CONCENTRATIONS OF ^{222}RN, ITS SHORT-LIVED DAUGHTERS AND ^{212}PB AND THEIR RATIOS UNDER COMPLEX ATMOSPHERIC CONDITIONS AND TOPOGRAPHY

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Abstract. Atmospheric activity concentrations of ^{212}Pb and short-lived ^{222}Rn daughters, together with meteorological elements, have been observed continuously at three sites at Kamisaibara Village in Japan. In addition, atmospheric activity concentration of ^{222}Rn, equilibrium-equivalent concentration of ^{222}Rn and conditions of the lower atmosphere were observed for three intensive observation periods at Akawase, one of the three sites in Kamisaibara Village. The equilibrium-equivalent concentration of ^{222}Rn is almost the same as the atmospheric activity concentration of short-lived ^{222}Rn daughters.

The activity concentrations of ^{212}Pb and the short-lived ^{222}Rn daughters and their ratio were low in the daytime owing to convective mixing, and high at night owing to the surface-based inversion during periods of no precipitation. Their variations have several patterns corresponding to the scale of the drainage wind or weak mixing.

Mechanical mixing due to strong winds through both day and night during the first and second observation periods made the atmospheric activity concentrations of ^{212}Pb and the short-lived ^{222}Rn daughters continuously low. However, their ratios were continuously high during the first period yet continuously low during the second period. This difference can be explained by the effect of

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extraction of ^{220}Rn and ^{222}Rn due to strong winds and snow cover. There were also cases in which the ratio of the atmospheric activity concentration of ^{212}Pb to that of the short-lived ^{222}Rn daughters at night was equal to or less than the ratio in the daytime. This inverse trend, as in the periods of no precipitation mentioned above, is considered to be due to near-neutral conditions on these nights.

We find a difference in the ratio of the equilibrium-equivalent concentration of ^{222}Rn (the activity concentration of short-lived ^{222}Rn daughters) to the activity concentration of ^{222}Rn during the first observation period and that during the second. The difference can be explained by snow cover on the ground. We also find differences among the ratios of the activity concentration of the short-lived ^{222}Rn daughters to that of ^{222}Rn during the three observation periods. These differences can be explained by the submergence of paddy fields.

Keywords: Concentration of ^{212}Pb, Concentration of ^{222}Rn, Concentration of short-lived ^{222}Rn daughters, Snow depth, Submergence of paddy fields.

1. Introduction

Airborne radon (^{222}Rn), thoron (^{220}Rn) and their decay products have been useful as suitable tracers in studies of the atmospheric boundary layer. Many reports have been published relating conditions of the lower atmosphere (especially meteorological elements) and concentration of airborne ^{220}Rn, ^{222}Rn and/or its daughters (e.g., Moses et al., 1963; Pearson and Moses, 1966; Ikebe, 1970; Druilhet and Fontan, 1973; Beck and Gogolak, 1979; Guedalia et al., 1980; Robé et al., 1992). Reports relating meteorological elements and exhalation rate of ^{220}Rn and/or ^{222}Rn from soil have also been published (e.g., Megumi and Mamuro, 1973; Clements and Wilkening, 1974; Guedalia et al., 1970; Schery et al., 1984).

With the above as background, 'A Study of Local Meteorology at Ningyo-toge and Its Vicinity' program was designed in 1994 and performed from 1995 to 1998 as part of 'Monitoring of Radiation around Ningyo-toge Environmental Engineering Center, Japan Nuclear Cycle Development Institute'. This monitoring of radiation has been sponsored by the Japanese Ministry of Education, Culture, Sports, Science and Technology. The focus of the above program was to make clear the relation between atmospheric concentrations of total α-activity (this is due to α-particles from two of ^{220}Rn daughters (^{212}Bi and ^{212}Po) and can be reduced to atmospheric activity concentration of ^{212}Pb, one of the ^{220}Rn daughters) and local meteorology. Included in the program were simultaneous observations of the atmospheric boundary layer and atmospheric activity concentrations of ^{222}Rn and its short-lived daughters at a location near one of the three observatories.

In a previous paper (Kataoka et al., 2000), we simply related time variations of atmospheric activity concentrations of ^{222}Rn, its short-lived daughters and ^{212}Pb and their ratios with some meteorological parameters such as wind speed, net radiative flux and snow depth. Following that early work, we have obtained additional examples including observations of the atmospheric boundary layer, and have used other methods for data analysis. In this paper, we present in detail the atmospheric activity concentrations of ^{222}Rn, its short-lived daughters and ^{212}Pb and their ratios and relate them to conditions within the atmospheric boundary layer.

We use the following terminology. Atmospheric activity concentrations of ^{212}Pb, ^{222}Rn, and of short-lived ^{222}Rn daughters (which is assumed to be in radioactive equilibrium with ^{222}Rn) will hereafter be denoted by, in order, concentrations of ^{212}Pb, ^{222}Rn and ^{222}Rn daughters. Also, the ratio of the concentrations of ^{212}Pb and ^{222}Rn daughters and the ratio of the concentration of ^{222}Rn daughters to the concentration of ^{222}Rn will hereafter be designated as the ^{212}Pb/^{222}Rn daughters ratio and ^{222}Rn daughters/^{222}Rn ratio, respectively. The equilibrium-equivalent concentration of ^{222}Rn (EC_{Rn}) is defined in ICRP (1981) as follows: the equilibrium-equivalent ^{222}Rn concentration of a non-equilibrium mixture of short-lived ^{222}Rn daughters in air is the activity concentration of ^{222}Rn in radioactive equilibrium with its short-lived daughters that has the same potential α-energy concentration as the non-equilibrium mixture to which the equilibrium-equivalent concentration of ^{222}Rn refers. It is almost the same as the concentration of ^{222}Rn daughters (Shimo, 1986) and will hereafter be designated as the concentration of ^{222}Rn daughters (EC_{Rn}) to avoid confusion. Furthermore, the ratio of the concentration of ^{222}Rn daughters (EC_{Rn}) and the concentration of ^{222}Rn will hereafter be designated as the ^{222}Rn daughters (EC_{Rn})/^{222}Rn ratio.

The rate of decay of a radionuclide is described by its activity, namely, by the number of atoms that decay per unit time. The unit of activity is the bequerel (Bq), defined as one disintegration per second: $1\ Bq = 1\ s^{-1}$.

2. Study Sites and Instrumentation

Since 1 April 1993, concentrations of ^{212}Pb and ^{222}Rn daughters have been measured simultaneously every three hours at observatories at three locations in Kamisaibara Village, Okayama Prefecture, Japan (35°18′ N, 133°35′ E). These locations are identified by the letters A, B and C in Figure 1a and are, in order, Tohge, Akawase and Tennoh. Tohge (A in Figure 1a) is at the top of a ridge in the Chuhgoku Mountains and is at an elevation of 740 m above sea level. At Tohge, the observatory is at the north side of a parking zone, which is 75 m long by 60 m wide and is laid with asphalt. Vegetation near this site is *Rubus crataegifolius-Aralia elata* community, *Fagus crenata-Quercus mongolica var. grosseserrata* community, plant communities in clear cut areas and a *Cryptomeria japonica* plantation. Ningyo-toge Environmental Engineering Center, Japan Nuclear Cycle Development Institute is situated close to the parking zone. Its site is about 750 m from east to west and about 500 m from north to south. Akawase (B in Figure 1a) is located in a basin (1200 m by 700 m) at the mid-point of Akawase Valley, and is at an elevation of 710 m. A detailed description of the Akawase site is given in Kataoka et al. (1998). The location of the observatory at Akawase is designated as B1 in Figure 1b (A in Kataoka et al., 1998). Tennoh (C in Figure 1a) is situated at the junction of Ikegoh Valley and Akawase Valley, and has an elevation of 540 m. The axis of this area is oriented from north to south. The floor of the area is 1100 m long by 400 m wide, and falls 240 m per km to the east and 60 m to the south. The observatory at Tennoh is at the northeast side of the area and is surrounded by paddy fields. On both sides of the area are walls having heights of 80–220 m and slopes of 35-37°, whose vegetation consists of plant communities in clear cut areas, *Quercus serrata* community, *Miscanthion sinensis*, a *Cryptomeria japonica* plantation. From the junction of the two valleys, a brook runs along the east side of the area at the base of the wall.

Some uranium deposits exist in this region, especially near Tohge. However, they contain low-grade ores and do not influence the concentration of ^{222}Rn daughters measured at these three sites (Kataoka et al., 1999).

The measurements of ^{212}Pb and short-lived ^{222}Rn daughters have been carried out with devices equipped with two α-scintillation counters. Details of the instruments are given in Kataoka et al. (1996). The reported concentration of ^{212}Pb is the average of a three-hour collection. However, as the effective half-life of short-lived ^{222}Rn daughters is about 40 min, the latter half of the three-hour collection has a major impact on the measurement of the concentration of ^{222}Rn daughters. Meteorological variables such as wind speed and direction have also been observed continuously at the three observatories. Net radiative flux, atmospheric pressure and snow depth have been observed continuously only at the Akawase observatory as a representative of the three sites.

At location B2 in Figure 1b (B in Kataoka et al., 1998), intensive observations of the atmospheric boundary layer were carried out during three periods. The first observation period was from 6 to 13 October 1995, the second was from 27 February to 1 March 1998 and the third was from 22 July to 2 August 1998. These intensive observation periods will hereafter be designated as, in order, IOP-1, IOP-2 and IOP-3. Table I provides a summary of the instrumentation used to obtain the data of ^{222}Rn, its daughters and meteorology during the three periods. The methodology of the measurements was provided in Kataoka et al. (1998).

3. Exhalation Rates of ^{220}Rn and ^{222}Rn and Atmospheric Pressure

Since the variation of atmospheric pressure was not large (the variation was 6 hPa) in work presented in a previous paper (Kataoka et al., 1998), exhalation rates of ^{220}Rn and ^{222}Rn were treated as constant. However, in this paper, we are concerned with the concentrations of ^{222}Rn, its daughters and ^{212}Pb (one of the daughters of ^{220}Rn) during a period in which the variation of atmospheric pressure is relatively large. We discuss here the influence of atmospheric pressure on exhalation rates of ^{220}Rn and ^{222}Rn.

