

Soil radon (^{222}Rn) monitoring in a forest site in Fukushima, Japan

Ryoko Fujiyoshi · Misato Ohno · Kazumasa Okamoto · Kikuo Umegaki

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Abstract Soil radon (^{222}Rn) has been monitored since August 2013 at three different soil depths on a campus forest of Fukushima University in Japan, where a large amount of fallout nuclides were released by the accident of Fukushima Daiichi Nuclear Power Plant in March 2011. The primary purpose of this study is to evaluate ^{222}Rn activity level, variability and factors controlling ^{222}Rn concentration in soil air using data obtained from August to December 2013. Time series of ^{222}Rn activity concentration showed depth-dependent variability with an equilibrium value ($^{222}\text{Rn}_{\text{eq}}$) during this observation period; 7.5, 14 and 23 kBq m^{-3} at 0.3, 0.6 and 1.0 m in depth, respectively. Two typhoons passing over the site had a great influence on soil radon level, which was practically used for evaluating effective diffusion coefficient of ^{222}Rn . Transport mechanism of ^{222}Rn in soil air was considered to be diffusion-controlled with data sets on changing ^{222}Rn concentration with time in selected cases that showed decreasing (or increasing) ^{222}Rn concentration with time at every depth. Important factors affecting soil ^{222}Rn variability are meteorological parameters, low-pressure front passing over the site, and subsequent precipitation. Time lags of decreasing ^{222}Rn concentration at different depths after rain indicate a certain relationship of ^{222}Rn level with moving water (and water vapor) in soil. The findings obtained in this study are important to evaluate the fate of fallout nuclides (radiocesium) in contaminated forest sites using soil radon as a tracer of moving soil air.

Keywords ^{222}Rn · Soil air · Monitoring · Gas transportation · Typhoon · Fukushima

Introduction

Radon (^{222}Rn) in the environment has been extensively studied to evaluate dose levels due to inhaling the radioactive gas into the body, to estimate its flux from the ground surface to the atmosphere (exhalation) as a tracer of air movement in the lower atmosphere, and to predict seismic activity in tectonically active areas (Dörr et al. 1983; Zahorowski et al. 2004; Vaupotič et al. 2010). Another important and basic aspect exists in the study of ^{222}Rn in soil air to elucidate mechanisms of its migration in soil and transportation to the ground surface (Nazaroff 1992; Neznal and Neznal 2005; Fujiyoshi et al. 2010, 2013). Soil radon monitoring has shown that ^{222}Rn activity concentration varies to a great extent depending on geological, meteorological and hydrological factors, and that its migration through the soil and rock is controlled not only by diffusion but also by convection (Yakovleva 2005; Perrier and Girault 2013). Monitoring soil radon for several years since 2004 gave the results that a major factor affecting soil ^{222}Rn concentration was soil temperature under high atmospheric pressure region in three seasons, except for winter on a campus forest site of the Hokkaido University, Japan (Fujiyoshi et al. 2005, 2006, 2010). In contrast, lower ^{222}Rn concentration with small variability appeared in soil under persisting snow in winter months from December to March. They further detected a small amount of ^{222}Rn releasing from the ground surface to the overlying snowpack with a mean flux density of $0.4 \text{ mBq m}^{-2} \text{ s}^{-1}$ at their observation site in Hokkaido (Fujiyoshi et al. 2013).

It is widely recognized that both liquid and gaseous water movements are fundamental factors controlling many processes in soil. Soil water dynamics are strongly linked to temperature variations and then biological activities. These processes, complicated due to

R. Fujiyoshi (✉) · M. Ohno · K. Okamoto · K. Umegaki
Faculty of Engineering, Hokkaido University, Kita 13, Nishi 8,
Sapporo 060-8628, Japan
e-mail: fuji@eng.hokudai.ac.jp

interrelations among controlling factors, have not been clarified thoroughly (Wells et al. 2007; Bittelli et al. 2008). Understanding radon transportation in soil is useful for evaluating soil air movement in the surface soil layers, because radon is chemically inert and radioactive.

