Soil radon in winter months under cool temperate deciduous stands in Hokkaido, Japan

Ryoko Fujiyoshi[1*], Masanori Okabayashi[1], Yousuke Sakuta[1], Kazumasa Okamoto[1], Takashi Sumiyoshi[1], Ivan Kobal[2], Janja Vaupotič[2]

[1]Faculty of Engineering, Hokkaido University, Sapporo 060-8628, Japan
[2]Radon Center, Jožef Stefan Institute, 1000 Ljubljana, Slovenia
* Corresponding author: Tel: +81 11 706 6674, Fax: +81 11 706 6675, Email: fuji@eng.hokudai.ac.jp

Abstract Soil radon (^{222}Rn) has been monitored during winter months under cool temperate deciduous stands of different surface geology in Tomakomai and in Sapporo, Hokkaido, Japan. Radon level was lower in Tomakomai of immature soil of porous volcanic ash emitted from an active volcano (Mt. Tarumae), compared with those in Sapporo of alluvial sediments. In Tomakomai mean value of the ^{222}Rn activity concentration was higher in winter (570 Bq m^{-3}) than in summer (350 Bq m^{-3}) at a depth of 1 m, which is consistent with the results in cold and dry winter reported in the literature. In contrast, soil radon decreasing with decreasing soil temperature from mid-September remained low level (2.6 kBq m^{-3}) under lingering snow in Sapporo, which had already been observed in the same location. Measurements of the activity concentrations of ^{222}Rn in snow and in snow air as well as in soil air indicate that small amount of ^{222}Rn is released from the ground surface to the overlying snowpack with a ^{222}Rn flux density of 1.0×10^{-3} Bq m^{-2} s^{-1} under thick snow cover in Sapporo.

Keywords Radon, Soil air, Snowpack, Transport

Introduction

Soil radon has been measured extensively in various places in the world not only to evaluate possible radiation hazard to the general public, but to study physical processes in lower atmosphere (Ghosh et al 2009; Etiope and Martinelli 2002). Analytical results are very important as primary data source for radiation protection and geophysical exploration. However, there are many factors affecting soil radon concentration, such as atmospheric pressure, temperature and humidity in the environment. Among various factors influencing soil radon variability, atmospheric (soil) temperature was found to be predominant under a cool-temperate deciduous stand in Sapporo, Japan (Fujiyoshi et al 2002, 2005, 2006, 2010). Regular diurnal variation in soil radon concentration was disturbed by approaching low pressure front and subsequent precipitation in three seasons except for winter.

There are not abundant data on the behavior of soil radon under snowpack in winter, which is often hampered by accessibility to remote observation sites. It has been believed that soil radon level is higher in winter than in summer due to a frozen top soil covered with snow (Winkler et al 2001). However, a different result to theirs was obtained in our previous measurements of ^{222}Rn concentration in soil air under snowpack in winter, in which ^{222}Rn level was relatively low with smaller variability compared with those in summer (Fujiyoshi et al 2002, 2010). There is a report by Perrier et al (2009) who monitored radon concentration in the saturated soil of Alpine grassland at altitudes of 1590 and 1792 m a.s.l. in winter. They found opposite seasonal variations of radon concentration at a depth of 70 cm between the two sites, which was ascribed to two types of water infiltration. It is therefore possible to find another behavior of radon under snowpack in winter.

The purpose of the present study is to compare the results of soil radon monitoring in winter under cool temperate deciduous stands at two different geological locations (Tomakomai and Sapporo) in Hokkaido, Japan, and to find possible reason(s) for winter low in soil ^{222}Rn concentration in Sapporo by measuring ^{222}Rn concentration in snow and in snow air.

Experimental procedure

Description of the sites

Fig.1

Table 1

Two forest sites of different geology (Tomakomai and Sapporo) were selected in this study. Figure 1 shows a map of these sites in Hokkaido Japan. Table 1 summarizes some data on geography, geology and meteorology of the sites. Tomakomai Experimental Forest of Hokkaido University is located in Tomakomai City (42.40 ºN, 141.36 ºE) with 2.715×10^{7} m^{2} in area. The forest belongs to the cool temperate deciduous stand of several tree species such as Japanese Larch (*Larix kaempferi*), Sakhalin Fir (*Abies sachalinensis*) *and* Sakhalin Spruce (*Picea glehnii*). Most of the area is covered by immature volcanic soil derived from an active volcano (Mt. Tarumae) located about 20 km north-western direction from Tomakomai city. Mean annual temperature is 6.4 ºC and annual precipitation is about 1,200 mm. Snowfall is relatively low about 40 cm in depth at most, which includes soil freezing during winter.

Another site was located on the campus of Hokkaido University of a semi-natural deciduous stand (43.08 ºN, 141.38 ºE). Predominant tree species are poplar (*Populus*), white birch (*Betula platyphylla*), oak (*Quercus crispula*) and elm (*Ulmus davidiana var japonica*). Geology of the site is alluvial sediments from Kotoni River. A dark green clay layer appeared at a depth of 40-50 cm. Mean annual temperature is 6 - 9 ºC and annual precipitation is 800 -1600 mm. Maximum snow depth is higher in Sapporo (~100 m) than in Tomakomai (~40 cm).

Soil properties

Several soil properties were investigated including dry density, porosity, humidity, ignition weight loss and pH. Data on dry density, humidity and porosity of a sample were obtained in the laboratory by a standard procedure (Japanese Society of Soil Science and Plant Nutrition, 1997). Ignition weight loss of a sample was obtained after heating at 500 ºC for 2 hrs. Soil pH was measured in the field using a pH meter (HI 99121, HANNA Instruments, US).

