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A study of the atmospheric boundary layer using ^{222}Rn, its short-lived daughters and ^{212}Pb

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INTRODUCTION

It is well known that activity concentrations of ^{222}Rn, ^{220}Rn and their daughters are suitable tracers for the study of diffusion processes in the atmospheric boundary layer (e.g.,1-4) and of long range transport in the troposphere (e.g.,5,6). Recently, it has become clear that the ratio of the activity concentration of ^{212}Pb and that of either ^{222}Rn or its short-lived daughters is also an indicator of the state of the atmospheric boundary layer (7-9).

In this paper, we select two periods which contain interesting meteorological conditions, namely one period contains weak mixing near the ground surface at night and the other does mechanical mixing which continued all day long. We discuss relations between activity concentrations of ^{212}Pb, ^{222}Rn and its daughters, and the ratio of the activity concentration of ^{212}Pb and the activity concentration of short-lived ^{222}Rn daughters under different meteorological conditions.

We use the following terminology. Atmospheric activity concentrations of ^{212}Pb, ^{222}Rn, and of short-lived ^{222}Rn daughters (which are 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 activity concentrations of ^{212}Pb and short-lived ^{222}Rn daughters will hereafter be designated as the ^{212}Pb/^{222}Rn daughters ratio. The equilibrium-equivalent ^{222}Rn concentration of a non-equilibrium mixture of short-lived ^{222}Rn daughters 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 will hereafter be designated as the equilibrium-equivalent concentration of ^{222}Rn.