A site concerned in this study was on the campus forest of the Fukushima University, Fukushima, Japan. Large amounts of radionuclides such as ^{131}I ($T_{1/2} = 8.04\text{d}$), ^{134}Cs ($T_{1/2} = 2.06\text{y}$) and ^{137}Cs ($T_{1/2} = 30.17\text{y}$) were released and deposited in the environment due to the accident of the Fukushima Daiichi Nuclear Power Plant in March 2011 as a consequence of a big earthquake (Great East Japan Earthquake) of magnitude 9.0 and following a 15 m tsunami. Total amounts of ^{131}I , ^{134}Cs and ^{137}Cs released to the atmosphere were estimated by several institutes and universities to be approximately 200, 20 and 20 PBq over March 12–31, 2011, respectively (Science Council of Japan 2012). The majority of the contaminated area (about 70 %) is covered by forests in which a large portion of deposited radiocesium existed in the canopies of coniferous forests, whereas fallen leaves on the ground surface contained most of the radiocesium in deciduous forests as of September, 2011 (Hashimoto et al. 2012). Much effort has been devoted to remove deposited fallout radionuclides in forest areas so far; however, a large part still remains without decontamination. Our previous results showed that most radiocesium deposited on fallen leaves of deciduous trees in March 2011 rapidly moved to the organic layer of the soil surface in November of this year (Hao et al. 2013). The above fact probably resulted from intensive (micro) biological activities in the organic layer in summer. Decomposing organic matter releases CO_2 and other gaseous components into pore spaces in soil. It is therefore important to evaluate potential effects of soil air (and water) movement on the fate of deposited radiocesium in the surface layer of forest soils.

The primary purpose of the present study is to elucidate basic behavior of soil radon (^{222}Rn) including activity level, variability and transportation mechanism(s) in forest soil in Fukushima contaminated with fallout nuclides. Special concern is to clarify effects of two typhoons passing over the observation site on soil radon transportation.

Experimental procedure

Description of the site

Figure 1 shows a location map of our observation site (37.68457N, 140.45347E), a campus forest of the Fukushima University, Fukushima Prefecture, Japan. The site (about 200 m a.s.l.) belongs to the Fukushima Basin which is surrounded by the Azuma Mountain Range in the west and Abukuma Highland in the east. The original sloping

terrain in this area was modified to construct a new university building in the 1980s (Fukushima Prefecture 1982).

It is a typical basin-specific climate of high temperature and high humidity in summer with annual mean temperature and precipitation, 12.8 °C and 1,105 mm, respectively. Annual mean of maximum snow depth in winter is about 8 cm.

There is a great variety of vegetation on the campus of the Fukushima University (Kurosawa et al. 2010). For example, tree species growing at the site are: oaks (*Quercus serrate*, *Quercus acutissima*), pine tree (*pinus densiflora*), chestnut tree (*Castanea crenata*) and Japanese laurel (*Aucuba japonica*).

Soil properties

Several soil properties were measured including porosity, humidity, soil organic matter and pH. Porosity (and humidity) was determined by measuring the weight of a sample in a container of known mass: (1) in the field (bulk weight), (2) after filling it with pure water (water-filled weight) and (3) after complete drying at 100 °C for more than 24 h (dried weight). The amount of soil organic matter was estimated as a difference of a sample weight before and after heating at 500 °C for 2 hours. Soil pH was measured in situ with a pH meter (HI 99121, HANNA INSTRUMENTS, USA), in which the electrode was inserted into the soil at a depth of about 10 cm (Fujiyoshi and Sawamura 2004).