Gamma spectrometry

Radioactivity of ^{226}Ra in soil was determined by gamma spectrometry with a HPGe system (SEIKO EG&G, Japan). Standard reference materials (IAEA 327 and IAEA-444) were used to evaluate the activity concentration of individual samples from counting data obtained with the same geometry under identical operating conditions. Energy and efficiency calibrations were periodically carried out as well as background checks. Precision of the measurement was estimated by considering the photo peak areas for both samples and standards. Interference of ^{235}U emitting photons with similar energy to that of ^{226}Ra were not considered, because there were no other peaks assigned to ^{235}U on the spectrogram. The minimum detectable activity (MDA) of ^{226}Ra was 0.7 Bq for a sample (approx. 100 g) in a plastic container (100 cm^3) for 40 hour counting time in our counting system (Fujiyoshi et al 2011).

Soil air sampling and ^{222}Rn measurement

Activity concentration of ^{222}Rn in soil air was monitored every 1 hour with a silicon detector (Barasol, Algade, France) at a depth of 30 cm in winter. The probe was set at a depth of 100 cm in other seasons. Soil temperature and barometric pressure were also measured with this probe. The sensitivity of the detector was declared as 0.02 pulses h^{-1} for 1 Bqm^{-3} with energy resolution of 60 keV for alpha particles emitting from ^{222}Rn (5.47 MeV). Appropriate temperature for the measurement was ranging from -20 to 60 ºC according to the manufacture.

Scintillation cell (300A, Pylon, Canada) and a radiation monitor (Model AB-5, Pylon, Canada) were used to measure ^{222}Rn concentration in snow air collected from pore space of the snowpack. Air was carefully collected with an evacuated scintillation cell (3.5×10^{-2} hPa) from a hole (2.5 cm in diameter and variable depth) dug in the snowpack. Depth of the hole was adjusted as close to the ground surface as possible. Air samples were kept in the laboratory for about three hours to allow the radon daughters being equilibrium with the parent radon gas in the cell (Neznal et al 2004).

Activity concentrations of ^{222}Rn in snow samples were determined with a liquid scintillation technique. A snow sample was collected in a Büchner funnel (1 L) from a snow layer close to the ground surface (within 5 cm in thickness from the ground). After measuring weight of the snow sample, 40 ml of a scintillator (Ecoscint O, National Diagnostics, US) was added to the container to melt it in a water bath at 80 ºC. The sealed funnel was then shaken vigorously with a mechanical shaker for 10 min. Liquid scintillation counting was conducted on an aliquot of the solution (20 ml) in a vial with a scintillation counter (LSC-5000, ALOKA, Japan). Activity concentration of ^{222}Rn was determined by an efficiency tracing method (Aloka, 2007) in which plots of counting data obtained within several window ranges were extrapolated to the whole window (y-axis) to find the value with 100 % count efficiency.

Results and discussion

Tomakomai experimental forest

Table 2 summarizes some of the soil properties under Tomakomai experimental forest investigated in this study. The soil was characterized as an immature soil of volcanic ash emitted from an active volcano (Mt. Tarumae) located about 20 km north-west direction from the site. The uppermost part of the soil was covered with an organic layer of about 5 cm in thickness. Laminar structures of volcanic ash with different color and grain size were observed at deeper portion of the soil resulting in high porosity of the soil (Table 2). Table 2

Figure 2 shows time series plots of ^{222}Rn activity concentration in soil air measured at a depth of 100 cm in winter months of 2010- 2011 (Nov. 13 2010 to Mar. 18, 2011). Soil temperature decreased continuously from November (~11 ºC) to March (~3.5 ºC) at this depth, whereas atmospheric temperature was relatively high (7~12 ºC) in November, but it suddenly decreased in mid-December when snow fall started on Dec. 18, 2010.

As shown in Fig.2, soil radon concentration was quite low (mean value; 570 Bq m^{-3}) at a depth of 100 cm in the whole winter 2010-2011. The low radon level in Tomakomai soil is supported by low ^{226}Ra concentration (<10 Bq kg^{-1}) in soil with high porosity. It should be noted that mean ^{222}Rn concentration was found to be higher in lingering snow (560 Bq m^{-3}) from Nov.18, 2010 to Mar. 6, 2011than that in other seasons of the year (350 Bq m^{-3}). Soil radon measurements with an alpha scintillation technique confirmed the low radon level in summer (mean value; 260 Bq m^{-3}) in the Tomakomai site (Vaupotič et al 2012). These results are consistent with those obtained in other countries of cold climates, in which soil radon levels become high due to frozen ground surface and thick snowpack (Winkler et al 2001). Similar winter high of ^{222}Rn was also observed at a Mediterranean test site (Valle della Caffarella) in Italy where the activity concentration was 100-200 kBq m^{-3} and 40-160 kBq m^{-3} at a depth of 80 cm in winter and in summer, respectively (Castelluccio M. et al 2010). According to them, the soil is drier, warmer and more permeable in summer than in winter, which causes lower concentrations in summer. Their result rather indicates a summer low due to Mediterranean weather conditions rather than a winter high observed in our site.