Reports concerning the relation between exhalation rates of ^{220}Rn and/or ^{222}Rn and atmospheric pressure include the following: Guedalia et al. (1970), Clements

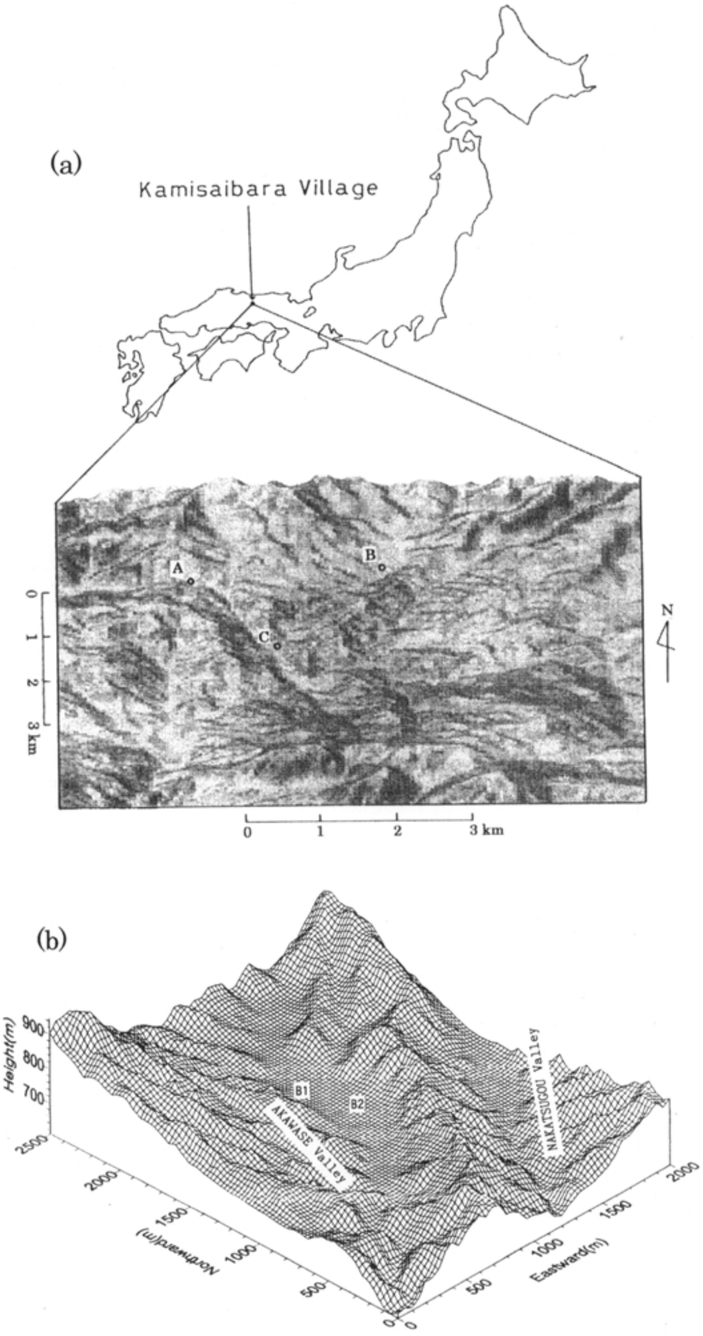

Figure 1. Topographic map around (a) three observatories (A: Tohge observatory, B: Akawase observatory, C: Tennoh observatory) and (b) Akawase observatory (B1: Akawase observatory, B2: A site where three intensive observations were carried out).

TABLE I

Instrumentation for measurement at Akawase (Location B2).

Instrumentation	Objective of measurement	Intense observation period		
		First (IOP-1) 6–13 October 1995	Second (IOP-2) 27 February–1 March 1998	Third (IOP-3) 22 July–2 August 1998
^{222}Rn monitor	Concentration of ^{222}Rn	○	○	○
WL monitor	Equilibrium-equivalent concentration of ^{222}Rn	○	○	×
ER system	Exhalation rate of ^{222}Rn	○	×	○
Low-level sonde	Vertical profile of meteorological elements up to about 1000 m	○	△	○
Tethersonde system	Vertical profile of meteorological elements up to about 600 m	○	×	○
Monostatic sodar	Inversion layer up to 400 m	○	×	×
Doppler sodar	Continuous wind profile up to 600 m	×	×	○
Surface eddy flux system	Sensible and latent heat fluxes at ground level	○	×	○
Infrared thermal imagery	Profile of slope temperature of basin floor	○	×	△
Meteorological observation station	Meteorological elements at ground level	○	×	○

○: Data were obtained for more than half of the period.
△: A few data were obtained.
×: No data were obtained.

and Wilkening (1974), Schery et al. (1984), Ishimori et al. (1998). We describe the relation between the exhalation rate of ^{222}Rn and atmospheric pressure firstly and the relation between the exhalation rate of ^{220}Rn and atmospheric pressure next. The experiments of Clements and Wilkening (1974) and Schery et al. (1984) were carried out at sites about 1 and 2 km west of the centre of the New Mexico Institute of Mining and Technology campus, respectively. Although soils at the two sites are slightly different, they are gravelly sandy loam, with a water table about 30 m below the surface. Since the climate is semi-arid (rainfall averages 200 mm y^{-1}), soil moisture is low. In accordance with Clements and Wilkening (1974), when changes in the atmospheric pressure are $+14.5$ hPa and -15.6 hPa after being relatively constant for at least 24 h, changes in the exhalation rates of ^{222}Rn are about -40% and about $+60\%$, respectively. A report by Schery et al. (1984) shows that, when the change in atmospheric pressure is slow and small, the exhalation rate of ^{222}Rn can be treated as constant, but that, when the change in atmospheric pressure is sharp or large, the exhalation rate of ^{222}Rn cannot be considered constant.

At our site, volcanic ash soil exists over decomposed granite soil, and the thickness of the soil layers varies within the experimental area. This pattern is typical, although there are several spots having sedimentary rocks of the Tertiary period. Since the depth of any private-dug well is 2–3 m, it is considered that there are shallow water veins in the area. The climate is humid temperate (rainfall averages 2600 mm y^{-1}). The rainfall and shallow water veins being taken into consideration, soil moisture is likely to be substantially higher at our experimental area than that at the sites used by Clements and Wilkening (1974) and Schery et al. (1984). Further, the soil in our area is also different from that at the sites used in the referenced studies. Therefore, the relation between the exhalation rate of ^{222}Rn and atmospheric pressure obtained by Clements and Wilkening (1974) and Schery et al. (1984) cannot be directly adopted here.

According to Ishimori et al. (1998), the exhalation rate of ^{222}Rn varies within $\pm 12\%$ and is considered to be constant during the period of 2–9 October 1994. The site used by Ishimori et al. (1998) is about 200 m east-northeast of the Tohge observatory. The height of the site above the sea level is almost the same as the height of the Akawase observatory. Therefore, the change in the atmospheric pressure is almost the same as that at Akawase observatory, which was 13 hPa for this period, with no rain. Therefore, the exhalation rate of ^{222}Rn can be assumed to be only weakly dependent on atmospheric pressure in the case of no rain, for the site and periods of data collection. As such we consider the exhalation rate to be dependent upon processes other than atmospheric pressure.

Atmospheric pressure varied between 934 and 945 hPa, between 928 and 939 hPa and between 927 and 938 hPa during the three periods of data analysis, from 7 to 18 October 1995, from 27 February to 1 March 1998 and from 22 July to 2 August 1998, respectively, as shown in Figure 2. Therefore, it is reasonable that the exhalation rate of ^{222}Rn is independent of the atmospheric pressure within each period for the reason above.

Table II gives exhalation rates of ^{222}Rn measured for the IOP-1 and IOP-3 at Akawase. The uncultivated lands (1, 2 and 3 in the table) are within 20 m of one another; the Akawase observatory is also within 20 m. Since the exhalation rates of ^{222}Rn were measured more than two days after rainfall, they are not influenced by the precipitation. Measurements of the ^{222}Rn exhalation rate were carried out on 9 and 12 October 1995 for the IOP-1. The difference in the atmospheric pressure was 3 hPa between the two measurements of the ^{222}Rn exhalation rate during the IOP-1. Errors being taken into account, the exhalation rates of ^{222}Rn agree at each point. The atmospheric pressure at 0503–0616 JST on 2 August 1998 was lower than that at 1352–1437 JST on 9 October 1995 by 3 hPa and was lower than that at 1108–1147 JST on 12 October 1995 by 6 hPa. The above explanation being taken into account, the difference between the ^{222}Rn exhalation rates at the uncultivated lands 1, 2 and 3 is due to the fact that they are different sites. Furthermore, the average of the ^{222}Rn exhalation rate at the uncultivated land 1 is about one third of the average of those at the paddy fields and at the ridge between the paddy fields on 9 and 12 October 1995. In addition, the average of the ^{222}Rn exhalation rates at the uncultivated lands 2 and 3 is also about one third of that at the forest area (uncultivated land 4 in Table II) on the opposite side of the paddy fields on 2 August 1998. Hence, it is reasonable to assume that the exhalation rate of ^{222}Rn at the area comprising the uncultivated lands 1, 2 and 3 near Akawase observatory is approximately one third of those at the paddy area and the forest area on the opposite side of the paddy fields during the three observation periods.

Next, we describe the relation between atmospheric pressure and the exhalation rate of ^{220}Rn. Guedalia et al. (1970) indicate that the exhalation rate of ^{220}Rn is independent of atmospheric pressure. Schery et al. (1984) showed that the exhalation rate of ^{220}Rn exhibited only one-fourth of the amplitude of ^{222}Rn for the same variation of atmospheric pressure. From these two facts and very weak pressure-dependence of the ^{222}Rn exhalation rate at our experimental area as mentioned above, it is reasonable that the exhalation rate of ^{220}Rn is also independent of atmospheric pressure at the area.

