AN AID TO THE STUDY OF THE BEHAVIOR OF RADON AND AIR POLLUTANTS IN THE ATMOSPHERIC BOUNDARY LAYER

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Abstract. Intensive observations of atmospheric activity concentration of ^{222}Rn, concentrations of air pollutants (NO, NO_2 and SO_2), and of the atmospheric boundary layer were carried out at Kabutonishi Town, Kasaoka City, Okayama Prefecture in Japan. Concentrations of ^{222}Rn at night were lower than those at Akawase and Uchio reported in previous papers. Smaller increases in the concentration of ^{222}Rn at night and slow decreases of the concentration of ^{222}Rn during morning calm could be clearly explained by meteorological observation including Doppler sodar. In the daytime, ^{222}Rn levels were almost the same as those at Akawase and Uchio. Although atmospheric conditions were near weak stability at night, it was possible to obtain information regarding advection using NO, NO_2 and SO_2 originating from motor vehicles. In addition, information regarding advection was obtained from observations of NO, NO_2 and SO_2 originating from factories and ships in the daytime, including a period of morning calm. These results support and supplement our opinion expressed in an earlier publication that a set of ^{222}Rn and air pollutants can provide useful information regarding local conditions of the atmospheric boundary layer.

KEYWORDS: Concentration of ^{222}Rn, Concentrations of air pollutants, Land and sea breezes, Surface-based inversion layer, Weak mixing, Doppler sodar

1. Introduction

It is suggested that observations of air pollutants can usefully supplement studies of the relation between the atmospheric activity concentration of ^{222}Rn and the local nature of the atmospheric boundary layer [1]. In this paper, first, we discuss the relation between the atmospheric activity concentration of ^{222}Rn and conditions of the atmospheric boundary layer at another site, where land and sea breezes are prevalent, namely Kabutonishi Town ($34^{O}28$'N, $133^{O}29$'E), Kasaoka City, Okayama Prefecture in Japan. Nest, we examine the relation between concentrations of NO, NO_2 and SO_2 and conditions of the atmospheric boundary layer at the site and within its vicinity.

We use the following abbreviations. Atmospheric activity concentration of ^{222}Rn will hereafter be denoted by concentration of ^{222}Rn. The experimental sites in Kabutonishi Town, in Akawase and in Uchio will hereafter be designated as K-site, A-site and U-site, respectively.