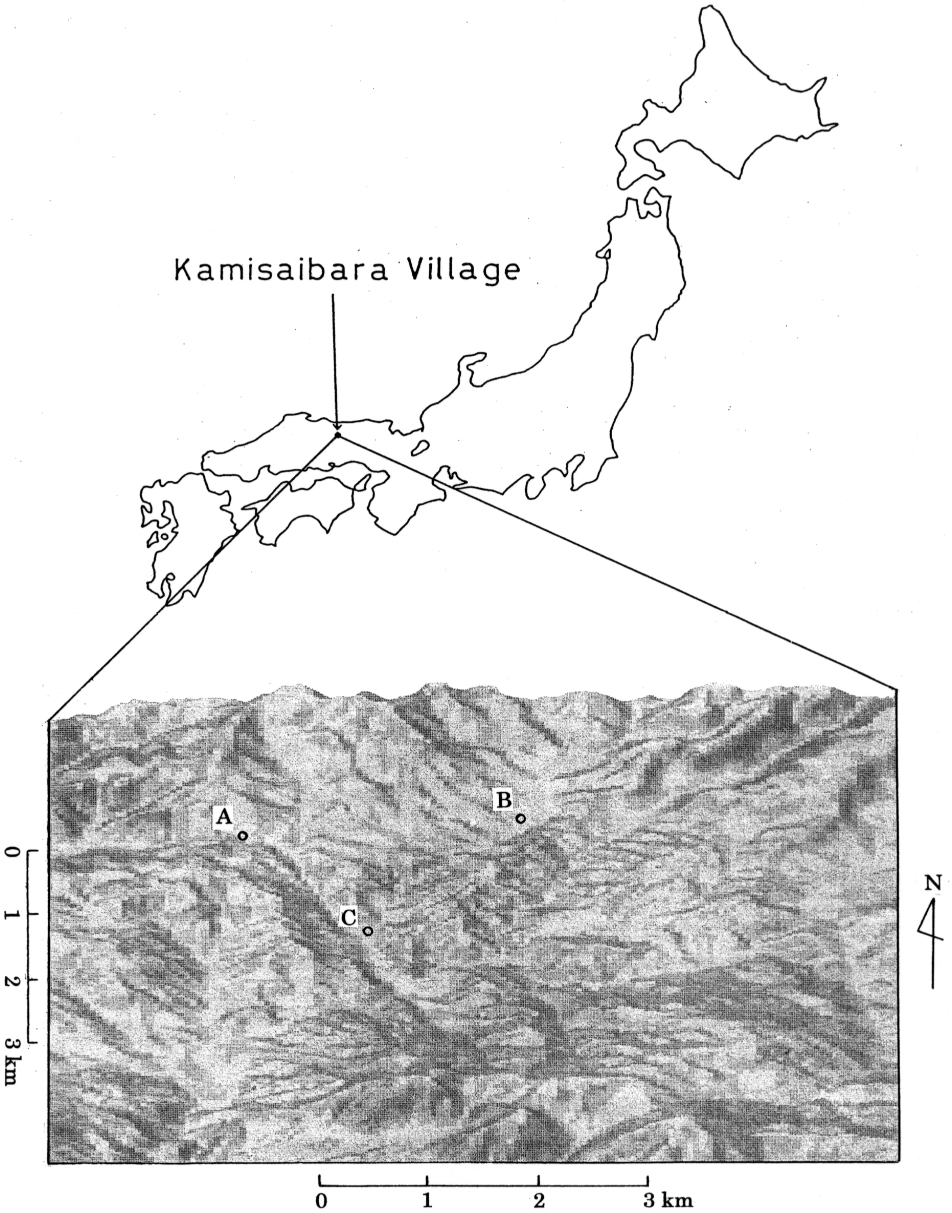

Figure 1. Location of observation sites.

SITE AND EXPERIMENT

Since 1 April 1993, measurements of concentrations of ^{212}Pb and ^{222}Rn daughters have been carried out simultaneously every three hours 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 1 and are, in order, Tohge, Akawase and Tennoh. The location of the measurement at Akawase is designated as B1 to distinguish this primary site from a nearby temporary site mentioned below. Tohge is at the top of a ridge in the Chuhgoku Mountains and is at an elevation of 740 m above sea level. Akawase 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. Tennoh is situated within a plain (1100 m by 400 m) at the junction of Ikegoh Valley and Akawase Valley, and has an elevation of 540 m. Some uranium deposits exist in this region. However, they contain low-grade ores and do not influence the concentration of the ^{222}Rn daughters measured at these three sites (9).

The measurements of ^{212}Pb and short-lived ^{222}Rn daughters have been carried out with devices having two α-scintillation counters. Details of such instruments are given in a previous paper (10). The reported concentration of ^{212}Pb is the average of three-hour collection but, since the effective half-life of short-lived ^{222}Rn daughters is about 40 minutes, the latter half of the three-hour collection is more important for 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 sites. Additional observations such as snow depth and net radiative flux have also been made continuously but only at the Akawase site.

At location B2 situated about 140 m southeast of the location B1 at Akawase, concentration of ^{222}Rn and equilibrium-equivalent concentration of ^{222}Rn were measured for two short periods. The first period was from 6 to 13 October 1995 and the second was from 27 February to 1 March 1998. The concentration of ^{222}Rn was measured with a ^{222}Rn monitor (Pylon Electronics Inc., Model PMT-TEL with Model AB-5), and the equilibrium-equivalent concentration of ^{222}Rn was measured with WL-monitor (Tracerlab Instruments, Model WLM-PLUS). The exhalation rate of ^{222}Rn from the soil surface was measured at three points at Akawase during the first of these short periods, one at location B1 and two at location B2.

RESULTS AND DISCUSSION

In a previous paper (9), we discussed, from a statistical point of view, whether the ^{212}Pb/^{222}Rn daughters ratio is useful as a supplemental tracer for determining the meteorological conditions of the atmospheric boundary layer. We concluded that it is indeed a useful supplemental tracer and, in this paper, provide further evidence based on individual cases.

