RADON AND ITS PROGENY IN OUTDOOR AIR

S. J. Hu and K. S. Tan

Physics Department, Faculty of Science
Universiti Brunei Darussalam

ABSTRACT

The diurnal variations of the outdoor ^{222}Rn and its progeny were measured over a period of four months starting from December 1998 till March 1999 in Bandar Seri Begawan, Brunei Darussalam. The set-up for ^{222}Rn measurement consists of an air filter pump, a ZnS detector that detect the alpha particles emitted, a photomultiplier tube and a counter. Both the morning and the afternoon data show that the mean radon activities for December and January are about 1.5 times higher than those of February and March. Also, the morning radon activities of all the four months are observed to be about twice those of the afternoon. A qualitative explanation based on the local meteorological conditions is given to account for the diurnal variations observed.

1. INTRODUCTION

In spite of the potential radiation hazards from the nuclear industry, nature is still by far the major contributor to the total collective radiation exposure of the world's population. The average radium concentration in soil is 40Bqkg^{-1} and it can reach values of hundreds¹ Bqkg^{-1} . Typical radon concentration in soil gas range from 10000Bqm^{-3} to 50000Bqm^{-3} . As the natural radionuclides in outdoor air are predominantly radon (^{222}Rn) and its short-lived decay products, the radon and its progeny represent a potential hazard to health.

Although the main source of radon is soil, several meteorological variables affect the radon concentration in outdoor air. A quite high radon concentration in the outdoor air has been reported in the case of particular weather condition such as very low precipitation². The potential for radon exhalation from the ground depends mainly on the activity level of ^{222}Rn in the subsoil and its permeability. After exhalation from the soil the gas ^{222}Rn and its short-lived decay products are transported vertically, mixed in the boundary layer, and dispersed horizontally by the wind. The decay products of radon may be removed from air by dry and wet deposition processes. In these processes, humidity of the air and rainfall play an important role.

The decay series of ^{222}Rn ends with stable lead isotope ^{206}Pb . However, as far as airborne ^{222}Rn progeny is concerned, the isotope ^{210}Pb with a half-life of 22.3 years is usually considered the meta-stable end product. All intermediate decay products are heavy metals,

which tend to attach to aerosols or to plate out onto surface areas. The airborne progeny is thus, in general, not in secular equilibrium with its parent.

The equilibrium-equivalent decay product concentration (EEDC), defined as that ^{222}Rn concentration which in secular equilibrium with all short-lived decay products, represents the same potential alpha energy as the mixture actually present. Due to physical processes like recoil, attachment to aerosols, and deposition of attached and unattached nuclides, ^{222}Rn and its decay products are seldom in secular equilibrium. The equilibrium factor E_p , defined as the quotient of the EEDC and the activity concentration a_0 of ^{222}Rn , i.e., $E_p = \text{EEDC}/a_0$, being less than 1 for non-equilibrium condition is rather typical for outdoor environment.

In recent years much attention have been given to the risks of prolonged exposure to low levels radon concentration and the exposure of the general population to radon in dwellings^{3,4}. The natural background radiation being very much influenced by the different environmental parameters is also not at a stable level. As the outdoor concentration of ^{222}Rn and its progeny may vary between virtually zero and levels that exceed the average indoor concentration, this study was carried out with the objective of monitoring the diurnal variations of the outdoor ^{222}Rn and its progeny in Bandar Seri Begawan, Brunei Darussalam. A qualitative explanation based on the local meteorological condition is given to account for the diurnal variation of the radon activities data obtained. However, no attempt is made in this study to correlate the radon concentration with the soil types. This is because in spite of the fact that the production of the radon isotopes and the exhalation rates of the radon do vary according to the soil types, the direct correlation between soil type and the ^{222}Rn concentration in air is not so obvious if one considers the atmospheric transport of ^{222}Rn where ^{222}Rn can be transported over considerable distances and be mingled with air masses with very different ^{222}Rn concentration, thereby completely losing all correlation with a specific soil type.

2. PROCEDURE FOR MEASURING RADON-222

The diurnal variations of the outdoor ^{222}Rn and its progeny were measured over a period of four months starting from December 1998 till March 1999. These four months were especially interesting for studying the diurnal variation of outdoor radon concentration in Brunei Darussalam. This is because December and January are generally the wet months here with lots of rainfall while February and March are generally very dry with little rainfall. The samplings and measurements were taken in the morning as well as in the afternoon. The morning samplings were generally carried out between 8am and 9am while the afternoon samplings were generally taken between 2pm and 3pm.

The set-up for ^{222}Rn measurement consists of an air filter pump, a ZnS detector that detect the alpha particles emitted, a photomultiplier tube (OKEN SP-20) and a counter (OKEN RC-101A). The system was calibrated with a known sample to give readings in the Bqm^{-3} activity units. The calibration was performed at the Department of Nuclear Sciences, University of Nagoya, Japan. The calibration graph obtained is as shown in

Figure 1. In the calibration curve, the activity is plotted against the counts per liter per minute. The counts per liter per minute data were obtained by taking the difference between the 40 minutes timer counts and the background counts.

Figure 1. Calibration curve for counts to activity.

The air was sampled at the rooftop of the Faculty of Science building of Universiti Brunei Darussalam, which is at a height of about 15 m above the ground level. The sampling was done by pumping the air through a piece of filter paper of pore size 1.2 μ m for 15 minutes as determined by the calibration procedure. The pump rate was set at 33 liter per minute. The filter was then inserted into a holder facing the ZnS detector and the counter set to count the emission of the α -particle emitted by the decay of ²²²Rn. The activity was read off from the calibration curve obtained.