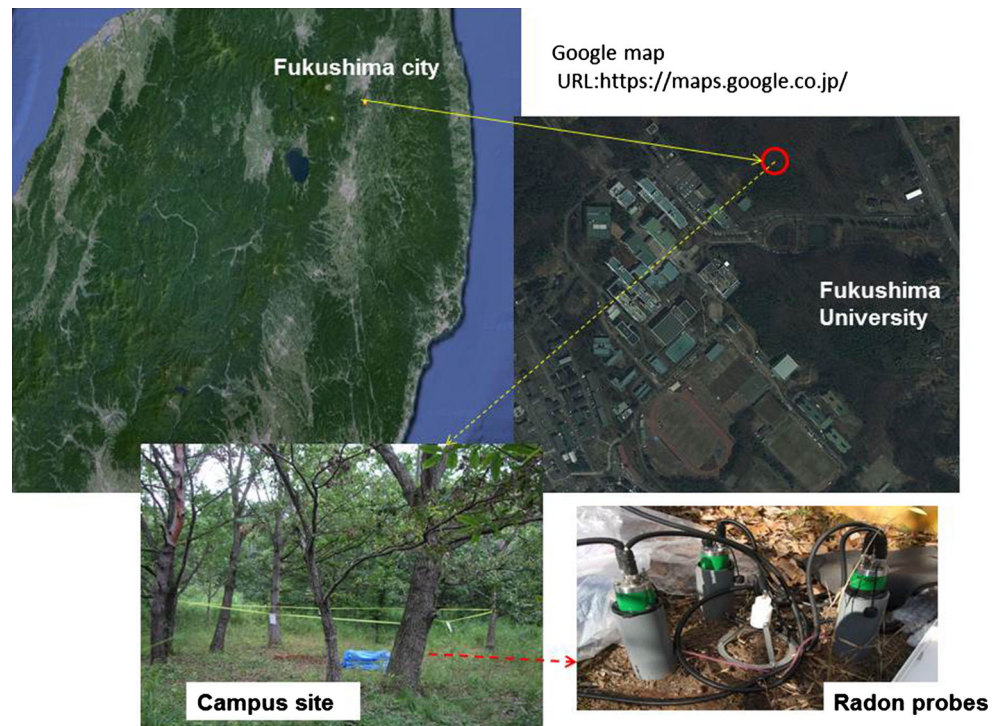
Soil radon (^{222}Rn) measurement

Activity concentration of ^{222}Rn in soil air was monitored at three soil depths with radon probes (VDG, Algade, France), in which silicon detectors of 450 mm² in the total detection area count alpha particles in the 0.7–6.1 MeV energy range, emitted from ^{222}Rn radioactive transformations. The radon probes in PVC housing tubes (0.5, 0.8 and 1.2 m in length) were buried in soil at 0.3, 0.6 and 1.0 m depths on August 21, 2013. Detection efficiency was 51.6, 51.8 and 45.8 Bq m⁻³/(impulse h⁻¹) at 0.3, 0.6 and 1.0 m, respectively. Probes were located close to each other in the ground, which could be assumed to be the same place of a local environment. A data logger stored hourly data on ^{222}Rn activity concentration, barometric pressure and temperature at each depth.

Monitoring differential barometric pressure

Differences in barometric pressure on the surface and at three depths were measured once an hour in the casing tubes of ^{222}Rn probes. The whole system prepared on demand by a private company (North One Co., Ltd.,

Fig. 1 Location map and instrumentation of radon probes in soil on the campus forest of Fukushima University, Fukushima, Japan



Sapporo, Japan) consists of a micro-barometric sensor (JP208, Yokogawa Electric, Japan) and a data logger (KADEC21-U4-C, North One Co., Ltd., Japan).

Gamma spectrometry

Activity concentration of several environmental radionuclides (^{40}K , ^{134}Cs , ^{137}Cs , ^{210}Pb) as well as ^{226}Ra , the parent nuclide of ^{222}Rn , in soil was determined by gamma spectrometry with a HPGe detection system (SEIKO EG&G, Japan). Standard reference materials (IAEA 327 and IAEA 444) were used to evaluate activity concentrations of individual radionuclides from counts obtained with the same geometry under identical operating conditions. Energy and efficiency calibrations were periodically carried out, as well as checking the background. Details of the measurements were described in Fujiyoshi et al. (2010, 2013).

Results and discussion

Figure 1 shows the monitoring point on the campus of the Fukushima University in Fukushima City, Japan, where the areas were contaminated with fallout radionuclides due to the accident of Fukushima Daiichi Nuclear Power Plant on Mar. 11 2011. As of 2013, most campus sites except for the forest parts have already been decontaminated. University

staff members have regularly measured and reported radiation dose rate at 1 m height above the ground at many selected points on the campus (<http://www.fukushima-u.ac.jp/guidance/top/fukudai-housyasen.html>).

Table 1 summarizes some of the soil properties including activity concentration of environmental radionuclides (^{40}K , ^{210}Pb , ^{226}Ra) in our test site where the original hilly landscape was modified to construct a new university building more than 30 years ago. This fact reflects relatively homogeneous distribution of ^{40}K and ^{226}Ra with depth as summarized in the table. Depth distribution profiles of ^{210}Pb , a ^{222}Rn progeny, showed a small surface enrichment, suggesting atmospheric lead deposition on the forest floor since 1981 when the construction of university buildings was finished. It should be noted here that much higher concentration of ^{210}Pb is usually observed in the surface layer of forest soils undisturbed for more than 100 years (Fujiyoshi and Sawamura 2004).

Figure 2 shows the depth distribution profiles of radiocesium (^{134}Cs and ^{137}Cs) activity concentration in soil, in which all the values in the figure were calculated on August 21, 2013. Two and a half years after the accident, activity concentration of short-lived ^{134}Cs ($T_{1/2} = 2.07$ y) was about a half of that of ^{137}Cs ($T_{1/2} = 30.17$ y). Activity concentration of radiocesium present within the surface portion of soil (depth of <5 cm) decreased exponentially with soil depth, thus suggesting no natural and/or anthropogenic intervention since then (Fujiyoshi et al. 2011).