Since we must deal with observations of ^{212}Pb and short-lived ^{222}Rn daughters obtained every three hours, we first compare measurements of ^{222}Rn and its short-lived daughters obtained every hour and every three hours at Akawase to determine to what degree the concentration of ^{222}Rn daughters follows changing meteorological conditions. Figure 2 presents time variations of the concentration of ^{222}Rn, the equilibrium-equivalent concentration of ^{222}Rn and the concentration of ^{222}Rn daughters at Akawase. The concentration of ^{222}Rn, the equilibrium-equivalent concentration of ^{222}Rn and the concentration of ^{222}Rn daughters were high as usual during the nights of 6-13 October 1995. Similarly, they were high due to weak winds during the night of 27-28 February. However, they remained low the following night due to a moderate wind between 19:00 JST on 28 February and 2:00 JST on 1 March, and due to a very strong wind between 3:00 JST and 9:00 JST on 1 March. Although the pattern of the concentration of ^{222}Rn daughters is smoother than those of the concentration of ^{222}Rn and the equilibrium-equivalent concentration of ^{222}Rn, they bear a considerable resemblance. This indicates that the concentration of ^{222}Rn daughters measured every three hours follows changes in meteorological conditions.

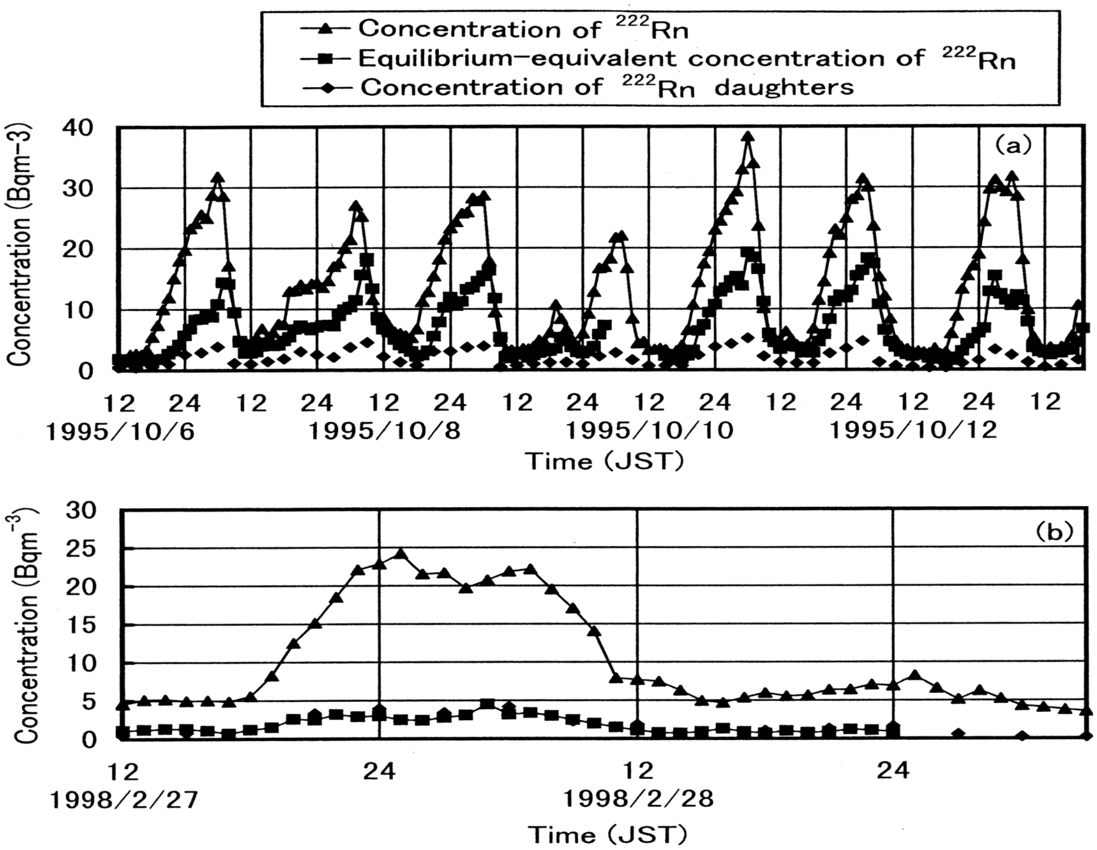

Figure 2. Time variations of the concentration of ^{222}Rn, the equilibrium-equivalent concentration of ^{222}Rn, and the concentration of ^{222}Rn daughters at Akawase for the periods (a) from 6 to 13 October 1995 and (b) from 27 February to 1 March 1998.