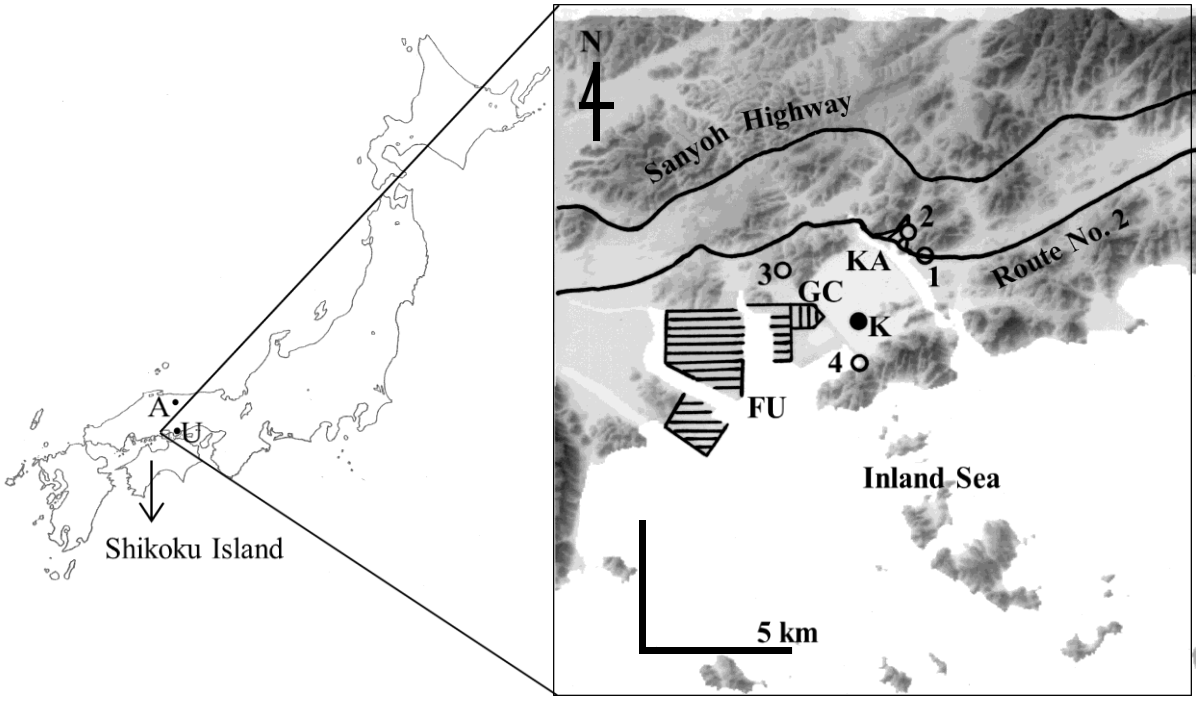

Figure 1. Locations of the experimental sites and observatories of Okayama Prefecture for monitoring air pollutants. ●K: Experimental site in Kabutonishi Town (K-site), ●A: Experimental site in Akawase (A-site), ●U: Experimental site in Uchio (U-site), ○1: Observatories for monitoring air pollutants emitted from motor vehicles driving along route No. 2, ○2–4: Observatories 2–4 for monitoring air pollutants, KA: Lightly populated region of Kasaoka City, FU: Fukuyama Industrial Complex, GC: Golf course

2. Description of Study Site and Instrumentation

We selected the K-site at Kasaoka Air-Station in Kabutonishi Town as a supplementary location and carried out intensive observation of the atmospheric boundary layer together with measurement of concentration of ^{222}Rn at the location from 31 August to 15 September 1998 (see Fig. 1).

Kabutonishi Town is on reclaimed land, which is 4 km long by 3 km wide and utilized for pastureland, stock farms, cornfields, flower gardens etc. To the west of this area, another region of reclaimed land about 5 km^2 in area is utilized for an ironworks (a small part of a large-scale iron-manufacturing company) and a golf course. Fukuyama Industrial Complex, which includes the large-scale iron-manufacturing company, is located to the west of these reclaimed lands over a waterway that is part of Fukuyama Port. There is also a 450 m wide waterway on the east side of the reclaimed land. The lightly populated region of Kasaoka City is located to the northeast of the reclaimed lands across the waterway. The K-site is located about 3 km north of the coast of the Inland Sea of Japan and about 1 km north-northeast of a cove (New Kasaoka Port) made by the reclaimed land. The part of the Inland Sea where the K-site is located is 30–40 km in width. National road route No. 2 having heavy traffic runs from east to west to the north of the K-site, and Sanyoh express highway having only light traffic also runs from east to west to the north of route No. 2. Main local road route No. 3 functioning as a by-pass of route No. 2 runs from north to south and takes a westward bend (see Fig. 2). There are four observatories to monitor air pollutants which are well suited to analyze the relation among concentrations of ^{222}Rn, air pollutants and atmospheric conditions, as were the observatories in the case of the U-site [1].

A sketch around the K-site is shown in Fig. 2. Radon observations were taken at the Air-Station, which consists mainly of a runway (800 m by 25 m), an apron (50 m by 40 m), a taxiway (37.5 m by 9 m) and roads that are all laid with asphalt. An intake from which air was drawn for determination of ^{222}Rn lay in the lee of the apron at night. The distance between the intake and the end of the apron was about 7 m. A ditch (2400 m by 30 m) is located about 20 m west-northwest from the runway and runs parallel with it. One end of the ditch takes a southeastward bend and is connected to a pond near New Kasaoka Port. The other end takes a northeastward bend and is connected to narrow ditches. A factory manufacturing sulfuric acid is located about 1.5 km to the southeast of observatory 4.