Table 1 Summary of soil properties (humidity, pH, porosity, activity concentrations of ^{40}K , ^{210}Pb and ^{226}Ra) on the campus forest of Fukushima University, Fukushima, Japan

| Depth (cm) | Humidity (%) | pH | ^{40}K (Bq kg ⁻¹) | ^{210}Pb (Bq kg ⁻¹) | ^{226}Ra (Bq kg ⁻¹) | Depth (cm) | Porosity (%) |
|------------|--------------|------|--|--|--|------------|--------------|
| 1 | 30.8 | 4.44 | 212 (5.2) | 49.2 (7.5) | 35.2 (2.8) | 2 | 20.6 |
| 3 | 24.3 | 4.70 | 197 (4.9) | 44.5 (3.7) | 30.7 (2.3) | 5 | 18.9 |
| 5 | 23.7 | 4.88 | 226 (5.5) | 30.4 (2.6) | 39.7 (2.5) | 8 | 19.5 |
| 7 | 24.5 | 5.01 | 221 (5.6) | 23.5 (2.0) | 33.4 (2.0) | 11 | 22.4 |
| 9 | 27.2 | 4.74 | 202 (5.0) | 13.1 (1.1) | 28.1 (1.7) | 14 | 28.9 |
| 11 | 27.3 | 5.21 | 205 (5.3) | 4.8 (0.4) | 29.7 (1.6) | 17 | 27.0 |
| 14 | 29.1 | 4.73 | 235 (5.8) | 16.6 (1.2) | 34.7 (1.7) | 20 | 25.1 |
| 18 | 27.2 | 4.77 | 198 (5.1) | – | 32.8 (1.6) | 23 | 23.8 |
| 23 | 28.5 | 4.98 | 214 (5.2) | 16.0 (1.2) | 22.8 (1.2) | 26 | 23.0 |
| 28 | 26.2 | 4.80 | 205 (5.2) | 16.5 (1.2) | 23.1 (1.2) | 29 | 22.7 |
| 33 | 24.3 | 4.87 | 203 (5.0) | 6.5 (0.5) | 25.2 (1.2) | 32 | 23.2 |
| 38 | 24.7 | 6.14 | 201 (6.8) | 14.7 (1.1) | 30.9 (1.5) | 35 | 15.9 |
| 43 | 23.2 | 4.56 | 188 (4.8) | 3.6 (0.3) | 27.3 (1.3) | 38 | 15.5 |
| 48 | 23.8 | 5.01 | 223 (5.5) | 12.2 (0.9) | 25.4 (1.3) | 41 | 16.9 |
| 53 | 24.1 | 5.04 | 263 (6.3) | 20.8 (1.6) | 38.5 (1.7) | 44 | 14.3 |
| | | | | | | 47 | 20.2 |
| | | | | | | 50 | 19.1 |

Parentheses under ^{40}K , ^{210}Pb and ^{226}Ra activity concentrations in the table denote to uncertainty of the measurements. Porosity was measured at every 3 cm of the soil horizon from the uppermost to a depth of 50 cm

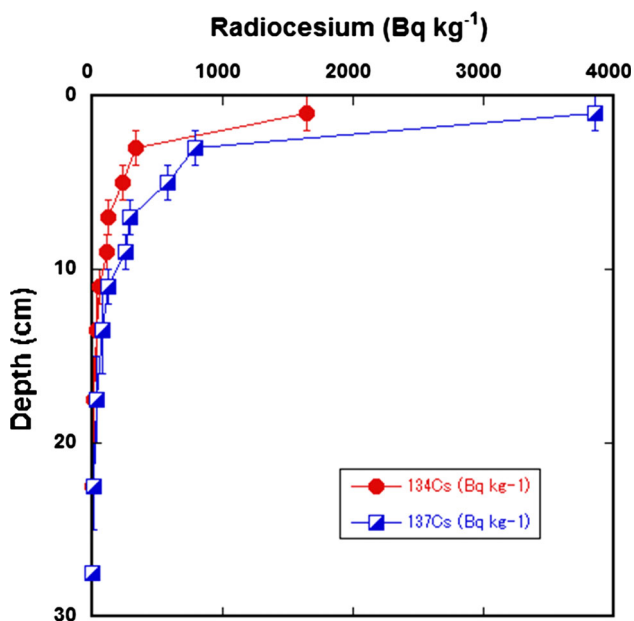


Fig. 2 Depth distribution profiles of radiocesium (^{134}Cs and ^{137}Cs) in soil collected on the test site on August 21 2013. Activity concentration of both nuclides was corrected to the values on the time of soil sampling (August 21 2013)