It is found from Figure 2(a) that an equilibrium factor ranged from 0.3 to 0.9 for ^{222}Rn and its short-lived daughters at the location B2. Since the concentration of ^{222}Rn daughters is 0.8 to 1 times that of the equilibrium-equivalent concentration of ^{222}Rn (11) for an equilibrium factor ranging from 0.1 to 1 (12), the equilibrium-equivalent concentration of ^{222}Rn is 80-100 % of the concentration of ^{222}Rn daughters. As shown in Table 1, the exhalation rate of ^{222}Rn from the soil at the location B2, where the concentration of ^{222}Rn and the equilibrium-equivalent concentration of ^{222}Rn were measured every hour, is about 3 times larger than that at the location B1, where the concentration of ^{222}Rn daughters was measured. Considering this difference and assuming an equilibrium factor of about 0.3 - 0.9 for ^{222}Rn and its short-lived daughters at the location B1, the difference between the equilibrium-equivalent concentration of ^{222}Rn and the concentration of ^{222}Rn daughters during the period 6 - 13 October 1995 is reasonable.

However, from Figure 2(b), the equilibrium factor was about 0.1 to 0.3 at location B2 between 27 February and 1 March 1998. The equilibrium factor at location B1 is conjectured to be in the range 0.3 to 0.9 for the same period, because the concentrations of ^{222}Rn and ^{222}Rn daughters were almost the same during the two short observation periods. Location B2 is situated in the centre of a rice paddy, while location B1 is at the edge of the paddy about 140 m northwest from its centre and borders on a forest area. Also, there was a snow to a depth of 0.2-0.3 m. Therefore, it is considered that any discrepancy between the equilibrium-equivalent concentration of ^{222}Rn and the concentration of ^{222}Rn daughters may be due to small differences in meteorological conditions caused by the difference between the snow of the paddy and forest areas. 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. The question thus remains whether the ratio of the concentrations of ^{222}Rn and its short-lived daughters is an appropriate supplemental tracer to the concentration of ^{222}Rn or its short-lived daughters.

Table 1. Exhalation rate of ^{222}Rn masured during the period from 6 to 13 October 1995.

Site	Exhalation rate of ^{222}Rn ($Bqm^{-2}s^{-1}$)
Uncultivated land (Location B1)	0.0059
Paddy field (Location B2)	0 .0100
Ridge between paddy fields (Location B2)	0.0198

The patterns of these three concentrations were similar, as mentioned above. Further, ^{212}Pb exhibits similar behaviour to ^{222}Rn and its short-lived daughters (Jacobi and André, 1963). It is therefore reasonable to believe that the concentrations of ^{212}Pb and ^{222}Rn daughters both follow changes in meteorological conditions.

Secondly, we discuss the relation between time variations of concentrations of ^{212}Pb and ^{222}Rn daughters, and their ratio, and the corresponding meteorological conditions at each of the three sites. We select two periods in which observations were carried out at location B2 at Akawase.

For the three locations, Tohge, Akawase and Tennoh, Figure 3 presents 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, for the period from 10 October to 13 October 1995. Figure 4 shows time variations of wind speed and net radiative flux for the same period. From a statistical point of view, the concentrations of ^{212}Pb and ^{222}Rn daughters and the ^{212}Pb/^{222}Rn daughters ratio at the three sites are well correlated. However, close examination of the time variations of the concentrations of ^{212}Pb and ^{222}Rn daughters reveals differences between the three sites. Since differences are most apparent during the night of 11-12 October, this period is inspected in more detail. At Tohge, the concentrations of ^{212}Pb and the ^{222}Rn daughters increased until 21:00 JST but then decreased. In contrast, concentrations continued to increase until after midnight at Akawase and Tennoh. This difference may have been caused by the fact that a moderate wind continued through much of the night at Tohge.