3. RESULTS AND DISCUSSION

Figure 2 shows the morning and afternoon diurnal variation of the radon activity in Bqm⁻³ from December 1998 to March 1999. Diurnal variation is observed at both the morning and afternoon radon activities. Morning data appear to fluctuate more compared

Figure 2. Diurnal variation of radon activity from December 1998 to March 1999.

Table 1: Monthly mean readings of radon activity.

Month	Morning reading, Bqm⁻³	Afternoon reading, Bqm⁻³
December 1998	1.41 ± 0.01	0.62 ± 0.01
January 1999	1.30 ± 0.01	0.67 ± 0.01
February 1999	0.84 ± 0.01	0.47 ± 0.01
March 1999	0.86 ± 0.01	0.40 ± 0.01

to the afternoon data. Table 1 gives the morning and afternoon monthly mean of the activity. In general, the morning radon activities of all the four months are about twice those of the afternoon. Both the morning and afternoon data show that the mean radon activities for December and January are higher than the mean radon activities of February and March. The December mean reading is highest (1.41 Bqm⁻³). However this value is still low compared to the average level of radon gas concentration in the atmosphere at ground level in the USA, usually assumed to be in the range⁵ of 4-15 Bqm⁻³. In fact, measurement that is more recent seems to indicate even higher values^{2,6,7,8,9,10}, reaching some tens of Bqm⁻³. December and January data are about 1.5 times higher than those of February and March. This is probably due to the more frequent rainfall that occurred during the rainy months of December and January.

After exhalation from the soil, the ²²²Rn gas and its progeny are transported horizontally with the prevailing winds and vertically within the mixing layer by turbulent processes. Blaauboer and Smetsers¹¹ reported in 1996 that during summer as much as 20% of the ²²²Rn is transported to heights above 5.5km, while 50% is transported above the boundary or mixing layer. In atmospheric air, the short-lived decays products of ²²²Rn rapidly attach to aerosols. Cloud droplets form through condensation around these aerosols. During rainy periods the amount of water in the lower atmosphere increases, thus the number of droplets formed during condensation also increases. The concentration of attached radon decay products in air is therefore expected to rise with humidity. Thus the ²²²Rn and its decay products are in a better equilibrium at higher humidity, thereby raising the EEDC for a constant ²²²Rn concentration, the equilibrium factor E_p and the total radon activity.

Wet deposition is often considered being proportional to the amount of rain that has fallen. The processes that are likely to occur to ²²²Rn and its decay products during rainfall is that the radon gas may dissolve in cloud droplets. After decay, its decay products may then be deposited on the ground during rainfall. The decay products may attach themselves to the aerosols that serve as condensation nuclei for cloud drops or they may be scavenged and swept from the air by falling raindrops. Although not much of the ²²²Rn will be washed out due to its low solubility in water, the decay products' activity fixed in the clouds is washed out during rainfall. As heavy downpour is rather common during the months of December and January, this explains why the mean radon activities for the rainy months of December and January are higher than the mean radon activities for February and March.

The morning activities being higher than those of the afternoon is possibly due to the temperature inversion that is commonly developed at night, which prevents exchange with higher air masses. The surface air directly above the soil at night are warm after being under the tropical sun the whole day while the soil underneath the air cools through back radiation. This helps to develop a temperature inversion very near the surface that inhibits vertical mixing largely at night as well as in the early morning hours. The temperature inversions at night near the ground surface would result in the built-up of radon and its decay products, giving higher radon activities. At later half of the day, i.e., after mid-day, when the sun warms the soil again, this inversion will disappear quickly, after which radon and its decay products will be mixed over greater vertical distances. This possibly explains why the afternoon readings are generally lower compared to those in the morning.

Higher radon emanation rate in the morning possibly also accounts partly for the higher activities morning data. When radium decays, the radon isotopes will be trapped in the soil particles. The interstitial radon in the upper soil layer will reach the atmosphere through exhalation. The rate of exhalation of radon from soil to the atmosphere is dependent on the emanation factor that increases with the moisture content of the soil. The temperature drop at night causes condensation in the soil. The interstitial water due to condensation will absorb the radon atoms recoiling from the soil particle and due to the low solubility of radon in water, the radon emanation rate will increase¹², thereby giving rise to higher radioactivity in the morning. In the afternoon when the sun gets very hot and the temperature rises, the water content in the soil reduces, thereby decreasing the emanation rate and giving a lower radioactivity level.

Since Bandar Seri Begawan is a coastal town, another possible reason why the afternoon radon activities are lower than those in the morning is the influence of the sea breeze. The sea breeze is more likely to occur in the later half of the day when low pressure develops over the landmass, which has been under the hot tropical sun the whole morning. Marine air is generally low in radon concentration (about 0.1 Bqm^{-3}) because of the minimum presence of radium in the seawater. Thus during the transport of the marine air masses to the coast region under low pressure in the afternoon, almost no enrichment of ^{222}Rn from soil can take place. The marine air masses that are brought in from the sea will tend to dilute the radon concentration activity of the landmass, giving rise to a lower radon concentration activity in the afternoon.

4. CONCLUSION

In conclusion the mean value of the outdoor radon concentration in Bandar Seri Begawan based on the 4 months data is very low compared to the values adopted by UNSCEAR¹ for which the coastal region value is reported to be somewhat less than the adopted values of $5\text{-}10 \text{ Bqm}^{-3}$ for continent area.

5. ACKNOWLEDGEMENT

The authors would like to thank Dr. S. Minato of NIRIN, Japan for lending the radon monitoring equipment for this work, and Dyg. Gandi Anggas and Azijah Mohamad Nasir for their help in collecting the data.

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