Semi-natural woods on the campus of Hokkaido University in Sapporo

Another monitoring site located on the campus of Hokkaido University was of alluvial sediments from Kotoni River in Sapporo under a cool-temperate deciduous stand. Table 2 summarized some of the soil properties, in which total porosity decreased with depth reflecting increasing amount of clay minerals in the soil. Clay was a major component of the soil deeper than 50 cm in depth. Soil pH showing the highest value at the soil surface (pH = 4.9), decreased to a value of about 4.6 in the clay layer at a depth of 50 cm.

The first snowfall was observed relatively earlier in Nov. 8, 2011 than years past, which resulted in a lingering snow of a few cm in depth till early December due to continuing low temperature around 0 ºC. Snow depth increased gradually to be a maximum value of 77 cm in February, 2012.

Figure 3 shows the time series ^{222}Rn activity concentrations in soil air at a depth of 30 cm during winter months from 2011 to 2012. The activity concentration varied from 1.0 kBq m^{-3} to 4.5 kBq m^{-3} with a mean value, 2.6 kBq m^{-3}. Soil radon levels were appreciably lower than those in summer (5.0 kBq m^{-3}), which had already been found in our previous work (Fujiyoshi et al 2010). Soil temperature at this depth remained constant about 0 ºC, which may result in small variability of ^{222}Rn concentration under the snow- pack from January to March. Soil radon level gradually increased with increasing atmospheric temperature in the end of March (Fujiyosni et al 2010).

Similar winter low of soil radon level was observed at the Sur-Frêtes ridge (French Alps) in France (Perrier et al 2009). They monitored radon in soil at a depth of 70 cm from 1995 to 1999 to find low values in winter with fluctuations associated with temperature variations. They evaluated the variations caused by water infiltration using a mixing model.

However, it is usually known that radon concentration is higher in winter than in summer as reported by Winkler et al. (2001). They investigated seasonal fluctuations of soil radon at two different depths (0.5 and 1.0 m) in an open glass land in Germany. They found maximum concentrations of ^{222}Rn in winter months with 6- 50 kBq m^{-3} and 8-34 kBq m^{-3} at the depths of 0.5 and 1.0 m, respectively. According to them, radon concentration in 0.5 m depth was consistently higher than that in 1.0 m depth, which could be explained by the depth concentration profiles of ^{226}Ra with the highest value of 100 kBq kg^{-1} in the top soil layer and low (about 25 Bq kg^{-1}) in the deeper portion. Due to the frozen top-soil, radon can’t easily escape to the atmosphere.

In order to elucidate possible consequences of low radon level in winter observed in our site both snow and air in snow were carefully collected from the bottom layer of the snowpack to determine ^{222}Rn activity concentrations with a solid (ZnS) scintillation and a liquid scintillation counters, respectively. The results are tabulated in Table 3 in which snow depth on the sampling day was also included. As shown in the table, ^{222}Rn concentration in snow on the basis of melted water volume (1 dm^{3}) varied from a value lower than the detection limit of our counting system (0.02 Bq dm^{-3}) to that of 1.20 Bq dm^{-3}. The result indicates that radon in soil air may partially dissolve in aqueous phase present in the snow pack close to the ground surface, since the solubility of ^{222}Rn in water is 0.5 cm^{3}/cm^{3} (H_2O) at 0 ºC. Winter low of soil radon level may be explained by a water-soluble property of radon gas, because it is likely to occur in our site free from the frozen subsoil close to the ground surface even in mid-winter. It should be noted that this is a distinct contrast to the soil in Tomakomai which is completely frozen down to a depth of about 30 to 40 cm in winter.

Also found in the table, ^{222}Rn concentrations in snow air were 2 to 6 times higher than those in snow itself except for one case resulting in an exceptionally high ^{222}Rn concentration (547 Bq dm^{-3}) on Jan. 27, 2012. Similar anomaly to those with high radon value (2.2 kBq dm^{-3}) was also observed in the same site on Apr. 7, 2011, when the snow on the ground was finally disappearing. The results together with the bulk density of snow samples suggest that there is a lot of pore space in snow to trap radon gas within. It is therefore considered that analytical results on ^{222}Rn concentration in snow may refer partly to radon contained in its pore space. Radon in soil air is not confined within the soil but to be released to the overlying snowpack. Sudden output of radon may sometimes occur under certain conditions not to be elucidated in this study.

Radon concentration in snow is affected by many factors, especially by meteorological ones in which atmospheric temperature is found a predominant factor (Fig. 3). Yamazawa et al (2005) measured radon concentrations in snow pore to find that they were highly variable depending on the wind conditions with values between 0.5 and 4.0 kBq m^{-3} at the middle of the snowpack of 1.22 m in depth. Without data on wind speed at our site but with those from Sapporo District Meteorological Observatory (http://www.jma-net.go.jp/sapporo/), no special relationship was obtained between ^{222}Rn in snow and wind speed in this study. Local weather conditions should be observed to evaluate possible relationships between radon and meteorological parameters.

Fig. 3

Transport of ^{222}Rn in soil air under snowpack in winter

Transport of ^{222}Rn in soil under snowpack in winter was considered using data on ^{222}Rn activity concentrations in soil air, snow and snow air obtained in this study. As shown in Fig.3, ^{222}Rn concentrations in soil air were lower and varied to a lesser extent in winter than in summer with the mean value of 2.6 kBq m^{-3} during lingering snow. There are many factors affecting variability of soil radon concentrations with time such as temperature, barometric pressure, moisture content and possibly sudden degassing of other gases than radon from biological and geological origins. The difference of mean ^{222}Rn concentration in summer and in winter is supposed to be attributable to changing water-filled porosity in soil, since depth profiles of ^{226}Ra activity concentration were virtually constant (13~15 Bq kg^{-1}) in the soil of our test site independent of the season. Changing water-filled porosity with time affects ^{222}Rn emanation and subsequent radon transport, which further results in varying soil radon flux to the ground surface (Fujiyoshi et al 2005). With respect to radon emanation in soils, Sakoda et al (2011) reviewed the results of emanation measurements for various geological materials including soils in the world to conclude a representative value of ^{222}Rn emanation for soil to be 0.13. They showed not only moisture but temperature of soil and grain size affect emanation of radon due to a reduction in physical adsorption of radon onto soil grains.