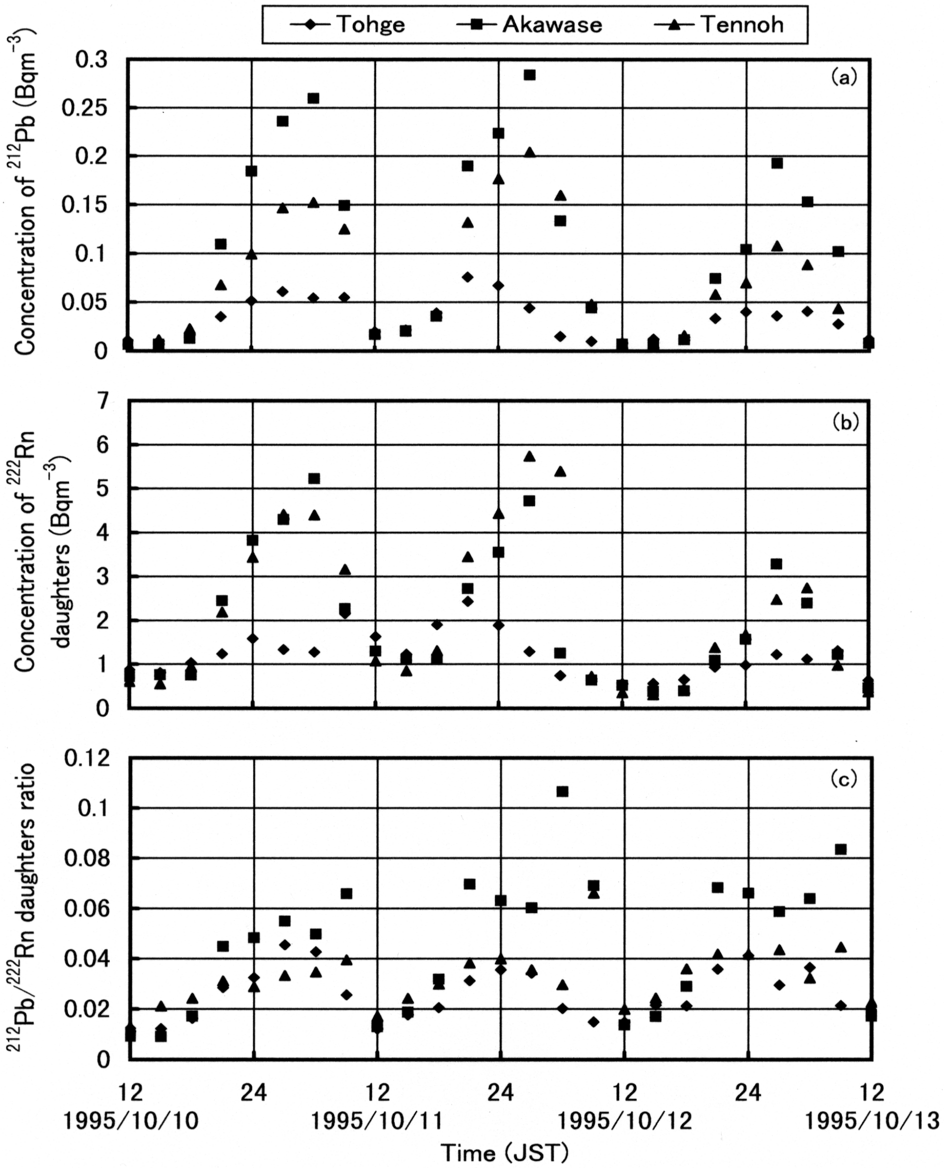

Figure 3. Time variations of (a) the concentration of ^{212}Pb, (b) the concentration of ^{222}Rn daughters, and (c) the $^{212}Pb/^{222}Rn$ daughters ratio at three locations for the period from 10 to 12 October 1995.