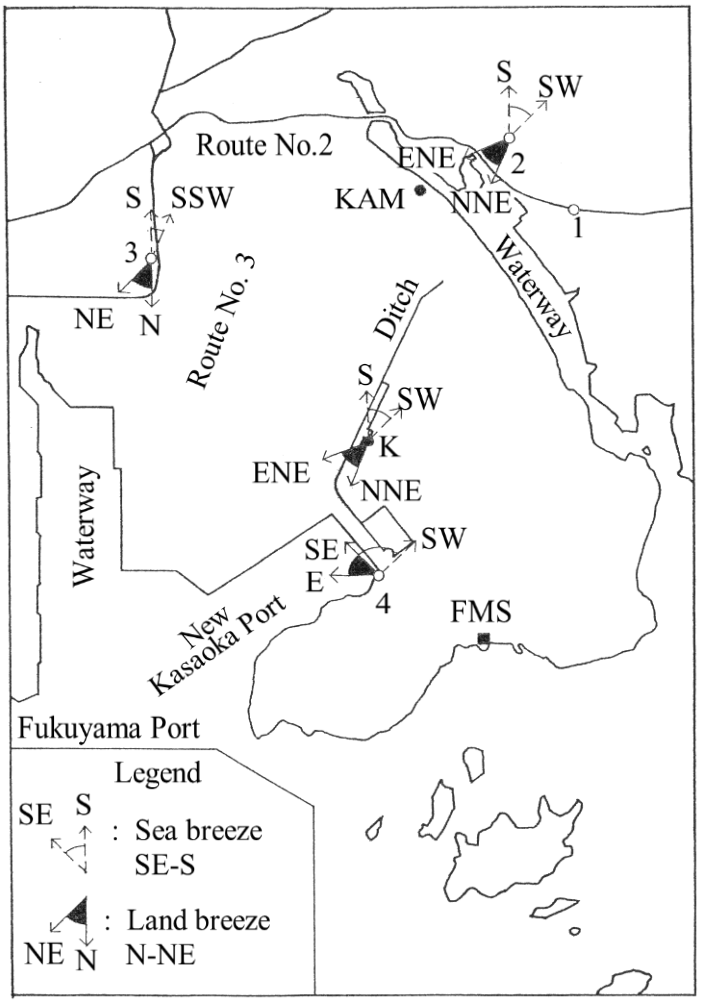

Figure 2. A sketch around the K-site showing observatories and predominant wind directions at the K-site during the period from 31 August to 15 September 1998. The wind directions shown are 10-minute averages. ●K: Experimental site in Kabutonishi Town (K-site), ○1: Observatories for monitoring air pollutants emitted from motor vehicles driving along route No. 2, ○2–4: Observatories 2–4 for monitoring air pollutants, ●KAM: Kasaoka station of Automated Meteorological Data Acquisition System, ■FMS: Factory manufacturing sulfuric acid. This sketch was made using JMC-Map of Japan Map Center.

Predominant wind directions at the K-site and at observatories 2, 3 and 4 in both the daytime and at night are also included in Fig. 2. Wind speed and direction was not measured at observatory 1. A land breeze between NNE and ENE and a sea breeze between S and SW were both common at the K-site. A sea breeze blew from the Inland Sea, but a land breeze blew to New Kasaoka Port at observatory 4. Land and sea breezes blew along the waterway at Kasaoka station of Automated Meteorological Data Acquisition System (KAM-station) [2].

The instruments used to obtain observations of ^{222}Rn and of meteorology are given in a previous paper [3], except that an instrumented tethered balloon, low-level sondes and infrared thermal imagery were not used. The instruments used to obtain data on air pollutants (NO, NO_2 and SO_2), wind speed and direction at the observatories of Okayama Prefecture for monitoring around the K-site are the same as (or another version of) those used at the observatories around the U-site and are described in a previous paper [1]. Observations of ^{222}Rn and air pollutants were obtained every hour.