In order to evaluate characteristics of radon transport in soil air, an equilibrium value of soil radon concentration ($^{222}Rn_{\infty}$) and relaxation depth (L) was estimated using a set of data on depth distribution profiles of soil radon concentrations ($^{222}Rn_{z}$) in summer (Yakovleva 2005; Vaupotič et al 2012). Mean values for $^{222}Rn_{\infty}$ and L were obtained to be 9.7 kBq m^{-3} and 1.37 m at the observation point, respectively. The equilibrium value of ^{222}Rn concentration thus obtained in summer was assumed to be similar in winter. Mean value of the relaxation depth of ^{222}Rn in winter was calculated to be 9.6 $\times 10^{-1}$ m. Using data on the relaxation depth and decay constant of ^{222}Rn (2.16×10^{-6} s^{-1}), effective diffusion coefficient of ^{222}Rn (D_e) was obtained to be 2.0 $\times 10^{-6}$ m^2 s^{-1} in winter. This value was about 50 % of that in summer (4.1×10^{-6} m^2 s^{-1}).

Among a variety of data accumulated on effective diffusion coefficients of radon in the literature, not so many reports have been concerned with the behavior of soil radon in winter (Hassan et al 2009). Yamazawa et al (2005) studied radon diffusion in soil air in winter both in the laboratory and in the field. Effective diffusion coefficients obtained in the laboratory decreased by a factor of about 2 in moist soils compared with those for dry soils. They concluded the existence of snow cover considerably reduced the radon exhalation to a few percent of that of no-snow seasons ascribing to wetting snow-soil interface.

The values for effective diffusion coefficient D_e obtained in this study may be reliable in a category of alluvial sediments in the data set published by Argonne National Laboratory in US (http://www.ead.anl.gov/resrad/datacoll/radon.htm). The results were then used to evaluate radon flux density from the ground surface (Yakovleva 2005).

The ^{222}Rn flux at the soil surface (F_{Rn}) was calculated with the following equation (1):

$$F_{Rn} = D_e p_a \frac{^{222}Rn_{\infty}}{L} \qquad (1)$$

where D_e, p_a, $^{222}Rn_{\infty}$ and L denote effective diffusion coefficient (m^{-2} s^{-1}), air-filled porosity (-), steady state ^{222}Rn activity concentration (Bq m^{-3}) and relaxation depth (m), respectively. The obtained values on ^{222}Rn flux at the soil surface of our site were 1.0 mBq m^{-2} s^{-1} and 2.9 mBq m^{-2} s^{-1} in winter and in summer, respectively. The result indicates that under a winter condition including low soil ^{222}Rn concentration and constant soil temperature (0 °C) in our site, transport of ^{222}Rn in soil was about 1/3 in magnitude compared in summer.

Seasonal variation in radon fluxes were evaluated by Grifffiths et al (2010). They constructed a time-dependent map of ^{222}Rn flux density at the Australian land surface with a spatial resolution of 0.05° and temporal resolution of one month. They obtained mean ^{222}Rn flux density over the period July 1979- to June 2010 to be 23.4 ± 2.0 mBq m^{-2} s^{-1}, which was consistent with previous estimate of 22 mBq m^{-2} s^{-1} by Schery et al (1989) in the same site.

Although there is no correlation between ^{222}Rn concentrations in soil air and in snow air measured simultaneously in our test site, about 5 % of ^{222}Rn in soil air may be degassing from the ground surface (Table 3). It means that a small amount of radon would have been released from soil pores to the overlying snowpack during winter months.

Table 3

Assuming that ^{222}Rn in soil air is transported steadily to the soil surface under snowpack, the time (t) for the radon to be in the same concentration in a definite volume of overlying snow could be calculated using the equation (2):

$$C_{Rn}(Bqm^{-3}) = \frac{F_{Rn}S}{V\lambda_{Rn}}\left[1-\exp(-\lambda_{Rn}t)\right] \qquad (2)$$

C_{Rn} is the ^{222}Rn activity concentration in snow. S and V are surface area (m^2) and volume of snow sample (m^3), respectively.

The result in Table 3 suggests that about five hours is necessary for ^{222}Rn in soil air transported to the ground surface under snowpack to be in the same concentration as those measured in snow. With respect to transportation of radon in soil, some carrier gases like CO_2 and CH_4 may be important in several forms (gas-phase flow, water displacement by a gas, gas slugs and bubbles) depending on “in situ” environment (Etiope and Martinelli 2002).

The result obtained in this study may be the first to show behavior of ^{222}Rn in soil under snowpack in winter. A simulation study is now proceeding to elucidate ^{222}Rn transport in the snowpack by considering physicochemical properties of snow itself. Long-term monitoring of ^{222}Rn concentration in soil air is still necessary to evaluate spatial variability of soil radon level in the whole seasons in detail for understanding behavior of radon in winter (Janja et al 2012).