4. Concentrations of ^{212}Pb and ^{222}Rn Daughters and their Ratio

4.1. A STANDARD NORMAL PERIOD – CYCLE OF CONVECTIVE MIXING AND SURFACE-BASED INVERSION

In a previous paper (Kataoka et al., 2000), we briefly introduced the time variation of concentrations of ^{212}Pb and ^{222}Rn daughters and ^{212}Pb/^{222}Rn daughters ratio during the IOP-1. In the paper, we showed a trend such that the concentrations and the ratio were low in the daytime and high at night, except for a day having precipitation and for a cloudy day. Adding data having the same trend, we show here

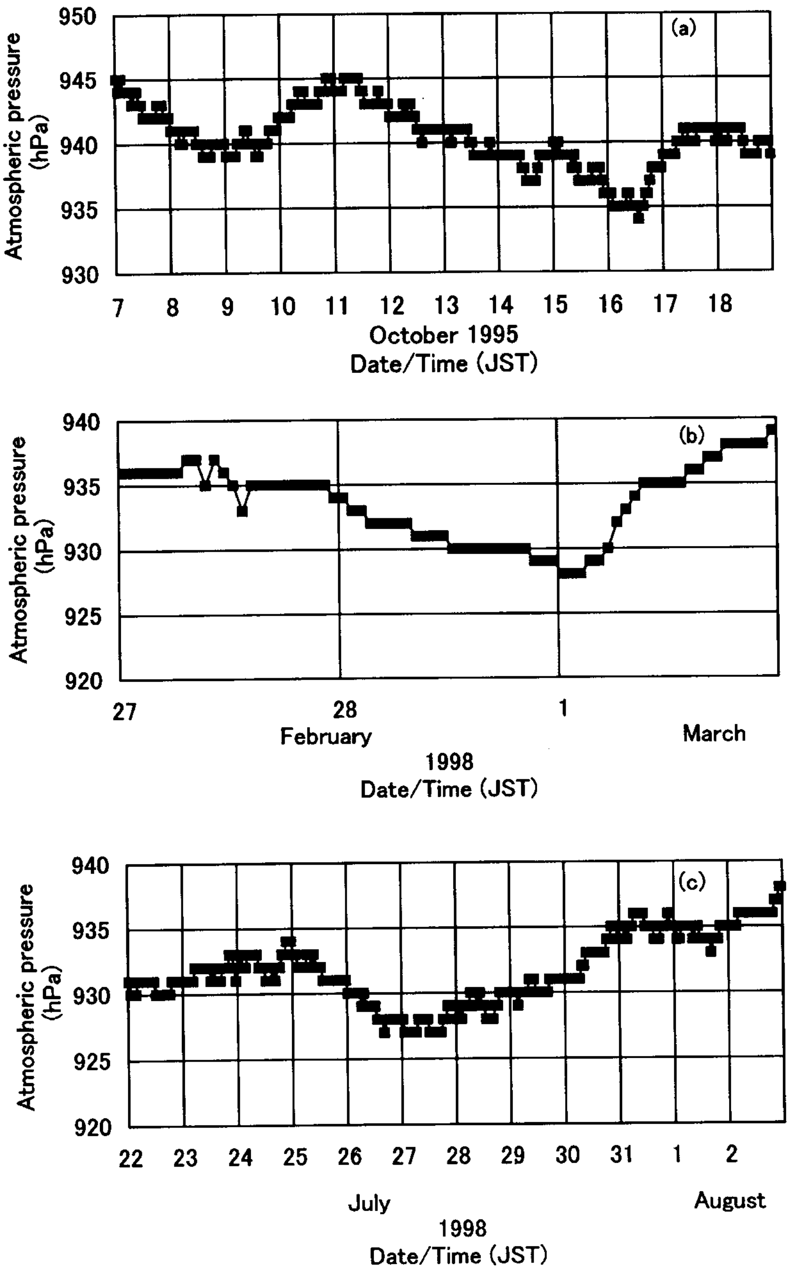

Figure 2. Time variation of atmospheric pressure for the periods (a) from 7 to 18 October 1995, (b) from 27 February to 1 March 1998 and (c) from 22 July to 2 August 1998.

TABLE II
Exhalation rate of ^{222}Rn.

Period	Soil type	Date/Time (JST)	Exhalation rate of ^{222}Rn (Bq m^{-2} s^{-1})	Atmospheric pressure (hPa)
First period	Paddy field	1352–1423, 9 October 1995	0.0103 ± 0.0016	939
		1120–1151,12 October 1995	0.0097 ± 0.0014	942
	Ridge between paddy fields	1400–1431, 9 October 1995	0.0187 ± 0.0018	939
		1116–1147, 12 October 1995	0.0209 ± 0.0017	942
	Uncultivated land 1	1406–1437, 9 October 1995	0.0063 ± 0.0009	939
		1108–1139, 12 October 1995	0.0055 ± 0.0015	942
Third period	Uncultivated land 2	0546–0616, 2 August 1998	0.0043 ± 0.0004	936
	Uncultivated land 3	0503–0533, 2 August 1998	0.0035 ± 0.0004	936
	Uncultivated land 4	0628–0658, 2 August 1998	0.0139 ± 0.0010	936

time variations of the concentrations of ^{212}Pb and ^{222}Rn daughters and ^{212}Pb/^{222}Rn daughters ratio at the three sites for the period of 7–18 October 1995 in Figure 3.

Kataoka et al. (1999) showed statistically that the levels of the concentrations of ^{212}Pb and ^{222}Rn daughters and ^{212}Pb/^{222}Rn daughters ratio at the three sites differ from each other. This reveals the difference of the ^{220}Rn and ^{222}Rn exhalation rates between the three sites. Time variations of the concentrations of ^{212}Pb and ^{222}Rn daughters, and of the ^{212}Pb/^{222}Rn daughters ratio indicate almost the same tendency for the three sites. We notice that the ^{212}Pb/^{222}Rn daughters ratio is small in the daytime when a convective mixing layer develops over the experimental area, while the ratio is large in the nighttime when a surface-based inversion layer develops. The decrease of the ^{212}Pb concentration with height is larger than that of the short-lived ^{222}Rn daughters over the duration of the surface-based inversion layer due to the difference in their half-lives (^{212}Pb: 10.64 h, ^{222}Rn: 3.82 days). Mixing after sunrise may cause the ^{212}Pb/^{222}Rn daughters ratio to decrease. During the daytime, the mixed-layer depth is mainly controlled by thermal convection and mechanical mixing. Thermal convection can be represented by solar radiation or simply, cloud amount, while, mechanical mixing is represented by wind speed. On sunny and windy days, the mixed layer develops to higher altitudes. At night, cloud amount and wind speed are both controlling factors for the development of a surface-based inversion. Unlike the daytime situation, high wind speeds lead to suppression of the inversion due to forced mixing. A well developed surface-based inversion can be observed during a clear and calm night. Diurnal ranges of the variations of the concentrations of ^{212}Pb and ^{222}Rn daughters and the ^{212}Pb/^{222}Rn daughters ratio at the three sites differ from each other. However, it can be summarized that they are small in the daytime and large at night. The cycle of this pattern is considered to be typical for continuous days with no rain.

Examining thoroughly the time variations of the concentrations of ^{212}Pb and ^{222}Rn daughters and ^{212}Pb/^{222}Rn daughters ratio for periods with no rainy days, we note that they have several different patterns at night. Examples of the patterns are picked up from the periods 7–18 October 1995 and 16–26 October 1997, and are shown in Table III. Since the examples are obtained more than two days after rain, and since the variation of the atmospheric pressure for each period is within 13 hPa, it may be assumed that the exhalation rates of ^{220}Rn and ^{222}Rn are constant. The observatory at Tohge is at the pass, and the observatories at Akawase and Tennoh are in the basin. The observation of the atmospheric boundary layer, including the measurement of ^{222}Rn, was carried out at the basin (Akawase), but was not done at the pass. Therefore, we here narrow down discussions to Akawase and Tennoh and leave the discussion about Tohge for the future.

Let us discuss each case from the top of Table III based on the accumulation of ^{212}Pb, ^{222}Rn and its daughters near the ground, the decrease of their concentrations with height, the surface-based inversion, and the mixing condition. The concentrations of ^{212}Pb and ^{222}Rn daughters, and ^{212}Pb/^{222}Rn daughters ratio, continued to increase from 1800 JST on 20 October to 0600 JST on 21 October 1997. This is a

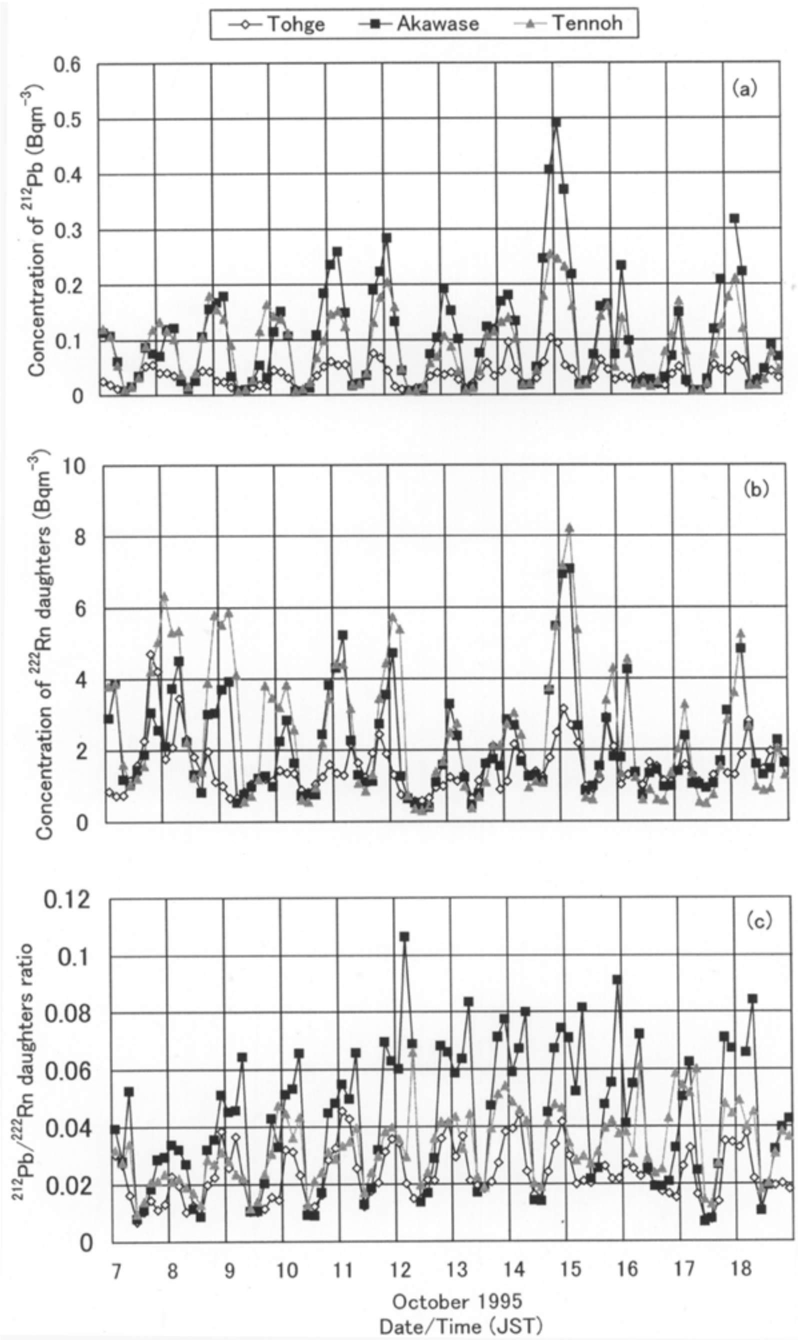

Figure 3. Time variations of (a) the concentration of ^{212}Pb, (b) the concentration of ^{222}Rn daughters and (c) the ^{212}Pb/^{222}Rn daughters ratio, at three sites for the period from 7 to 18 October 1995.

TABLE III

Changes of the concentrations of ^{212}Pb and ^{222}Rn daughters and ^{212}Pb/^{222}Rn daughters ratio and meteorological conditions.

