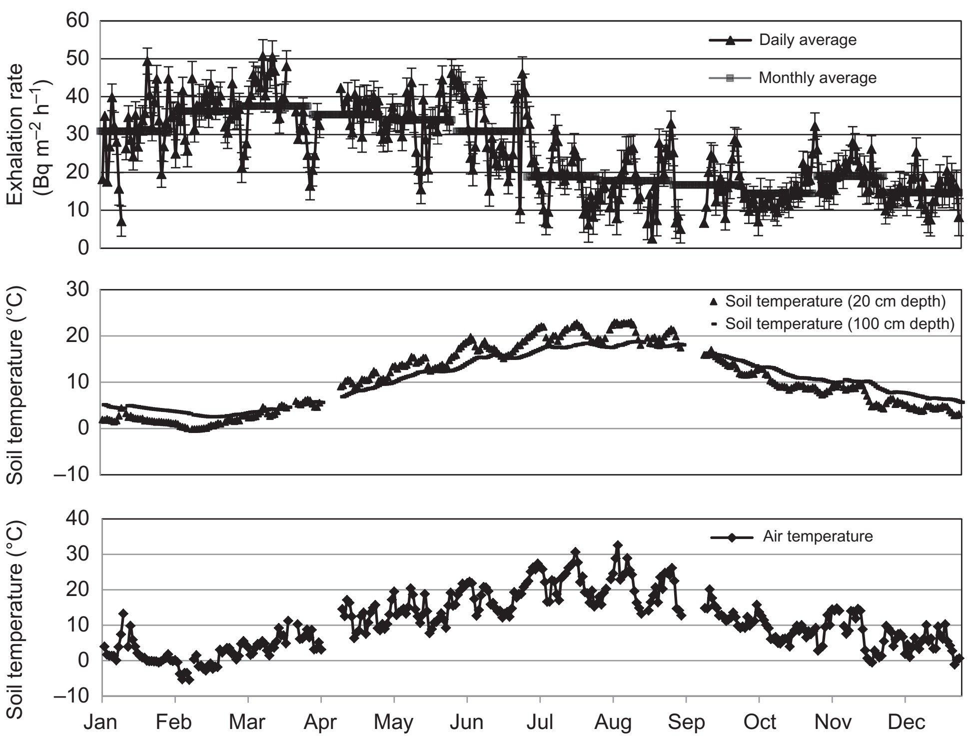

Figure 3. Radon exhalation rate, soil temperature at different depths and air temperature in 2015.

Due to the different results of the temperature effects on exhalation between general and stable soil humidity condition, it is supposed the soil humidity plays an important role in it and to a certain extent conceals the influence of temperature.

CONCLUSION

The exhalometer is an improvement of the traditional accumulation method. It helps to avoid the soil surface to suffer from the interference induced by the accumulation chamber. The performance indicates that it is suitable for long-term measurement. The system is compact in order to be used at different sites.

The daily average radon exhalation rate varied from 2.5 to 50.7 Bq m^{-2} h^{-1}. Exhalation rate is high in the afternoon and low in the early morning. Exhalation rate in spring and winter is higher than in summer and autumn.

Air temperature showed a moderate negative correlation with exhalation, while air humidity and surface soil temperature showed a weak negative correlation. The soil humidity also played an important role. However, these findings are associations observed at an individual sampling site. More data at different sites are necessary for further generalization. In the future, other environmental parameters such as soil humidity, air pressure and precipitation should also be taken into consideration. Further investigations are needed to describe thoroughly the mechanism how the environmental parameters affect the radon exhalation rate.

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