Conclusion

Radon (^{222}Rn) in soil air was found to be released from the pore space of soil to overlying snowpack in winter in our test site in Sapporo, Japan. The amount is so small that it takes about four hours to be the same concentration in snow measured independently. It may be the first observation that radon is not only accumulated in soil air under snowpack in winter, but released to the ground surfaces.

Acknowledgements

The present research was supported by Japan Society for the Promotion of Science (JSPS) and Slovenian Research Agency under Japan-Slovenia Research Cooperative Program in 2010-2011. The present authors would appreciate Prof. Tsutomu Hiura and his group for helping field works in the Tomakomai Experimental Forest, Hokkaido University.

References

Castelluccio M, Moroni M, Tuccimei P, Neznal M, Neznal M (2010) Soil gas radon concentration and permeability at “Valle della Caffarella” Testi Site (Roma, Italy), 10th International Workshop on the Geological Aspects of Radon Risk Mapping, Sep. 22-25, 2010 (Prague, Czech Republic)

Etiope G, Martinelli G (2002) Migration of carrier and trace gases in the geosphere: an overview. Phys. Earth Planet. Inter. 129:185-204

Fujiyoshi R, Morimoto H, Sawamura S (2002) Investigation of the soil radon variation during the winter months in Sapporo, Japan. Chemosphere 47: 369–373.

Fujiyoshi R, Kinoshita M, Sawamura S (2005) Variation of ^{222}Rn activity concentration in soil gas at a site in Sapporo, Japan. Environ. Geochem. Health 27:539–547

Fujiyoshi R, Sakamoto K, Imanishi T, Sumiyoshi T, Sawamura S, Vaupotič J, Kobal I (2006) Meteorological parameters contributing to variability in ^{222}Rn activity concentration in soil gas at a site in Sapporo, Japan. Sci.Total Environ. 370:224–234

Fujiyoshi R, Haraki Y, Sumiyoshi T, Amano T, Kobal I, Vaupotič J (2010) Tracing the sources of gaseous components (^{222}Rn, CO_2 and its carbon isotopes) in soil air under a cool-temperate deciduous stand in Sapporo, Japan. Environ. Geochem. Health 32(1):73–82

Ghosh D, Deb A, Sengupta R (2009) Anomalous radon emission as precursor of earthquake. J. Appl. Geophys. 69: 67-81

Griffiths AD, Zahorowski W, Element A, Werczynski S (2010) A map of radon flux at the Australian land surface. Atmos. Chem. Phys. 10:8969-8982

Hassan NM, Hosoda M, Ishikawa T, Sorimachi A, Sahoo S, Tokonami S, Fukushi M (2009) Radon migration process and its influence factors; Review. Jpn. J. Phys. 44(2):218-231

Argonne National Laboratory, US. 7.EFFECTIVE RADON DIFFUSION COEFFICIENT. http://www.ead.anl.gov/resrad/datacoll/radon.htm Accessed May 5 2012

Japan Meteorological Agency. http://www.jma-net.go.jp/sapporo/ Accessed Apr. 20 2012

Japanese Society of Soil Science and Plant Nutrition (1997) Analytical Methods in Soil Environment. Konno T (ed) Hakuyusha Japan (in Japanese)

Neznal M, Matolin M, Just G, Turek K (2004) Short-term temporal variations of soil gas radon concentration and comparison of measurement techniques, Radiat. Prot. Dosim. 108(1): 55-63

Perrier F, Richon P, Sabroux JC (2009) Temporal variations of radon concentration in the saturated soil of Alpine grassland: The role of groundwater flow. Sci.Total Environ. 407: 2361-2371

Sakoda A, Ishimori Y, Yamaoka K (2011) A comprehensive review of radon emanation measurements for mineral, rock, soil, mill, tailing and fly ash. Appl. Radiat. Isot. 69:1422-1435

Schery S, Whittlestone S, Hart K, Hill S (1989) The flux of radon and thoron from Australian soils. J. Geophy. Res. 94:8567-8576

Vaupotič J, Gregorič A, Kobal I, Fujiyoshi R, Okabayashi M, Sakuta Y (2012) Radon in soil gas at selected sites in Hokkaido, Japan. International Symposium on Natural Radiation Exposures and Low Dose Radiation Epidemiological Studies (NARE 2012), Hirosaki, Japan.

Winkler R, Ruckerbauer F, Bunzl K (2008) Radon concentration if soil gas: A comparison of the variability resulting from different methods, spatial heterogeneity and seasonal fluctuations. Sci. Total Environ. 272:273-282

Yakovleva VS (2005) A theoretical method for estimating the characteristics of radon transport in homogeneous soil. Annals Geophys. 48(1):195-198

Yamazawa H, Miyazaki T, Moriizumi J, Iida T, Takeda S, Nagara S, Sato K, Tokizawa T (2005) Radon exhalation from a ground surface during a cold snow season. Int. Cong. Ser. 1276:221-222