Period	Location	Case			Conditions
		^{212}Pb concentration	Concentration of ^{222}Rn daughters	^{212}Pb/^{222}Rn daughters ratio	
1800 JST 20 Oct.–0600 JST 21 Oct. 1997	Akawase	+	+	+	Surface-based inversion
0300–0600 JST 11 Oct. 1995	Akawase	+	+	−	Surface-based inversion
0000–0300 JST 12 Oct. 1995	Akawase	+	+	−	Surface-based inversion
2100–2400 JST 11 Oct. 1995	Akawase	+	+	−	Middle accumulation Small-scale of drainage wind
0300–0600 JST 12 Oct. 1995	Akawase	−	−	+	Large accumulation Strong surface-based inversion Large-scale of weak mixing
0300–0600 JST 12 Oct. 1995	Tennoh	−	−	−	Large accumulation Small-scale of weak mixing
2100–2400 JST 9 Oct. 1995	Akawase	−	−	−	Poor accumulation Drainage wind
0300–0600 JST 25 Oct. 1997	Akawase	−	−	−	Large accumulation Weak surface-based inversion Large-scale of drainage wind
0300–0600 JST 14 Oct. 1995	Akawase	+	−	+	Intermediate-scale of drainage wind or weak mixing
2100–2400 JST 21 Oct. 1997	Tennoh	+	−	+	Intermediate-scale of drainage wind or weak mixing
0300–0600 JST 15 Oct. 1995	Akawase	−	+	−	Intermediate-scale of drainage wind or weak mixing
0300–0600 JST 25 Oct. 1997	Tennoh	−	+	−	Intermediate-scale of drainage wind or weak mixing

+: Increase, −: Decrease.

good example to indicate the accumulation of ^{212}Pb, ^{222}Rn and its daughters near the ground by a surface-based inversion.

The concentrations of ^{212}Pb and ^{222}Rn daughters increased but the $^{212}Pb/^{222}Rn$ daughters ratio decreased at Akawase during the periods of 0300–0600 JST on 11 October and 0000–0300 JST on 12 October 1995. The ^{222}Rn concentration increased less during the period of 0255–0355 JST on 11 October and much less during the period of 0055–0155 JST on 12 October 1995 (Kataoka et al., 1998). Surface-based inversion layers were caught by a sodar during these two periods. Since the ^{222}Rn concentrations increased, there must have existed surface-based inversion layers lower than 25 m, which is the lowest level of detection by the sodar. To our deep regret, it is difficult to explain why the smaller increases happened during the two nights using the meteorological data simultaneously obtained. The concentrations of ^{212}Pb and ^{222}Rn daughters and $^{212}Pb/^{222}Rn$ daughters ratio during 2100–2400 JST on 11 October 1995 behaved the same as those during the two periods. However, the ^{222}Rn concentration decreased a little during the period of 2155–2255 JST on 11 October (Kataoka et al., 1998). The sodar caught a surface-based inversion layer during the period. The analysis of the meteorological data indicates that a small-scale drainage wind occurred. The decrease of the ^{222}Rn concentration may have been provoked by the small-scale drainage wind.

^{222}Rn, its daughters and ^{212}Pb accumulated near the ground due to a surface-based inversion by 0300 JST on 12 October 1995. Cloud cover at Akawase for the period of 0300–0530 JST on 12 October resulted in a net radiative flux that was less negative than usual and an atmospheric surface layer that was less stable than normal. Combined with slightly higher than normal wind speeds for the period of 0400–0600 JST, this is likely to have resulted in weak mixing, and the observed decreases in concentrations of ^{212}Pb and ^{222}Rn daughters. The scale of the weak mixing is presumed to be fairly large from the large decrease of the concentrations of ^{212}Pb and ^{222}Rn daughters. This weak mixing also produced an extremely large value of the $^{212}Pb/^{222}Rn$ daughters ratio at 0600 JST. At the same time, at Tennoh, the concentration of ^{212}Pb decreased by 25% while the concentration of ^{222}Rn daughters decreased by only 5%. Considering the accumulation of ^{212}Pb and short-lived ^{222}Rn daughters near the ground, the mixing is likely to have been much weaker than at Akawase. This resulted in a more normal value to the $^{212}Pb/^{222}Rn$ daughters ratio at Tennoh.

In the period immediately after the formation of the surface-based inversion layer, ^{222}Rn, its daughters and ^{212}Pb are not greatly accumulated near the ground. Therefore, the decrease of the concentration of ^{212}Pb with height is not much larger than that of the ^{222}Rn daughters. Weak mixing or drainage winds may decrease the $^{212}Pb/^{222}Rn$ daughters ratio, since the difference of the concentrations of ^{212}Pb and ^{222}Rn daughters with height is small. The case of 2100–2400 JST on 9 October 1995 at Akawase (see Figure 3) is an example of a drainage wind situation.

After sufficient accumulation of ^{222}Rn, its daughters and ^{212}Pb near the ground surface, both concentrations of ^{212}Pb and ^{222}Rn daughters and their ratio decrease considerably due to developed mixing. The case of 0300–0600 JST on 25 October 1997 at Akawase corresponds to this phenomenon; strong winds were observed during the period of 0300–0500 JST. During the nighttime, the net radiative flux ranged from -72 to -78 W m^{-2}, under clear skies and large radiative cooling. Under such conditions, the strong wind is symptomatic of a drainage wind. With the degrees of decrease of the concentrations with time taken into account, this phenomenon is considered to be almost the same scale as that at Akawase for the period of 0300–0600 JST on 12 October 1995. However, the ^{212}Pb/^{222}Rn daughters ratio of the former decreased while that of the latter increased. This may be due to the difference between the two cases in the vertical profiles of the concentrations of ^{212}Pb and ^{222}Rn daughters provoked by the formation of the surface-based inversion layer. At Tennoh, moderate winds blew from 0300 to 0600 JST on 25 October (this case will be mentioned in the next paragraph).

The other two cases happened at night according to the degrees of decrease of the concentrations of ^{212}Pb and ^{222}Rn daughters with height and the scale of mixing. One is a case in which the concentration of ^{212}Pb increased and the concentration of ^{222}Rn daughters decreased (0300–0600 JST on 14 October 1995, Akawase; 2100–2400 JST on 21 October 1997, Tennoh). The other is a case in which the concentration of ^{212}Pb decreased and the concentration of ^{222}Rn daughters increased (0300–0600 JST on 15 October 1995, Akawase; 0300–0600 JST on 25 October 1997, Tennoh). There might have existed an intermediate scale of drainage wind or weak mixing during the period of these four events. However, since observations of the atmospheric boundary layer were not carried out during these periods, it is difficult to determine the meteorological cause of these concentration changes.

The patterns of the concentrations of ^{222}Rn and ^{222}Rn daughters (EC_{Rn}) (Kataoka et al., 2000) measured at location B2 for the IOP-1 are similar to the concentration of ^{222}Rn daughters for the same period shown in Figure 3b. We discuss their relations in detail in Section 5.

4.2. Periods of Meteorological and Surface Conditions Differing from a Standard Normal Period

Figure 4 shows time variations of the concentrations of ^{212}Pb and ^{222}Rn daughters and ^{212}Pb/^{222}Rn daughters ratio at the three sites during the IOP-3. The time variation of the concentration of ^{222}Rn measured at location B2 is also shown in the figure. Although the concentration of ^{222}Rn was measured at the submerged paddy field, its pattern of variation resembles that of the concentration of ^{222}Rn daughters (Figure 3b) measured at location B1. However, since further analysis of the data shows the effect of submergence, we describe the effect in Section 5.2.

The observation period has two interesting features. One is that the concentrations of ^{222}Rn, its daughters and ^{212}Pb are low through the day and night during the periods from 0800 JST on 24 July to 1800 JST on 27 July and from 1000 JST

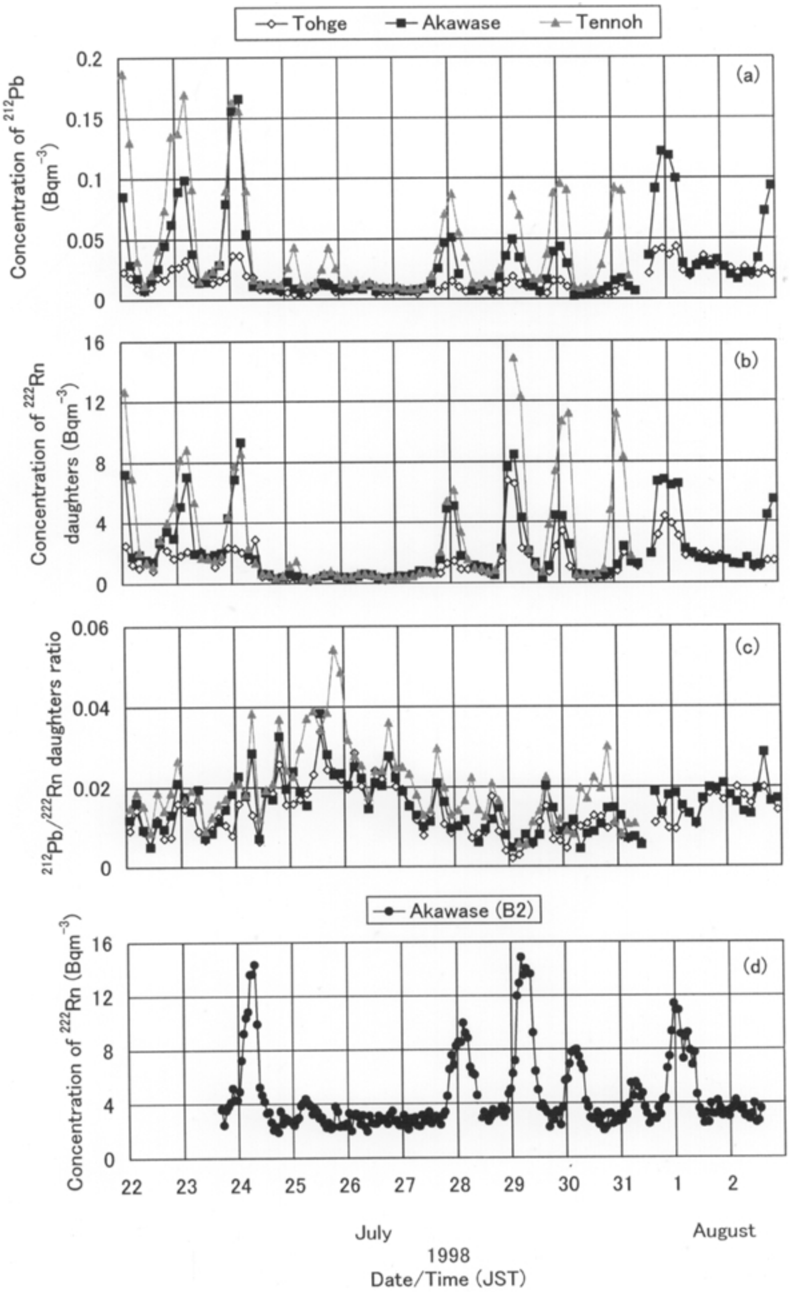

Figure 4. Time variations of (a) the concentration of ^{212}Pb, (b) the concentration of ^{222}Rn daughters, (c) the ^{212}Pb/^{222}Rn daughters ratio at three sites and (d) the concentration of ^{222}Rn at Akawase (location B2) for the period from 22 July to 2 August 1998.