Soil radon (^{222}Rn) monitoring started in August, 2013 on the observation site of known basic information on geology, climate and soil properties, already described in the site description in the “[Experimental procedure](#)” and also in Table 1 Materials and methods). Figure 3 shows time series changes in ^{222}Rn activity concentration in soil air at different

soil depths (0.3, 0.6, and 1.0 m) at the observation point, together with atmospheric pressure from August 21 to December 6, 2013. Soil radon level varied to a great extent depending on various factors, including meteorological and soil parameters. Different soil radon levels appeared at different soil depths, indicating equivalent ^{222}Rn concentrations ($^{222}\text{Rn}_{\text{eq}}$) to be 7.5, 14 and 23 kBq m^{-3} at 0.3, 0.6 and 1.0 m in depth, respectively (Dörr and Münnich 1990). Plotting $^{222}\text{Rn}_{\text{eq}}$ values against soil depth gave an infinite ^{222}Rn concentration ($^{222}\text{Rn}_{\infty}$) as 53.6 kBq m^{-3} at our observation site (Dörr and Münnich 1990). Using this value of $^{222}\text{Rn}_{\infty}$ (53.6 kBq m^{-3}), soil density (ρ), ^{226}Ra activity concentration in soil (^{226}Ra) and total porosity (p) shown in Table 1, emanation coefficient of ^{222}Rn (ε) was estimated with the following equation:

$$^{222}\text{Rn}_{\infty} = ^{226}\text{Ra} \cdot \varepsilon \cdot \rho \cdot (1 - p) / p \quad (1)$$

Values of ε obtained at different depths were then used for evaluating ^{222}Rn generation rate (v_s) from the source with the decay constant of ^{222}Rn (λ_{Rn}) as follows:

$$v_s (\text{Bq m}^{-3} \text{ s}^{-1}) = \varepsilon \cdot \rho \cdot ^{226}\text{Ra} \cdot \lambda_{\text{Rn}} \cdot (1 - p) / p \quad (2)$$

Figure 4 gives the v_s values at different soil depths giving a minimum value (0.64 $\text{Bq m}^{-3} \text{ s}^{-1}$) at a depth of 0.2 m, and increasing values up to 2.2 $\text{Bq m}^{-3} \text{ s}^{-1}$ down to a depth of about 0.5 m. The results may suggest that it takes several hours for ^{222}Rn to be in an equilibrium state in soil.

Now, it should be noted that two big typhoons (Typhoons No. 18 and No. 26) passed over the site in mid-September and in mid-October, 2013, respectively, as shown in Fig. 3. Atmospheric pressure decreased drastically, and it then recovered within a short period of time

Fig. 3 Time series plots of ^{222}Rn activity concentration at different depths (0.3, 0.6 and 1.0 m) and of atmospheric pressure from August 21 to December 6 in 2013. Two typhoons struck the test site on September 16 (Typhoon No. 16) and October 16 (Typhoon No. 23) in 2013. Upward (E1, E2, E3, E4, E5, E6, and E7) and downward (E8 and E9) changes in ^{222}Rn concentration with time were observed at all the depths, in which E denotes to an abbreviation of event

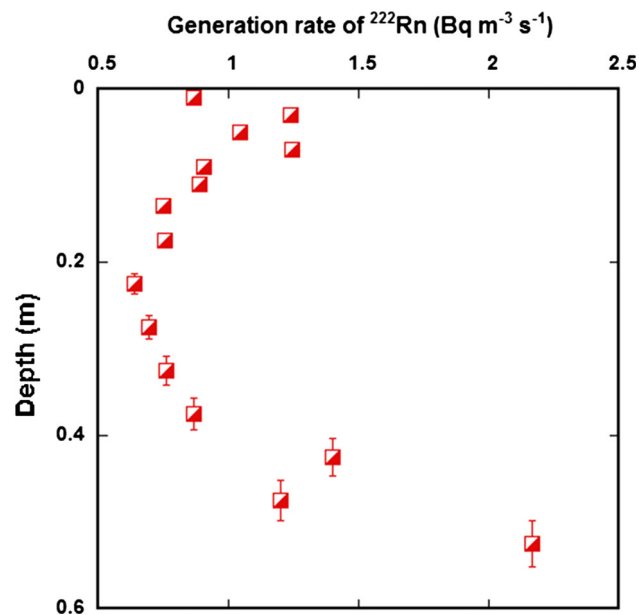
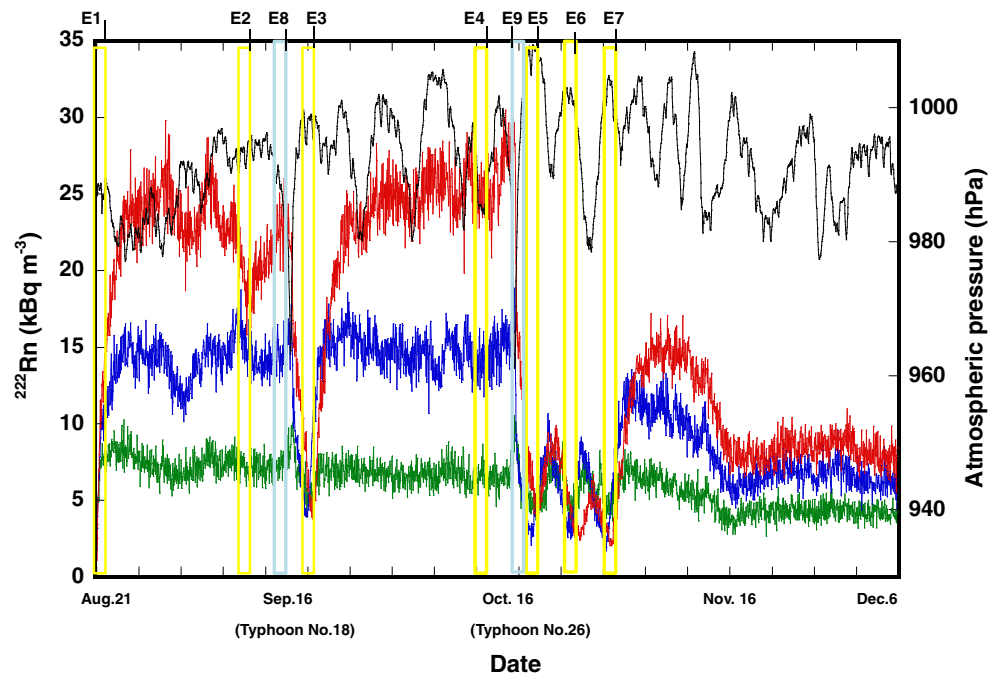