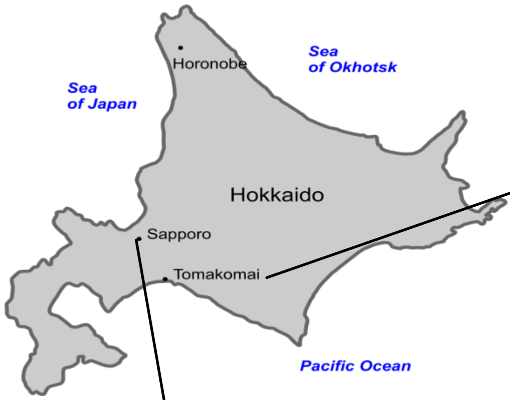
Figure Caption

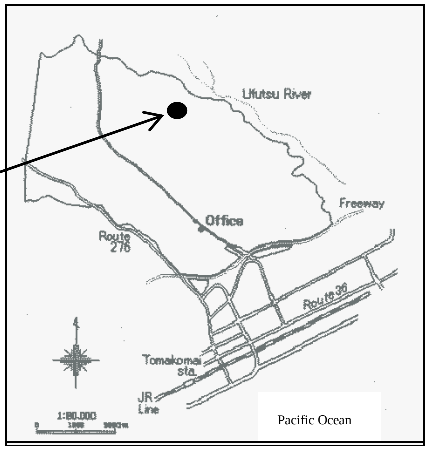
Figure 1 Location map of the sites investigated in this study (Tomakomai and Sapporo) in Hokkaido, Japan.

Figure 2 Time series plots of ^{222}Rn activity concentrations and soil temperature in soil in winter (Nov. 13, 2010 - Mar. 18, 2011)

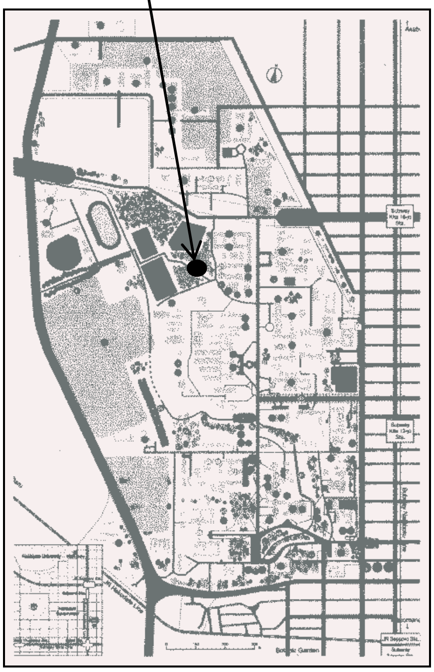
Figure3 Time series plots of ^{222}Rn activity concentrations and soil temperature in soil in winter (Jan. 8 - Mar. 18, 2012).

Figure 4 Relationship between ^{222}Rn in snow air and atmospheric temperature in January, 2012.