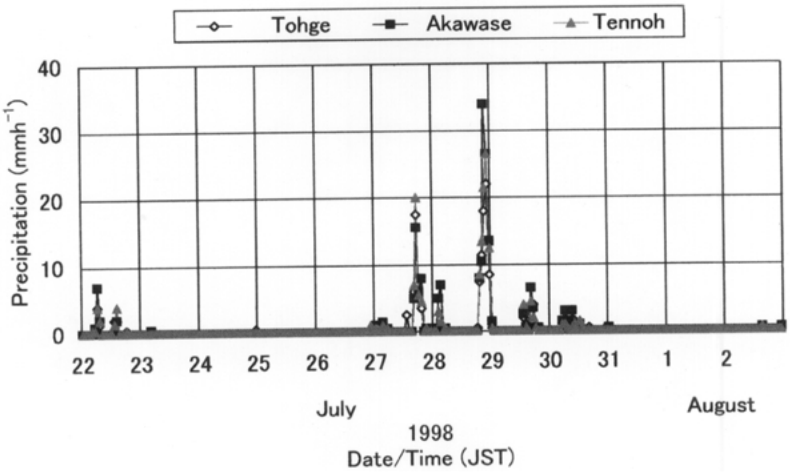

Figure 5. Time variations of precipitation at three sites for the period from 22 July to 2 August 1998.

on 1 August to 1400 JST on 2 August. Another is that the ^{212}Pb/^{222}Rn daughters ratios at night during the period from 27 to 31 July are equal to or less than those in the daytime. This trend is the inverse of the situation in which the ratios are low in the daytime and high at night, as in the standard normal period. We explain these phenomena respectively in the following (Sections 4.2.1 and 4.2.2).

Furthermore, the IOP-2 with a snow depth of 0.2–0.3 m is interesting. Since we discussed the time variation of the concentrations of ^{222}Rn daughters and ^{212}Pb and ^{212}Pb/^{222}Rn daughters ratio in detail for this period previously (Kataoka et al., 2000), we present only a summary in the following (Section 4.2.3).

4.2.1. *Period of Continuous Low Concentrations of ^{222}Rn Daughters and ^{212}Pb*

Figure 5 shows the occurrence of precipitation for the IOP-3. At Akawase, the amounts of rainfall were 10 mm, 2 mm and 0.5 mm during the periods of 0500–0800 JST and 1400–1500 JST on 22 July, and 0400–0500 JST on 23 July, respectively. There was no rain at that location from 0500 JST on 23 July to 2400 JST on 26 July. Precipitation at Tohge and Tennoh was almost the same as that at Akawase. The exhalation rate of ^{222}Rn largely recovers one day after rain (Ishimori et al., 1998), whilst the exhalation rates of ^{220}Rn and ^{222}Rn reveal almost the same trends of recovery after rain (Megumi and Mamuro, 1973). Hence, the continuously low concentrations of ^{212}Pb and ^{222}Rn daughters and the continuously high ^{212}Pb/^{222}Rn daughters ratios for the period of 0800 JST on 24 July to 2400 JST on 26 July at the three sites are considered to be due to meteorological conditions other than precipitation.

Strong winds blew continuously from 0800 JST on 24 July to 1800 JST on 27 July because of a stationary front near the Japanese Island. In the daytime of 24 and 25 July, mixing layers developed due to the strong winds and intense solar radiation. In the daytime of 26 July, the strong winds again led to the development of a mixing layer. During the nighttimes of 24–25 and 25–26 July, the net radiative flux was small and negative, and the wind was strong. Therefore, it is likely that a mixing layer existed during the nighttime. The wind was weak at Tennoh for the periods of 0100–0500 JST and 1800–2100 JST on 25 July, and a surface-based inversion layer is considered to have formed for these two short periods. This is supported by the increase in the concentrations of ^{212}Pb and ^{222}Rn daughters.

On 27 July, rainfall amounted to 4 mm from 0000 to 0600 JST at Akawase and 2.5 mm from 0000 to 0600 at Tennoh. The strong winds continued for the period of 0800 JST on 24 July to 1800 JST on 27 July. Therefore, the low concentration of ^{212}Pb and ^{222}Rn daughters may have been caused by the strong winds and rainfall (mainly the strong winds) from 0000 to 1800 JST on 27 July.

As mentioned in Section 4.1, a cycle such that the ^{212}Pb/^{222}Rn daughters ratio is large in the nighttime and small in the daytime was repeated in the case of the continuous days with no rainy and no cloudy days. In the case of the continuous windy days, the ^{212}Pb/^{222}Rn daughters ratio may be conjectured to retain its daytime value. However, in reality, the ^{212}Pb/^{222}Rn daughters ratios were larger than those of the daytime. This shows that ^{220}Rn is mixed up more from the ground into the atmosphere by the strong winds than is ^{222}Rn. Therefore, the ^{212}Pb/^{222}Rn daughters ratio, which normally decreases by mixing, may have increased due to the extraction of ^{220}Rn and ^{222}Rn from the ground. Furthermore, the fact that the ^{212}Pb/^{222}Rn daughters ratio increased to a slightly larger value and became almost constant, indicates that the effect of the extraction of ^{220}Rn and ^{222}Rn by the strong winds and the convective mixing became stationary.

The observations with the Doppler sodar as shown in Figure 6 indicate upper level strong winds from 0800 JST on 24 July to 1800 JST on 27 July. The wind was moderate (0.5–2 m s^{-1}) near the ground from the night of 1 August to the daytime of 2 August, but the wind in the upper air was not strong, as shown in Figure 7. Under these conditions, the concentrations of ^{212}Pb and ^{222}Rn daughters did not increase and the ^{212}Pb/^{222}Rn daughters ratio remained at a middle value (about 0.02) during the period from the night of 1 August to the daytime of 2 August.

4.2.2. *Period of Low ^{212}Pb/^{222}Rn Daughters Ratio at Night*

We note from Figures 4 and 5 that the period exhibiting the inverse trend of the ^{212}Pb/^{222}Rn daughters ratios (27–31 July 1998), when the ratios at night are equal to or less than those in the daytime, corresponds to a period of rainfall. With this in mind, the inverse trend is thought to be caused by the following possible factors: (1) a variation of exhalation rates of ^{220}Rn and ^{222}Rn from the ground due to the permeation of rain water into the soil, (2) washout of ^{212}Pb and ^{222}Rn daughters from the atmosphere by precipitation, (3) a degree of mixing by winds simultaneously occurring with precipitation, and (4) an increase or decrease of the exhalation rates of ^{220}Rn and ^{222}Rn due to a change in atmospheric pressure. We examine these factors in order as follows:

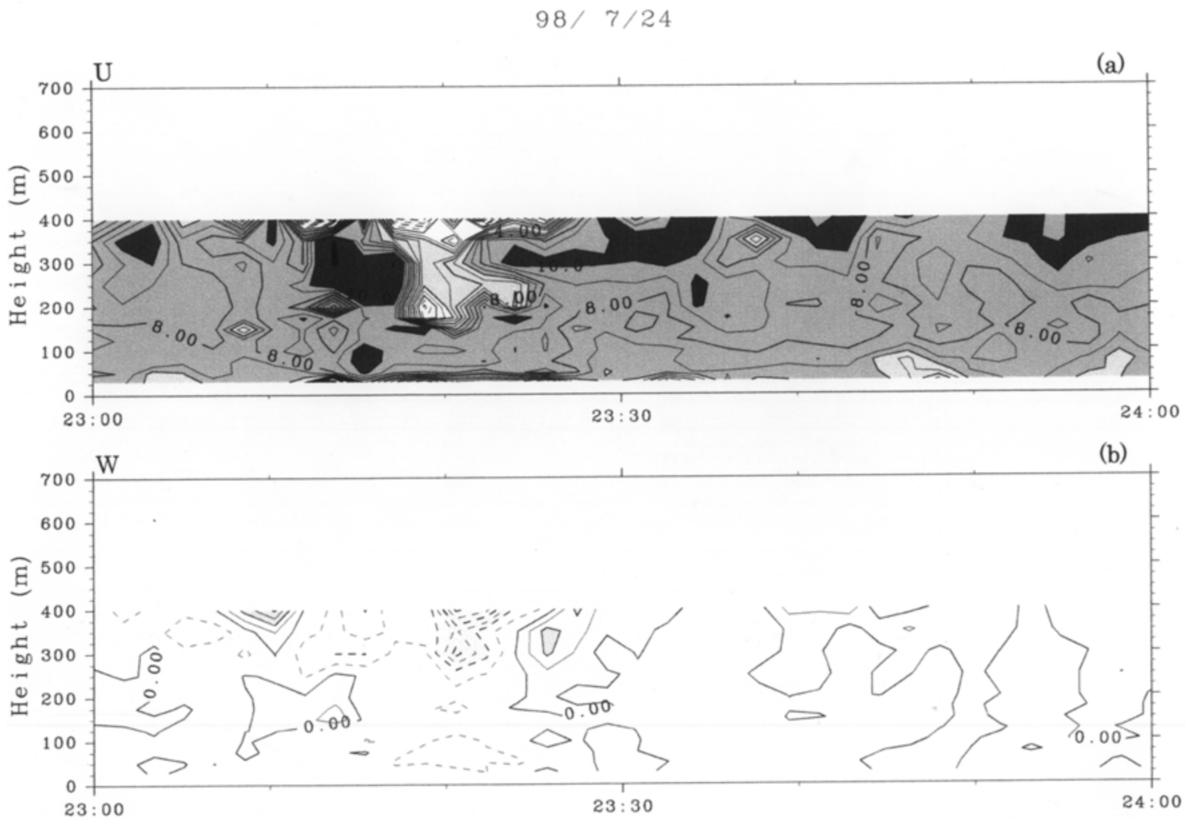

Figure 6. An example of vertical and temporal variations of (a) horizontal component (U) and (b) vertical component (W) of wind speed observed by Doppler sodar on 24 July 1998.