Cloud cover at Akawase between 3:00 and 5:30 JST on 12 October resulted in a net radiative flux that was less negative than usual (Figure 4) and an atmospheric surface layer that was less stable than normal. Combined with slightly higher than normal wind speed between 4:00 and 6:00 JST, this is likely to have resulted in weak mixing, and the observed decreases in concentrations of ^{212}Pb and the ^{222}Rn daughters. This weak mixing also produced an extremely large value of the $^{212}Pb/^{222}Rn$ daughters ratio at 6:00 JST. At the same time, at Tennoh, the concentration of ^{212}Pb decreased by 25% and 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 that at Akawase. This also resulted in a normal value to the $^{212}Pb/^{222}Rn$ daughters ratio at Tennoh.

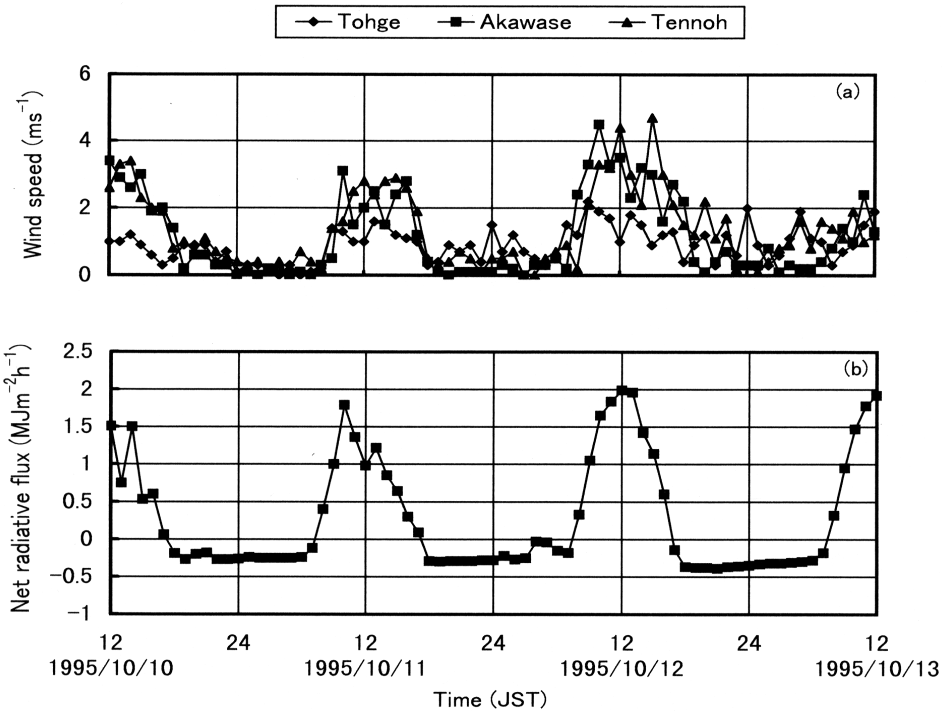

Figure 4. Time variations of (a) wind speed at three locations and (b) net radiative flux at Akawase for the period from 10 to 12 October 1995.

Figure 5 gives 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 for the period from 27 February to 3 March 1998. Figure 6 shows time variations of (a) wind speed and (b) depth of snow for the same period. The wind speed at Tohge was stronger than usual from around 2:00 JST to around 24:00 JST on 1 March. That at Akawase was stronger than usual from around 3:00 JST on 1 March to around 10:00 JST on 2 March. Winds at Tennoh were stronger than usual from around 2:00 JST to around 22:00 JST on 1 March. The depth of snow at Akawase increased from 0.21 m at 23:00 JST on 28 February to 0.32 m at 8:00 JST on 1 March, remained constant until 14:00 JST on 2 March, after which it decreased to 0.28 m. The depth of snow at Tohge was similar to that at Akawase. Tennoh initially had no snow on the ground prior to the snowfall late on 28 February, after which the depth of snow was about 0.1 m.