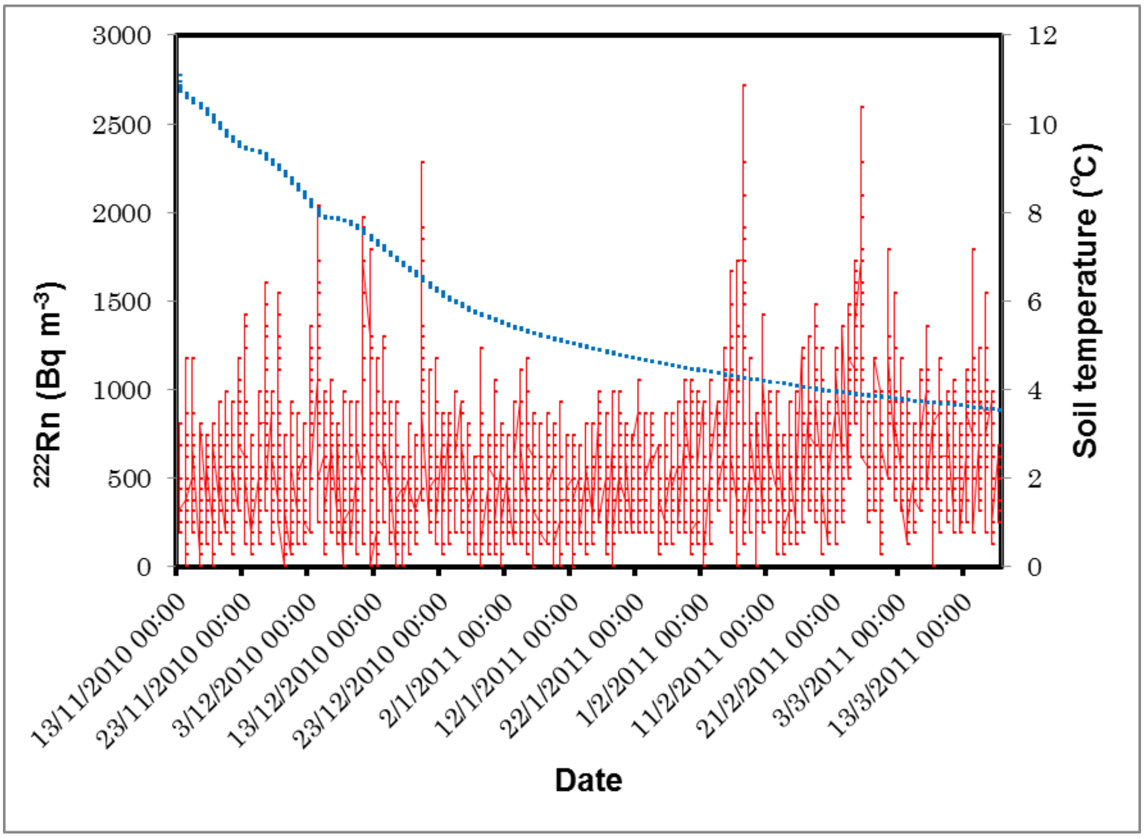




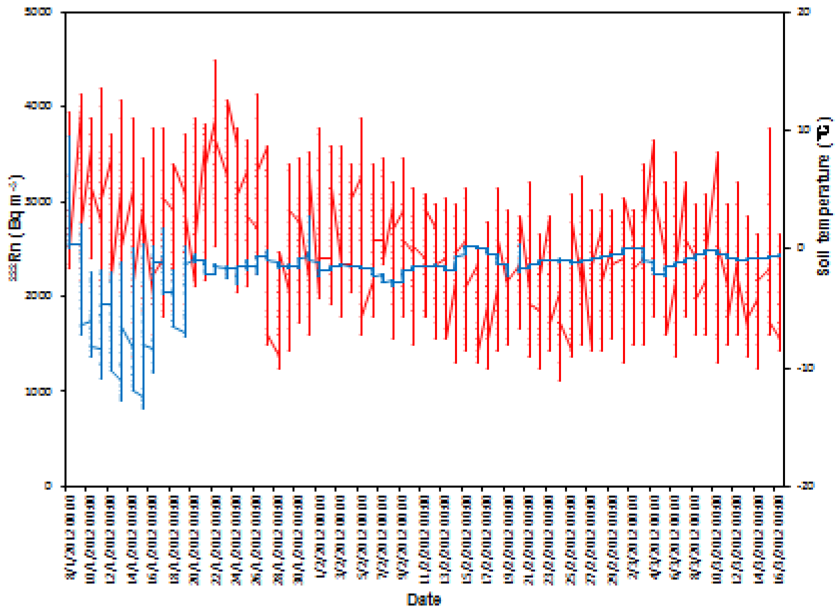
http://forest.fsc.hokudai.ac.jp/~exfor/Toef/toma-e/out-to-e.htm



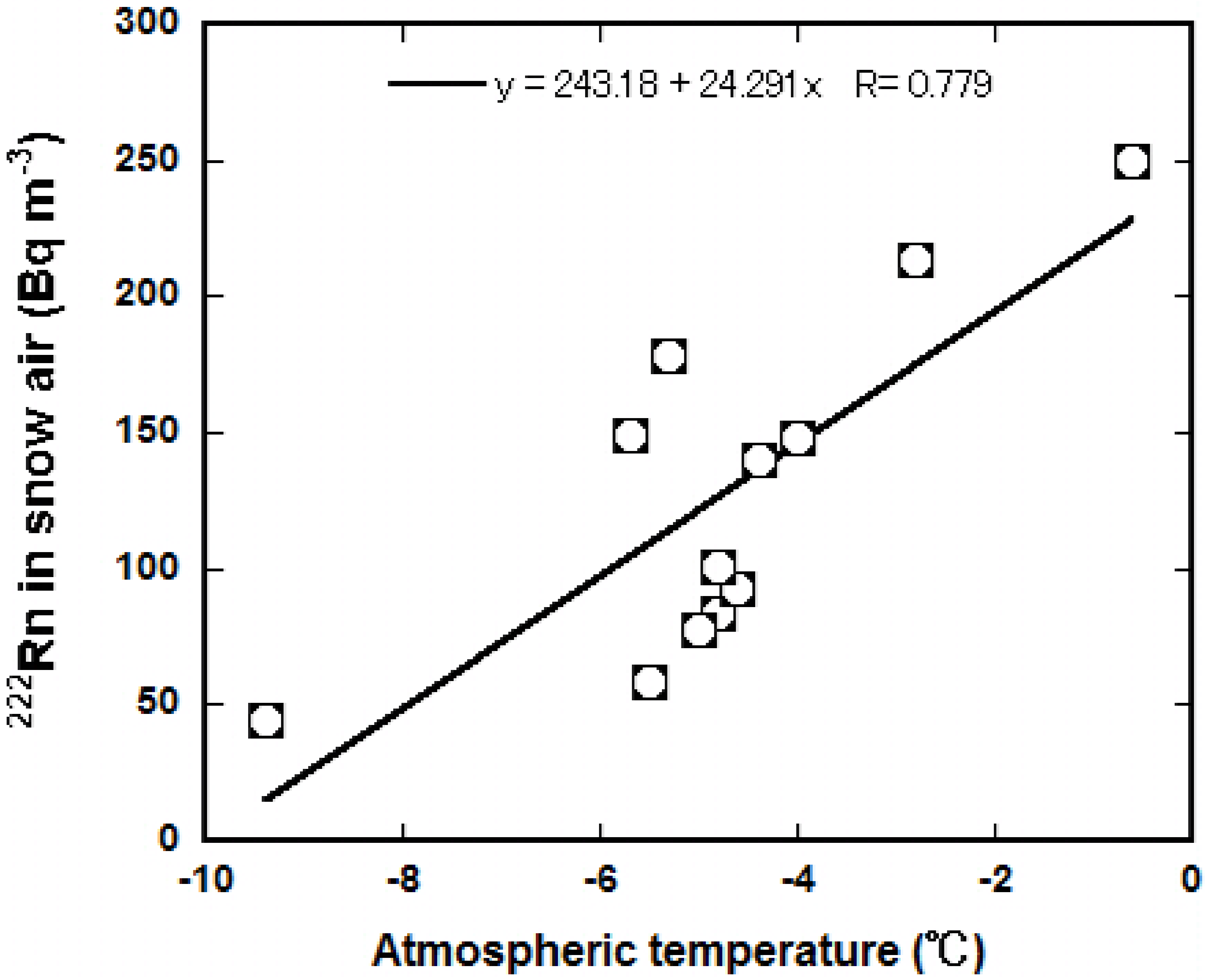
http://www.hokudai.ac.jp



3000
2500
2000
1500
1000
500
0
12
10
8
6
4
2
0
222Rn (Bq m-3)
Soil temperature (ºC)
13/11/2010 00:00
23/11/2010 00:00
3/12/2010 00:00
13/12/2010 00:00
23/12/2010 00:00
2/1/2011 00:00
12/1/2011 00:00
22/1/2011 00:00
1/2/2011 00:00
11/2/2011 00:00
21/2/2011 00:00
3/3/2011 00:00
13/3/2011 00:00
Date