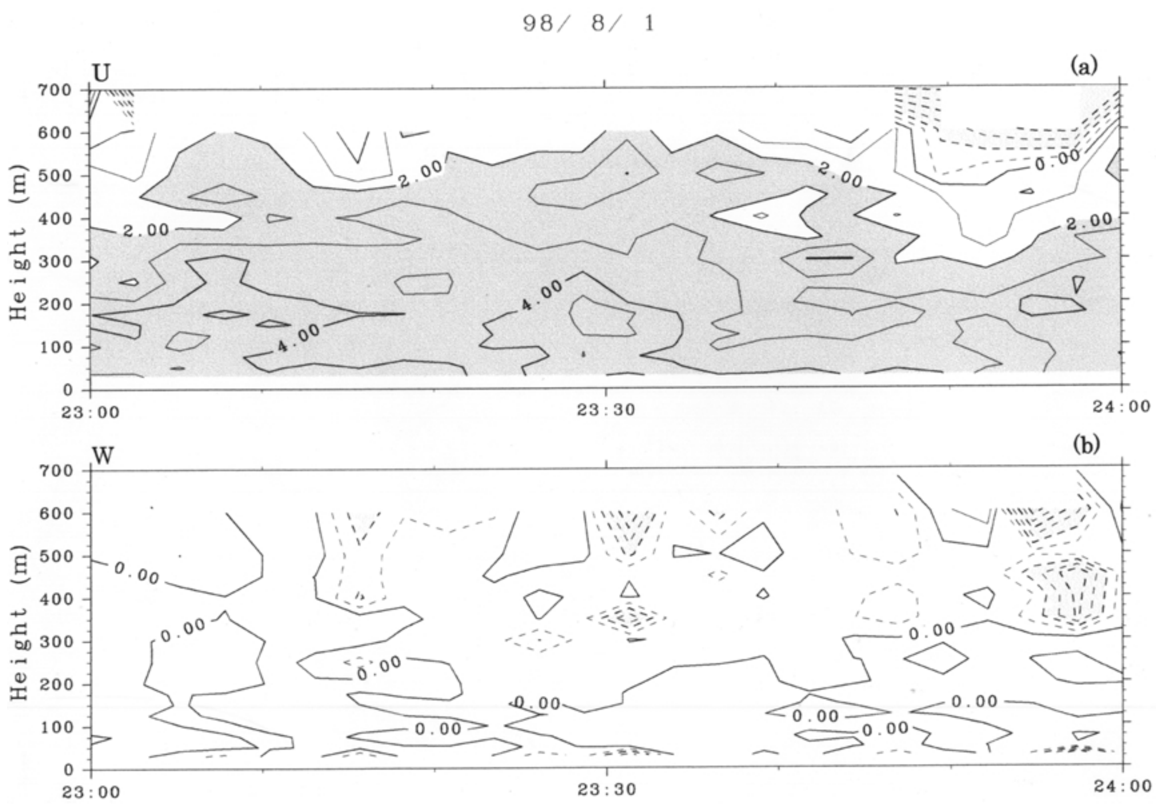

Figure 7. An example of vertical and temporal variations of (a) horizontal component (U) and (b) vertical component (W) of wind speed observed by Doppler sodar for the night of 1 August 1998.

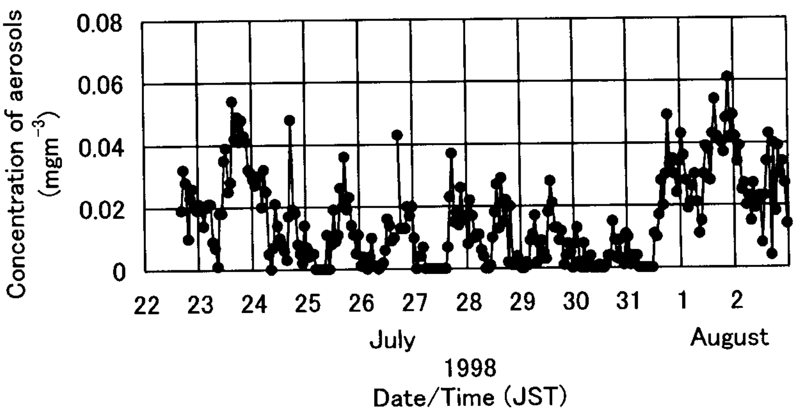

Figure 8. Time variation of the concentration of aerosols.

1. According to Megumi and Mamuro (1973), Ishimori et al. (1998) and Koarashi et al. (2000), the exhalation rates of ^{220}Rn and ^{222}Rn considerably decrease due to rain. Although the ^{220}Rn concentration in soil air becomes constant at a much shallower depth than does the ^{222}Rn concentration in soil air (Israël, 1962), the exhalation rate of ^{220}Rn decreases and recovers, having almost the same trend as that of ^{222}Rn (Megumi and Mamuro, 1973). Therefore it is difficult to believe that this is the reason for the inverse trend of the ^{212}Pb/^{222}Rn daughters ratio at night.
2. In Figure 8, we show the time variation of the concentration of aerosols at Akawase (location B2) for the IOP-3. Since the concentration of aerosols was lower during the rainy period than that with very little or no rain, as shown in Figures 5 and 8, some of the aerosols may have been eliminated from the atmosphere due to the rain. It is reported that rainout (the short-lived ^{222}Rn daughters attach to the raindrops during the process of formation of the raindrops in the upper air) is much more important than washout (the raindrops collide with the aerosols containing the short-lived ^{222}Rn daughters) (Jacobi, 1961; Bhandari and Rama, 1963)). Therefore it is difficult to evaluate the short-lived ^{222}Rn daughters removed by washout using the ground-based measuring system. According to Jacobi (1961), the change in the concentration of ^{222}Rn daughters in the atmosphere by washout is at most 5%. The rate of elimination of ^{220}Rn daughters by washout may be almost the same as that of the ^{222}Rn daughters. Even if it is much different, the half-lives of ^{220}Rn, ^{222}Rn and their daughters being taken into account, the decrease of the concentrations of ^{212}Pb and ^{222}Rn daughters recover near the ground within a few hours. Therefore it is unlikely that washout is the cause of the inverse trend of the ^{212}Pb/^{222}Rn daughters ratio at night.
3. From the night of 27 July to the dawn of 31 July, the weather was variable with periods of rain. The net radiative flux ranged between −25 and −19 W

m^{-2} for the period of 0200–0500 JST on 29 July. Unfortunately, there were no observations of the vertical profile of temperature during this period. However, it is considered that a surface-based inversion formed, since the concentrations of ^{212}Pb and ^{222}Rn daughters increased substantially. During the nights of 27–28 and 29–31 July, it was rainy or cloudy, and the net radiative flux was nearly zero. From the time variation of the vertical profiles of temperature obtained with the tethered instrumented balloon, it was observed that surface-based inversion layers were not present during the nights of 29–31 July. The wind was weak and the atmospheric conditions were near neutral (weakly stable) during these four nights. This near-neutral condition in the nighttime is considered to be the cause of the inverse trend of the $^{212}Pb/^{222}Rn$ daughters ratio at night.

4. Since the atmospheric pressure increased by 8 hPa during the period of 27–30 July and maintained the same level until the evening of 2 August, the exhalation rate of ^{222}Rn may have been independent of atmospheric pressure for the period, as mentioned above. Therefore, it is very unlikely that the inverse trend of the $^{212}Pb/^{222}Rn$ daughters ratio at night is due to an increase of atmospheric pressure.

From the above discussions, the inverse trend of the $^{212}Pb/^{222}Rn$ daughters ratio at night is considered to be due to the near-neutral condition.

4.2.3. *Period of Snow Cover*

The pattern of variation of the concentrations of ^{222}Rn daughters and ^{212}Pb and the $^{212}Pb/^{222}Rn$ daughters ratio for the first half of the IOP-2 is similar to that during the standard normal period. The snow depth was 0.2–0.3 m for the IOP-2. The ground in the forest area having very little snow cover, the atmospheric condition near the surface may have been almost the same as that with no snow cover.

In the latter half of the period, the concentrations of ^{222}Rn daughters and ^{212}Pb and the $^{212}Pb/^{222}Rn$ daughters ratio was continuously low through both day and night from 0300 JST on 1 March. The $^{212}Pb/^{222}Rn$ daughters ratio during this period was much lower than those during the period of strong winds (from 24 to 27 July) for the IOP-1. Snow cover existed on the paddy fields, while there was very little in the forest area. Considering the half-life of ^{220}Rn (55.4 s), it is thought that the snow decreased the effect of the extraction of ^{220}Rn. Hence, it is believed that the decrease of ^{212}Pb concentration was due to convective mixing provoked by the strong winds and due to the decrease of the extraction of ^{220}Rn from the ground by snow. With a snow depth of 0.2–0.3 m, a large number of ^{222}Rn atoms that reach the snow-ground interface pass through the snow and exhale from the air-snow interface into the atmosphere. It is considered that the decrease of concentration of ^{222}Rn daughters was due only to the convective mixing provoked by the strong winds. Considering these factors, the difference between the $^{212}Pb/^{222}Rn$ daughters ratios in the latter half of the IOP-2 and during the period of strong winds of the IOP-1 may be due to the fact that a snow cover of 0.2–0.3 m decreased the extraction of ^{220}Rn by the strong winds but did not decrease that of ^{222}Rn.

5. Concentrations of ^{222}Rn and its Daughters and their Ratio

Figure 9 shows the relations between the concentration of ^{222}Rn above the paddy field (location B2) and that of ^{222}Rn daughters above the uncultivated land (location B1) during the three observation periods. Figure 10 illustrates the time variations of ^{222}Rn daughters/^{222}Rn ratio for the three periods; the concentration of ^{222}Rn daughters (EC_{Rn}) could not be measured for the IOP-3 owing to instrument problems. Therefore, we compare the concentrations of ^{222}Rn and ^{222}Rn daughters (EC_{Rn}) and their ratios for the IOP-1 and IOP-2 only. Figure 11 shows the relations between the concentrations of ^{222}Rn and ^{222}Rn daughters (EC_{Rn}) for the IOP-1 and IOP-2, and Figure 12 shows time variations of the ^{222}Rn daughters (EC_{Rn})/^{222}Rn ratio for the two periods.

We also treat here the IOP-1 as a standard normal period as in Section 4. The snow depth was 0.2–0.3 m at location B for the IOP-2; the paddy fields were submerged for the IOP-3.

5.1. Period of Submergence of Paddy Fields

Figure 13 shows time variations of the sensible and latent heat fluxes on one of the paddy fields (location B2) during the IOP-1 and IOP-3. The sensible and latent heat fluxes were both about 100 W m^{-2} for the IOP-1, but the latent heat flux was about 200 W m^{-2}, and the sensible heat flux only about 50 W m^{-2} for the IOP-3. The former period represents a case of no water over the paddy fields, while the latter represents water cover on the paddy fields and a large amount of transpiration from the rice plants. This is representative of all the paddy fields at Akawase. The paddy fields were covered with water for the IOP-3, and therefore the exhalation rate of ^{222}Rn is considered to have been much less than that for the IOP-1. The ^{222}Rn concentration measured at the ridge in the centre of the paddy fields became not so low because of the advection of ^{222}Rn from the forest area due to the moderate to strong winds. From this, it is reasonable to assume that the concentrations of ^{222}Rn and its daughters in such a condition were almost the same for these two periods, as shown by the low concentrations in Figures 9a and 9c.