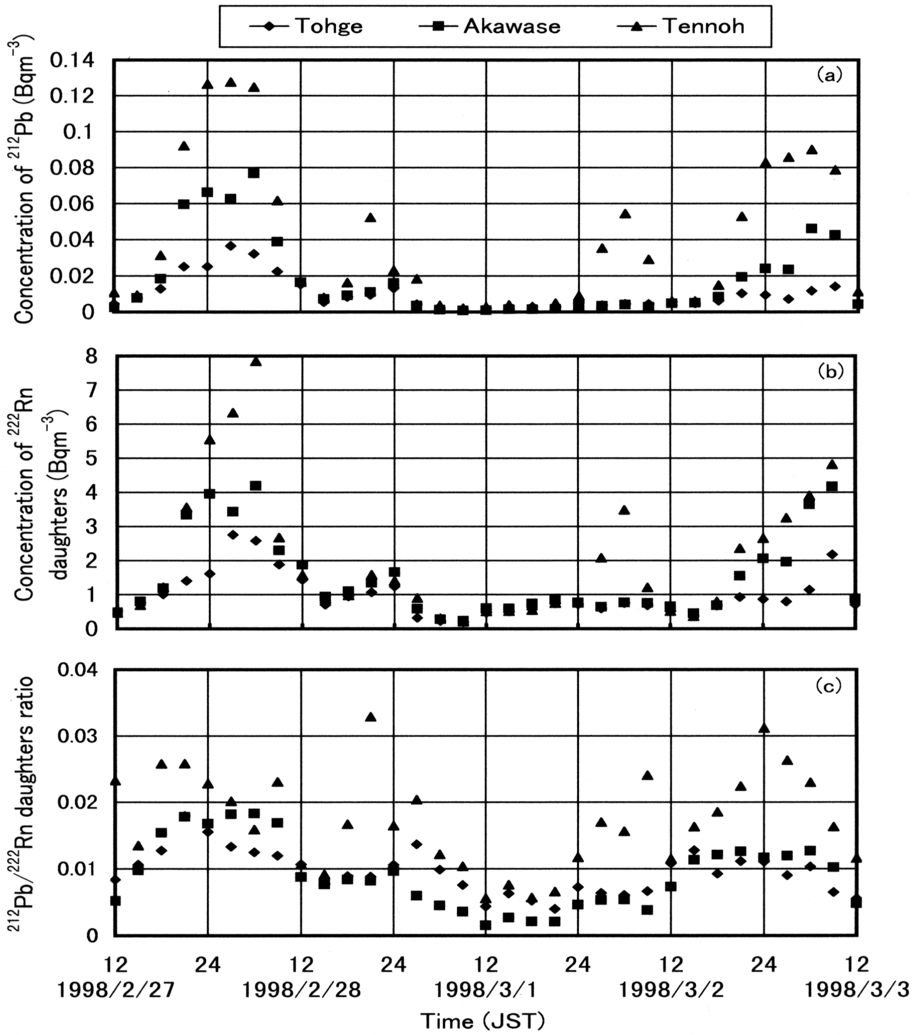

Figure 5. Time variations of (a) the concentration of ^{212}Pb, (b) the concentration of ^{222}Rn daughters, and (c) the ^{212}Pb/^{222}Rn daughters ratio at three locations for the period from 27 February to 3 March 1998.

Concentrations of ^{212}Pb and of ^{222}Rn daughters at all three sites decreased at 3:00 JST on 1 March and maintained low levels through the remainder of the day. All of the ^{212}Pb/^{222}Rn daughters ratios at the three sites remained lower than about 0.01 from 6:00 to 24:00 JST on 1 March. At Tennoh, the wind became weak at about 23:00 JST on 1 March, with the consequence that concentrations of ^{212}Pb and ^{222}Rn daughters and ^{212}Pb/^{222}Rn daughters ratio increased. At Akawase, the low concentrations of ^{212}Pb and ^{222}Rn daughters and the low ^{212}Pb/^{222}Rn daughters ratio resulted from the mechanical mixing caused by the continued strong wind that night. Although the wind speed was also not strong (0.5 – 0.7 ms^{-1}) at Tohge, being located at the top of a ridge in the Chuhgoku mountains, mechanical mixing may have been sufficient to maintain low concentrations. Furthermore, the concentrations of ^{212}Pb and ^{222}Rn daughters and the ^{212}Pb/^{222}Rn daughters ratio increased during the night of 2-3 March in spite of there being almost the same depth of snow as during the night of 28 February to 1 March. It is concluded that the decrease was provoked not by the increase of the snow cover, but by the strong wind.