^{222}Rn (Bq m^{-3})
Soil temperature (°C)
Date



y = 243.18 + 24.291x R= 0.779
300
250
200
150
100
50
0
-10
-8
-6
-4
-2
0
222Rn in snow air (Bq m-3)
Atmospheric temperature (℃)

Table 1 Data on surface geology, climate, stand property of the sites (Tomakomai and Sapporo) investigated in this study

Site	Location	Surface Geology	Climate (Annual mean)			Stand description (typical species)
			Temperature (ºC)	Precipitation (mm)	Snow depth February (cm)	
Tomakomai	141.36ºE 42.40ºN	Volcanic ash	6.4	1200	50	Cool temperate deciduous stand (Quercus mongolica var. grosseserrata, Acer sp., Betula sp.)
Sapporo	141.36ºE 42.40ºN	Alluvial sediment	8.9	1100	100	Cool temperate deciduous stand (Populus, Betula sp., Quercus crispula)

Table 2 Properties of soil (dry density, humidity, total porosity, Soil organic matter, pH and ^{226}Ra activity concentration) at the monitoring points in Tomakomai and in Sapporo, Hokkaido, Japan.

Site	Depth (cm)	Dry density (g cm^{-3})	Humidity (%)	Porosity (%)	SOM (%)	pH	^{226}Ra (± uncertainty) (Bq kg^{-1})
Tomakomai							
(2010-2011)	0-2	1.27	56.8	0.74	20.0	4.88	9.9 (1.2)
	2-4	1.38	49.0	0.62	15.4	4.81	10.2 (1.1)
	4-6	1.51	33.1	-	6.2	4.93	9.6 (1.0)
	6-8	1.76	25.1	0.54	3.1	5.12	7.7 (0.8)
	8-10	1.67	19.0	-	1.8	5.27	3.4 (0.5)
	10-12	1.76	18.7	0.39	1.3	5.52	4.7 (0.6)
	12-15	1.98	21.5	0.44	1.5	5.42	5.6 (0.7)
	15-20	1.66	22.3	0.65	0.7	5.36	8.0 (0.9)
	20-25	1.86	19.9	0.73	0.5	5.58	4.0 (0.5)
	25-30	1.76	19.7	0.67	0.6	5.50	6.7 (0.7)
	30-35	1.67	21.6	0.68	0.6	5.48	7.0 (0.7)
	35-40	1.68	23.0	-	1.1	5.40	5.1 (0.6)
Sapporo							
(2006-2011)	0-2	0.85	33.7	17.8	23.1	5.04	14.2 (1.4)
	2-4	0.95	34.9	16.8	20.5	5.02	18.8 (1.6)
	4-6	0.95	36.0	16.3	20.0	4.99	11.9 (1.5)
	6-8	0.97	33.0	15.8	19.2	4.98	15.0 (1.5)
	8-10	0.99	31.6	14.0	18.1	4.93	16.0 (1.5)
	10-12	1.00	33.4	13.6	17.8	4.94	18.0 (1.5)

	12-15	1.03	32.7	15.8	17.2	4.76	16.6 (1,5)
	15-20	1.03	32.3	16.1	16.0	4.76	17.4 (1.5)
	20-25	1.09	34.2	14.9	15.6	4.70	19.1 (1.6)
	25-30	1.07	33.8	15.6	14.4	4.75	19.2 (1.6)
	30-35	1.16	32.3	16.0	11.9	4.70	19.2 (1.6)
	35-40	1.13	30.5	15.2	9.0	4.79	21.3 (1.9)

Table 3 Meteorological data and results of ^{222}Rn concentration in snow and snow air samples collected in January, 2012 Results of soil radon (^{222}Rn) concentration on the day specified in the table are also included.

Date	Snow depth (cm)	Atmospheric pressure (hPa)	Atmospheric temperature (°C)	^{222}Rn in soil air (Bq m^{-3})	^{222}Rn in snow (Bq m^{-3})*	^{222}Rn in snow air (Bq m^{-3})
Jan. 06	41	1010.1	0.8	3702	13.8	104
10	44	1014.0	-4.0	3210	n.d.	148
11	45	1007.1	-5.3	2903	n.d.	178
12	45	1009.2	-9.4	3026	18.4	44
13	49	1005.6	-4.8	2411	46.0	84
14	50	1013.2	-4.6	2657	50.6	93
15	51	1013.3	-4.8	2534	n.d.	101
16	52	1014.9	-2.8	2349	36.8	213
17	53	1015.1	-4.4	2288	n.d.	140
18	54	1017.9	-0.6	3149	n.d.	250
26	61	1009.7	-.05	2472	46.0	78
27	62	1011.1	-5.7	2288	547.4	149
28	62	1013.8	-5.5	2472	n.d.	58

* The notation (n.d.) in the table denotes to an analytical value lower than the detection limit (< 9.0 Bq m^{-3}) of our liquid scintillation counting system.