^{222}Rn was not supplied from the submerged paddy fields during the IOP-3, but was supplied only from the ridges between the paddy fields. There existed a surface-based inversion or near neutral (weakly stable) condition in the night-time for the period. The concentrations of ^{222}Rn at the submerged paddy fields were lower than those when the paddy fields had no surface water. At location B1, the concentrations of ^{222}Rn daughters at night for the IOP-1 were a little lower than those for the IOP-3. It may be due to these conditions that the ^{222}Rn daughters/^{222}Rn ratios were more than 0.4 at night for the IOP-3 (see Figure 10c), contrary to the observation that they were less than 0.4 for the IOP-1 (see Figure 10a).

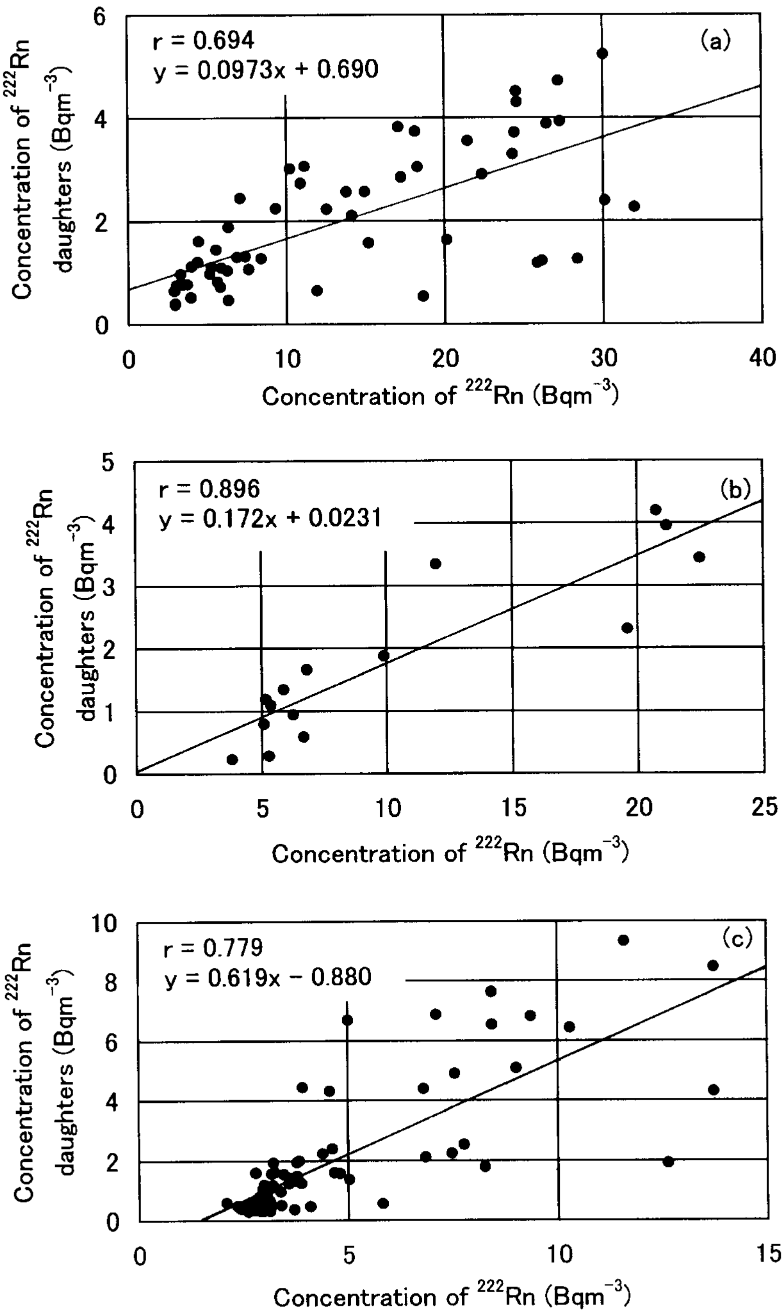

Figure 9. Relations between the concentration of ^{222}Rn at location B2 and the concentration of ^{222}Rn daughters at location B1. Correlation coefficient (r), regression equation and regression line are shown in each figure. (a) 6–13 October 1995; (b) 27 February–1 March 1998; (c) 23 July–2 August 1998.

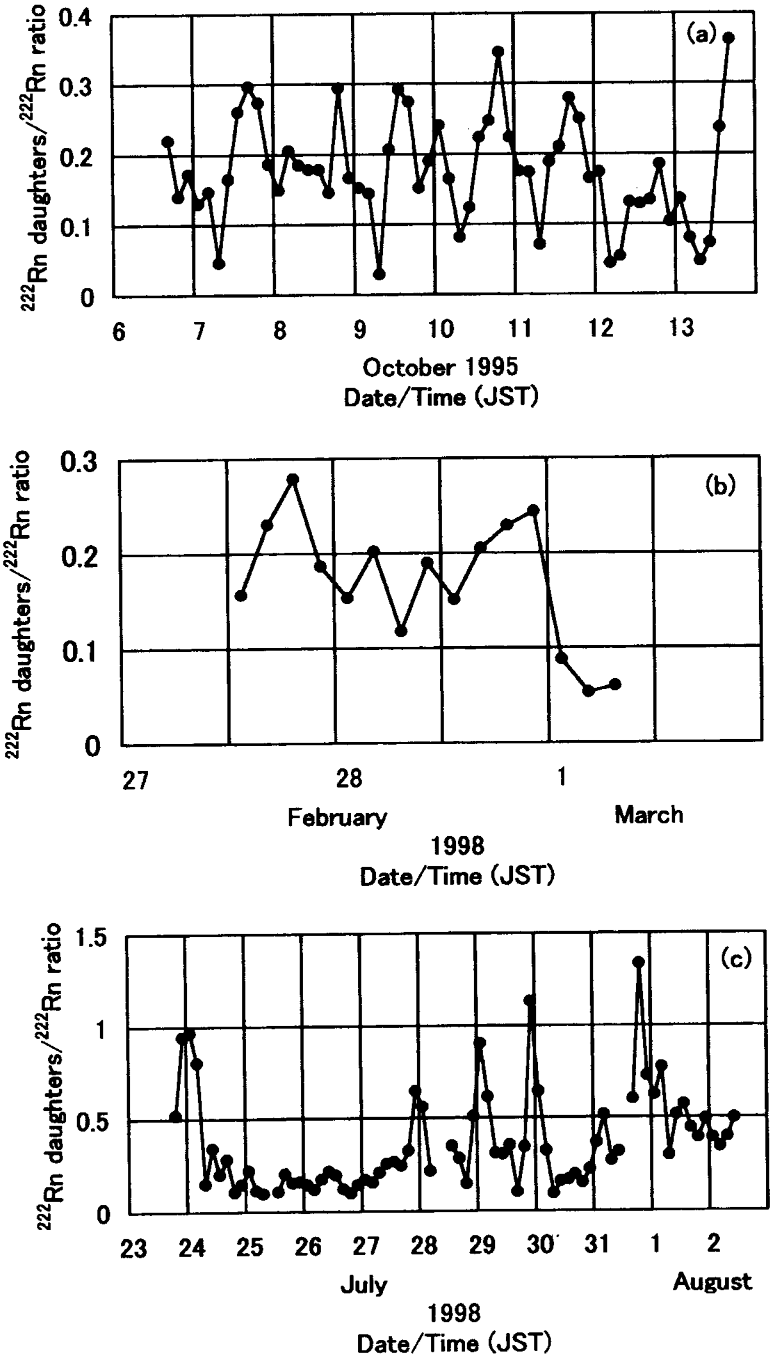

Figure 10. Time variations of the ratio of the concentration of ^{222}Rn daughters at location B1 to the concentration of ^{222}Rn at location B2 (^{222}Rn daughters/^{222}Rn ratio). (a) 6–13 October 1995; (b) 27 February–1 March 1998; (c) 23 July–2 August 1998.

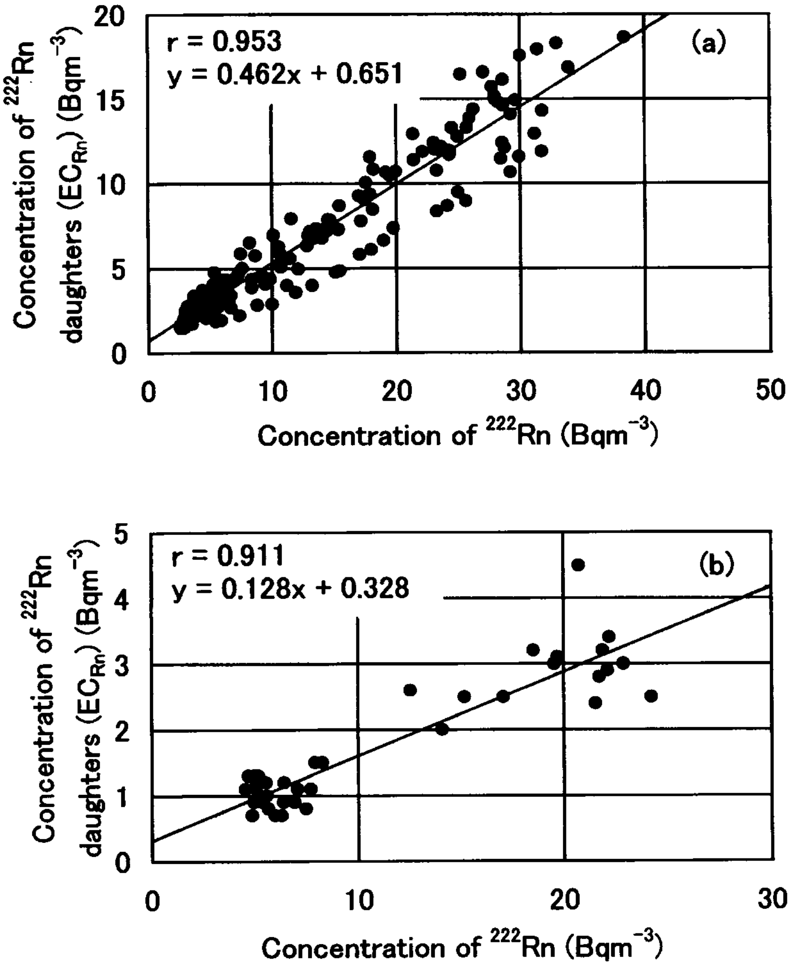

Figure 11. Relations between the concentration of ^{222}Rn and the concentration of ^{222}Rn daughters (EC_{Rn}). Correlation coefficient (r), regression equation and regression line are shown in each figure. (a) 6–13 October 1995; (b) 27 February–1 March 1998.