Fig. 4 Change in ^{222}Rn generation rate ($\text{Bq m}^{-3} \text{s}^{-1}$) with soil depth calculated from sets of observed data on ^{226}Ra activity concentration and porosity of the soil

depending on the moving speed of a typhoon. In contrast, soil ^{222}Rn activity concentration decreased slowly to a bottom value ($\sim 4 \text{ kBq m}^{-3}$) at all the depths (0.3, 0.6 and 1.0 m) during the typhoon periods. It is probably because supplying rate of ^{222}Rn from the parent ^{226}Ra in soil was too low to catch up with the concentration in a steady-state level in this period.

There is a lack of information in the literature concerning effects of typhoon on soil radon concentration. It may be the only one that Richon et al. (2003) monitored soil radon (^{222}Rn) at Taal volcano in Philippines from 1993 to 1996 to investigate possible relationship between ^{222}Rn and an earthquake. They concluded that a ^{222}Rn anomaly (extremely high ^{222}Rn level) appeared 22 days before the M 7.1 Mindoro earthquake in 1994, being a precursor of the quake, not resulting from typhoon Teresa passing a few days before. According to them, the only proof for the above conclusion was that ^{222}Rn level was not affected so seriously by another super typhoon (Angela) striking the island just one year later. Findings of theirs were obviously different from ours in the present study, in which soil radon concentration was affected greatly by the passing typhoon.

To elucidate the mechanism of ^{222}Rn transportation in soil air, nine periods of time (E1–E9) depicted in Fig. 3 were selected, in which upward (E1–E7) and downward (E8 and E9) changes in ^{222}Rn concentration with time appeared at all three depths. Here, it was assumed that increasing ^{222}Rn concentration after the Typhoon No. 18 (Event 3) in the figure should be a diffusion-controlled process due to a ^{222}Rn concentration gradient between shallow and deeper portions of the soil under a recovering high-pressure region in the atmosphere. This assumption leads to obtaining effective diffusion coefficient (D_e) of ^{222}Rn with the equation below, where ^{222}Rn , F and x denote to ^{222}Rn concentration (Bq m^{-3}), ^{222}Rn flux ($\text{kBq m}^{-2} \text{s}^{-1}$) obtained at E3 in Fig. 3 and soil depth (m), respectively. A differential part ($d^{222}\text{Rn}/dx$) in Eq. (3) was

evaluated using an equilibrium concentration of ^{222}Rn at each depth.

$$F = D_e \left(\frac{d^{222}\text{Rn}}{dx} \right) \quad (3)$$

Mean D_e value thus obtained as $5 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ is reasonable by considering homogeneous and well-drained soil in our test site (Nazaroff 1992; Sakoda et al. 2011). Diffusion-controlled ^{222}Rn flux in soil air was therefore estimated to be about $1 \times 10^{-1} \text{ Bq m}^{-2} \text{ s}^{-1}$ during the observation period from August to September, 2013. It is much higher than that obtained in a forest site of Sapporo under thick snowpack in winter (Fujiyoshi et al. 2013).