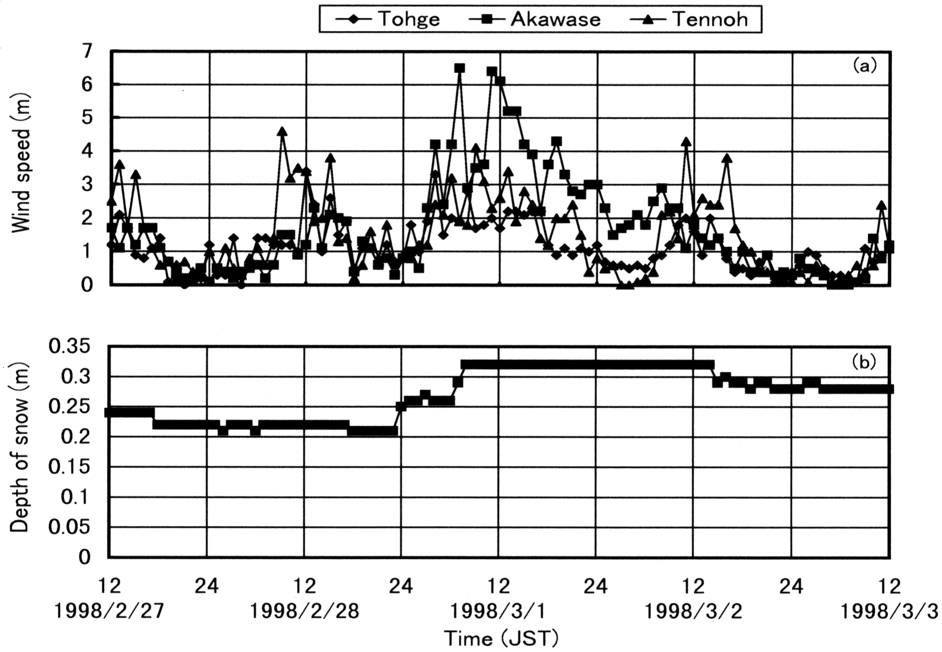

Figure 6. Time variations of (a) wind speed at three locations and (b) depth of snow at Akawase for the period from 27 February to 3 March 1998.

Thus, in the mountainous region, variations in the concentrations of ^{212}Pb and ^{222}Rn daughters and the ^{212}Pb/^{222}Rn daughters ratio are different from each other at the three sites, even within an area as small as 1.5 km radius. It is also observed from Figures 3 and 5 that the ^{212}Pb/^{222}Rn daughters ratio ranged from 0.003 to 0.11 even during these two short periods. This indicates that, since the half-life of ^{220}Rn is much shorter than those of ^{222}Rn, ^{212}Pb and ^{222}Rn daughters, they behave somewhat differently in a mountainous region. Hence, ^{212}Pb/^{222}Rn daughters ratio is a good supplemental tracer to the concentrations of ^{222}Rn, ^{222}Rn daughters and/or ^{212}Pb for exposing the meteorological conditions of complex terrain.

CONCLUSIONS

The variations of concentration of ^{222}Rn and the equilibrium-equivalent concentration of ^{222}Rn observed every hour generally resembles those of the concentrations of the short-lived ^{222}Rn daughters observed at three-hour intervals. Hence, it is demonstrated that the concentrations of short-lived ^{222}Rn daughters measured every three hours followed changes in atmospheric condition.

Furthermore, by adding the ^{212}Pb/^{222}Rn daughters ratio as a supplemental tracer, we reveal conditions of the atmosphere that can not be elucidated using concentrations of ^{222}Rn, ^{212}Pb and/or short-lived^{222}Rn daughters alone, as we conjectured from prior statistical analyses.

It is a subject for future inquiry whether or not the ratio of concentrations of ^{222}Rn and its short-lived daughters is also useful as a supplemental tracer to their concentrations.

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