5.2. Period of Snow Cover

We find from Figures 9a and 9b that the range of concentrations of ^{222}Rn and ^{222}Rn daughters (EC_{Rn}) during the IOP-2 is almost the same as that during the IOP-1. No snow was observed over the paddy fields and forest area for the IOP-1, but there was a snow depth of 0.2–0.3 m during the IOP-2. With a snow depth of 0.2–0.3 m, a large number of ^{222}Rn atoms that reach the snow-ground interface pass through the snow and exhale from the air-snow interface into the atmosphere. Therefore, the exhalation rate of ^{222}Rn is considered to have been almost the same for these two periods. The small amount of snow over the forest area during the IOP-2 made the surface meteorological condition similar to that with no snow cover. This may be

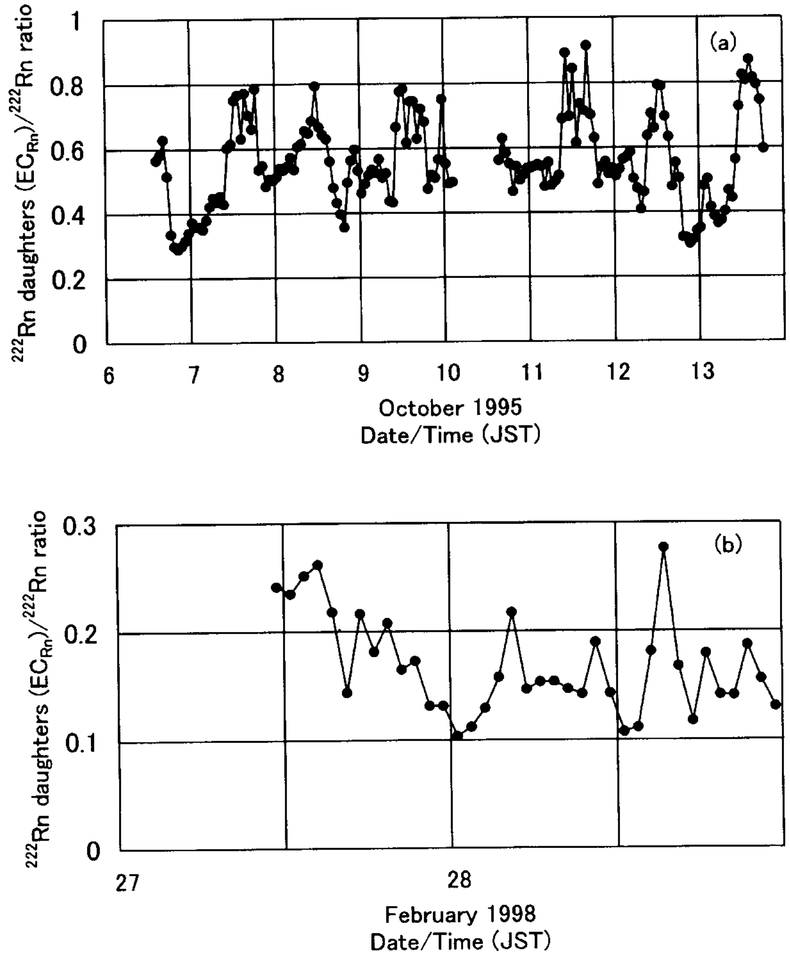

Figure 12. Time variations of the ratio of the concentration of ^{222}Rn daughters (EC$_{Rn}$) to the concentration of ^{222}Rn (^{222}Rn daughters (EC$_{Rn}$)/^{222}Rn ratio). (a) 6–13 October 1995; (b) 27 February–1 March 1998.

the cause of the fact that the range of concentration of ^{222}Rn daughters for the IOP-2 was almost the same as that for the IOP-1. Consequently, ^{222}Rn daughters/^{222}Rn ratios during the IOP-2 were almost the same as those during the IOP-1, as shown in Figures 10a and 10b.

From Figure 11, we see that the concentrations of ^{222}Rn at location B2 during the IOP-1 and during the IOP-2 are within the same range. We also find from the figure that when the concentration of ^{222}Rn daughters (EC$_{Rn}$) at location B2 is low, the IOP-1 and IOP-2 are similar, but when the concentration is high, the concentration during the IOP-2 is about one fourth of that during the IOP-1. The range of the ^{222}Rn daughters (EC$_{Rn}$)/^{222}Rn ratio for the IOP-1 was 0.3–0.9 at location B2 (see Figure 12a). However, the range of the ^{222}Rn daughters (EC$_{Rn}$)/^{222}Rn ratio for

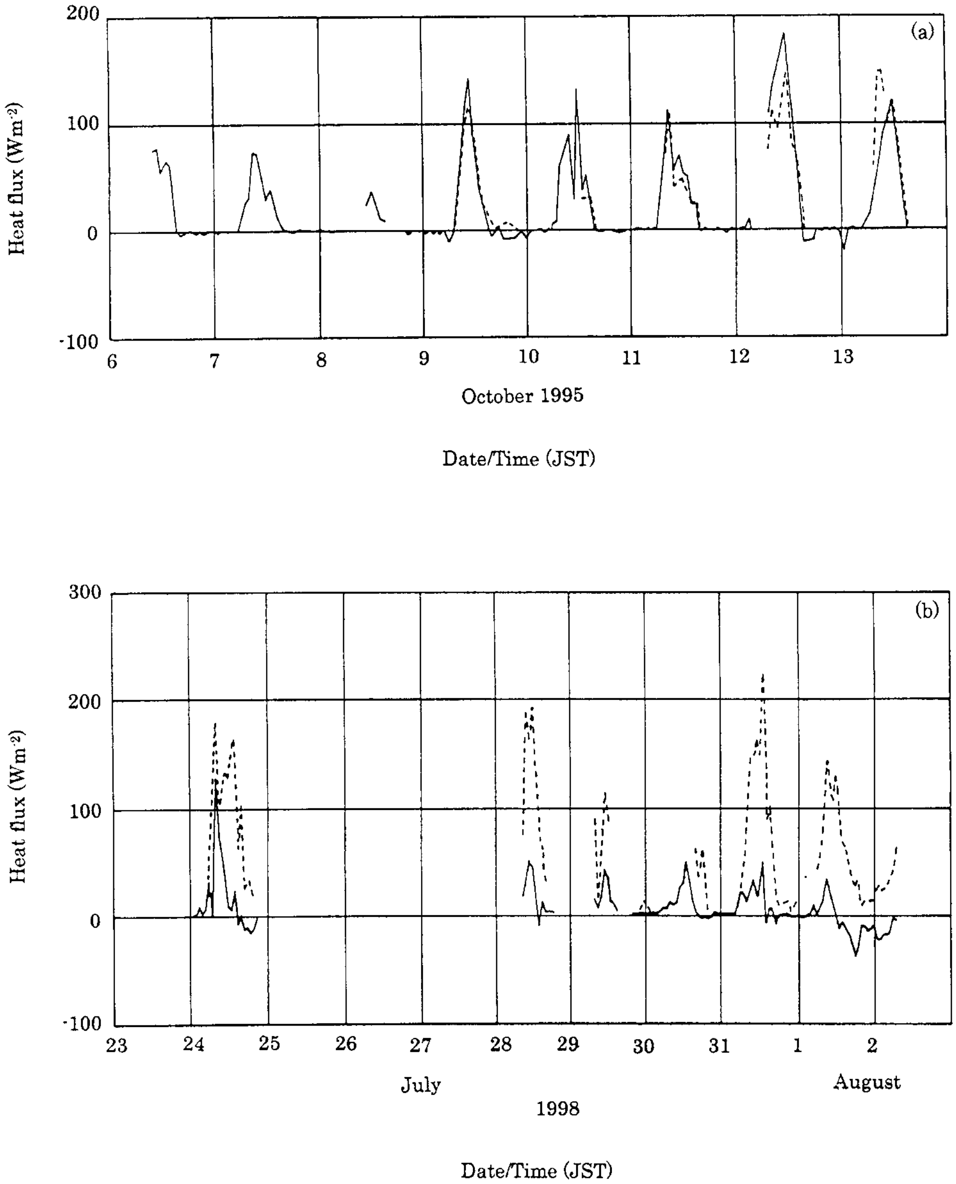

Figure 13. Comparison of sensible heat flux (continuous curve) and latent heat flux (dashed curve) between (a) 6–13 October 1995 and (b) 23 July–2 August 1998.

the IOP-2 was 0.1–0.3 at location B2 (see Figure 12b) and outside the range for the IOP-1. The exhalation rate of ^{222}Rn at location B1 is about one third of that at the other area (see Table II). We surmise that the ^{222}Rn daughters/^{222}Rn ratio at location B1, where the meteorological condition near the ground was similar to that with no snow, varies from 0.2 to 0.9 from Figure 10b. Thus, the equilibrium factor in the case of snow cover is different from that in the case of no snow cover. This may be due to small differences in meteorological conditions caused by the difference between the snow of the paddy and forest areas.

6. Conclusions

We presumed in prior statistical analysis (Kataoka et al., 1999) that the ^{212}Pb/^{222}Rn daughters ratio is a good supplemental tracer for a study of detailed atmospheric conditions that cannot be elucidated using concentrations of ^{222}Rn, ^{212}Pb and/or short-lived ^{222}Rn daughters alone. The observations described in this paper clearly support the assumption.

There are very few studies in which the influence of meteorological conditions on a lack of radioactive equilibrium between ^{222}Rn and its short-lived daughters is discussed in detail. From the observations described in this paper, it is suggested that the ratio of the concentration of ^{222}Rn daughters to the concentration of ^{222}Rn measured at the same location is useful as a supplemental tracer to their concentrations for a study of detailed atmospheric conditions. It is also suggested that the ratio of the concentration of ^{222}Rn daughters at a given location to the concentration of ^{222}Rn in a nearby area is useful as a supplemental tracer to their concentrations in order to study the difference in atmospheric conditions and topography between the two locations.

Recently, new methods have been developed for estimating variations of concentrations of airborne ^{222}Rn and ^{212}Pb (one of the ^{220}Rn decay products), but the results of computation do not always agree with observations (Segawa et al., 1993; Chino and Yamazawa, 1996; Sakashita et al., 1997). Such methods involve diffusion and transport models of ^{220}Rn, ^{222}Rn and their daughters using constant exhalation rates of ^{220}Rn and ^{222}Rn from the ground. The data and discussion presented in this paper may be fundamental to improving computational models, such that they accommodate a wider range of weather related variations in ^{220}Rn and ^{222}Rn exhalation rates.

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