Transportation mechanism of ^{222}Rn in soil air was considered with a set of data on ^{222}Rn flux at each period of time (E1–E9). As shown in Fig. 5, ^{222}Rn in soil air moves by a diffusion-controlled mechanism in most of the periods shown in Fig. 3. However, there are two cases that mass flow of soil air controls ^{222}Rn transport as in cases E8 and E9 in which an approaching low-pressure front caused strong upward movement of air from the soil to the atmosphere resulting in a lack of ^{222}Rn in soil air during the typhoon periods. The difference in the ^{222}Rn flux between downward (E8) and recovering (E3) time affected by the Typhoon No. 18 (Sept 16 2013) is probably due to mass flow of soil air in the former.

After the typhoon No. 26 (October 16, 2013) passed on, ^{222}Rn concentration was not recovered up to the previous level at each depth (Fig. 3). Small up and down changes in radon level appeared for about 10 days after this typhoon. Such a behavior of ^{222}Rn in soil air is supposed to be associated with changing meteorological conditions and soil properties in autumn season when atmospheric temperature gradually decreases. Figure 6a shows consequences of ^{222}Rn concentration at three depths and of precipitation (mm h^{-1}). As shown in the figure, it rained several times ranging from 1 to 9 mm h^{-1} , which clearly affected subsequent soil radon level to a great extent. Temperature in soil air (and also on the ground surface) did not show clear diurnal variability in this period (Fig. 6b). Barometric pressure at different depths showed small differences, especially when the surface portion (0.3 m) is compared with the other two (0.6 and 1.0 m), as shown in Fig. 6c. However, it is clear in Fig. 6d that barometric pressure difference ($P_g - P_s$) between ground surface (P_g) and soil air (P_s) differed in time series profile at a depth of 0.3 m if compared with other depths. As depicted from Figs. 6a and b, properties of surface soil was directly affected by meteorological conditions, such as air movement and intensity and amount of precipitation. Frequent precipitation in autumn season gave some retarding effect on ^{222}Rn concentration at different depths, in which a deeper

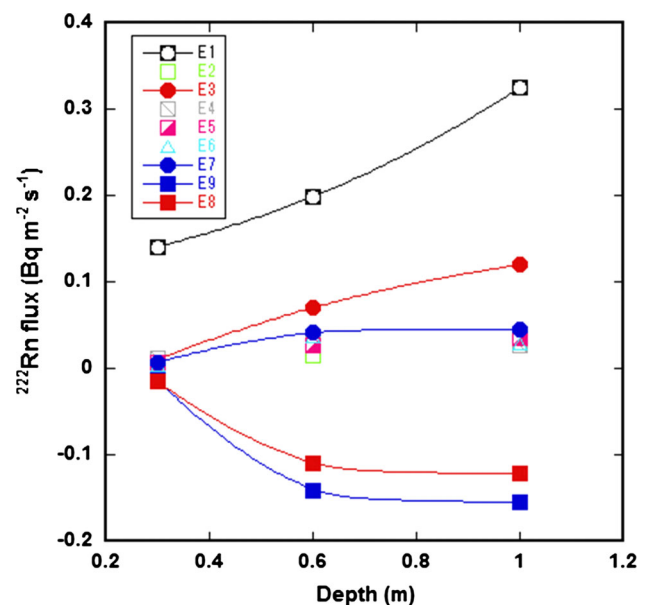


Fig. 5 Flux of ^{222}Rn as a function of soil depth obtained on nine events shown in Fig. 3. Most of the events except for E1, E8 and E9 followed diffusion-controlled ^{222}Rn transportation in soil air. Mass flow of soil air was predominant in the ^{222}Rn transportation only in the cases of E8 and E9, when extremely low atmospheric pressure front was passing over the observation site

layer of the soil responded much slower to changing meteorological conditions (Fig. 6a). Dynamic behavior of ^{222}Rn in surface soil found in this study suggests that further investigation should be necessary for evaluating effect of humidity on ^{222}Rn variability in soil air. This may be further required to elucidate effects of water (and water vapor) transportation on the fate of fallout nuclides deposited on the forest floor. Monitoring has still been continued, aimed at evaluating water (water vapor) movement in soil as well soil ^{222}Rn concentration at three depths in the present test site.

Conclusions

Soil radon (^{222}Rn) was monitored from mid-summer to early winter in 2013 at three depths in a forest site contaminated with fallout radionuclides derived from the accident of the Fukushima Daiichi Nuclear Power Plant in 2011. Different ^{222}Rn levels appeared depending on soil depths, which gave an equivalent ^{222}Rn concentration ($^{222}\text{Rn}_{eq}$) of 7.5, 14 and 23 Bq m^{-3} at the depths of 0.3, 0.6 and 0.7 m, respectively. Two big typhoons passing over the observation site showed a great effect on soil radon variability, in which upward transportation of ^{222}Rn associated with passed typhoon was governed by diffusion-controlled mechanism. Effective diffusion coefficient of ^{222}Rn (D_e) was easily evaluated using time series ^{222}Rn data at three

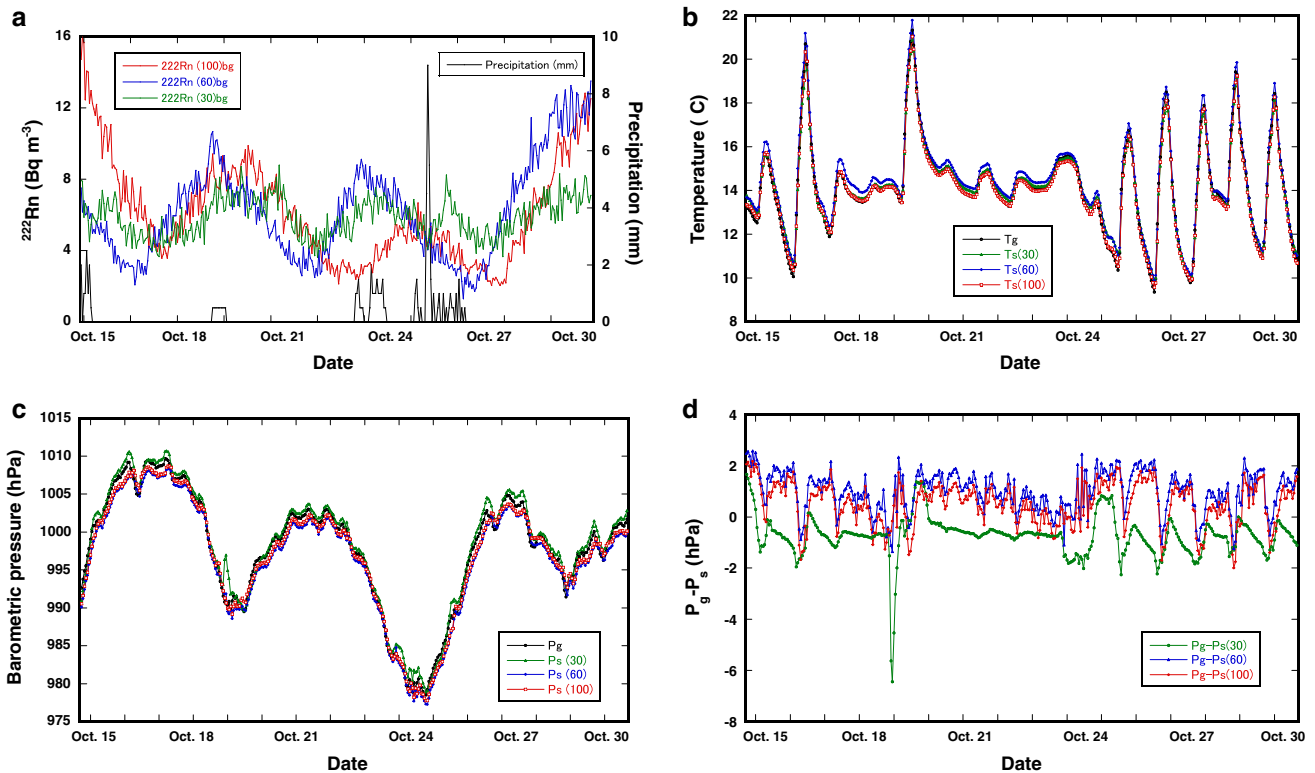


Fig. 6 a, b, c, d Time series plots of ^{222}Rn concentration with precipitation (a), ^{222}Rn concentration with soil temperature (b), barometric pressure (c) and difference in barometric pressure on the ground surface from those in soil air at different depths (d) obtained from October 15 to 30, when Typhoon No. 23 had passed over the test

site. In a, variability of ^{222}Rn concentrations with time did not show similar patterns at individual depths. It is clear in d that pressure difference between the ground surface and soil air was quite different only at the surface portion (0.3 m in depth) of the soil

different soil depths during a typhoon event in September, 2013. This is important for investigating dynamic behavior of ^{222}Rn in soil air with an “in situ” diffusion coefficient. Diffusion-controlled ^{222}Rn flux was obtained in selected time regions using this effective diffusion coefficient. Precipitation and subsequent change in soil humidity also affected ^{222}Rn level to a great extent, depending on soil depth, which remains to be investigated thoroughly. The authors have been further studying dynamic behavior of gaseous and water components in soil using ^{222}Rn as a radiotracer for tracking the fate of deposited radiocesium on the forest floor.

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