3. Source of ^{222}Rn for calculation of concentration of ^{222}Rn

We start by making the following five points about our observations of exhalation rate of ^{222}Rn and of the meteorological and site conditions:

1. The exhalation rates of ^{222}Rn were measured at three points within the K-site during the intensive observation period. The average of the measured values is 0.0082 $Bqm^{-2}s^{-1}$. These are within the range of the exhalation rates of ^{222}Rn measured elsewhere in Japan [4].
2. The amount of rainfall was 6 mm for the period of 0700–0900 JST on 6 September at the K-site during the intensive observation period. Soil moisture increased by rainfall reduces the exhalation rate of ^{222}Rn but the rate recovers within 1.5–2 days after the rainfall [5]. Therefore, when calculating the concentrations of ^{222}Rn assuming that the exhalation rate of ^{222}Rn is constant, we eliminate the period from 0700 JST on 6 September to 0900 JST on 8 September.
3. Atmospheric pressure varied between 1005.5 hPa and 1015.0 hPa at the K-site during the intensive observation period. We treat the exhalation rate of ^{222}Rn as being independent of atmospheric pressure [6].
4. Other meteorological factors such as wind speed, temperature of the ground surface, and humidity have only a minor effect on the exhalation rate of ^{222}Rn [7].
5. The Inland Sea is shallow, and the exhalation rate of ^{222}Rn from the surface of the Inland Sea is at least one order of magnitude lower than that from the ground surface near the shore of the Inland Sea [8]. The part of the Inland Sea where the K-site is located is 30–40 km (average about 35 km) in width with Shikoku Island to the south. It is considered that the Inland Sea affects the concentration of ^{222}Rn at the K-site under such conditions, especially in the case of sea breeze. However, despite the fact that there is a substantial difference between exhalation rates, we assume here that the exhalation rate of ^{222}Rn for the Inland Sea is the same as that for land in order that we might apply a one-dimensional (1D) transport model.

Based upon the above five points, we treat the exhalation rate of ^{222}Rn as a constant, the average value, for the periods when there is no effect of rainfall at the K-site during the intensive observation period.

4. Concentration of ^{222}Rn, and land and sea breezes

Time variation of the measured concentration of ^{222}Rn is shown in Fig. 3a. The figure also shows calculations of diurnal variations of concentration of ^{222}Rn ranging from neutral stability in the daytime (NNN) to strong stability at night (IWN), and from strong instability in the daytime (SSN) to weak stability at night (WNN) made using the one-dimensional (1D) transport model proposed by Beck and Gogolak [9] under the conditions of Section 3. Such calculations use the vertical profiles of diffusion coefficient proposed by Jacobi and André [10] (NNN, IWN, SSN and WNN are notation of Jacobi and André [10]), and are calibrated by the average value of the exhalation rate of ^{222}Rn (0.0082 $Bqm^{-2}s^{-1}$). A period subject to the effects of rainfall is omitted from the calculated results.

Considering the complexity of the site, it would be more appropriate to apply a three-dimensional (3D) flow and dispersion model taking upwind sources of ^{222}Rn from Japan and east Asia into account to the K-site. Since there are very few observations of exhalation rate of ^{222}Rn within an 80-km radius of the K-site, and since the three-dimensional (3D) flow and dispersion model is not developed for the K-site, we apply a one dimensional (1D) flow and dispersion model to the K-site despite its obvious imperfection.

4.1. NIGHTTIME

It is noted, from Fig. 3a, that the measured concentrations of ^{222}Rn lie from the midpoint of the concentrations calculated using the method of Beck and Gogolak [9] for the case of strong stability (IWN) and for the case of weak stability (WNN) to those for the case of weak stability (WNN) at night. These concentrations were lower than those at Akawase [3] and at Uchio [1].

In Fig. 3b, we show the time variation of wind speed in 1-hour averages measured with the ultrasonic anemometer near the surface. The wind speed varied between 0.5 and 7.6 ms^{-1} with the

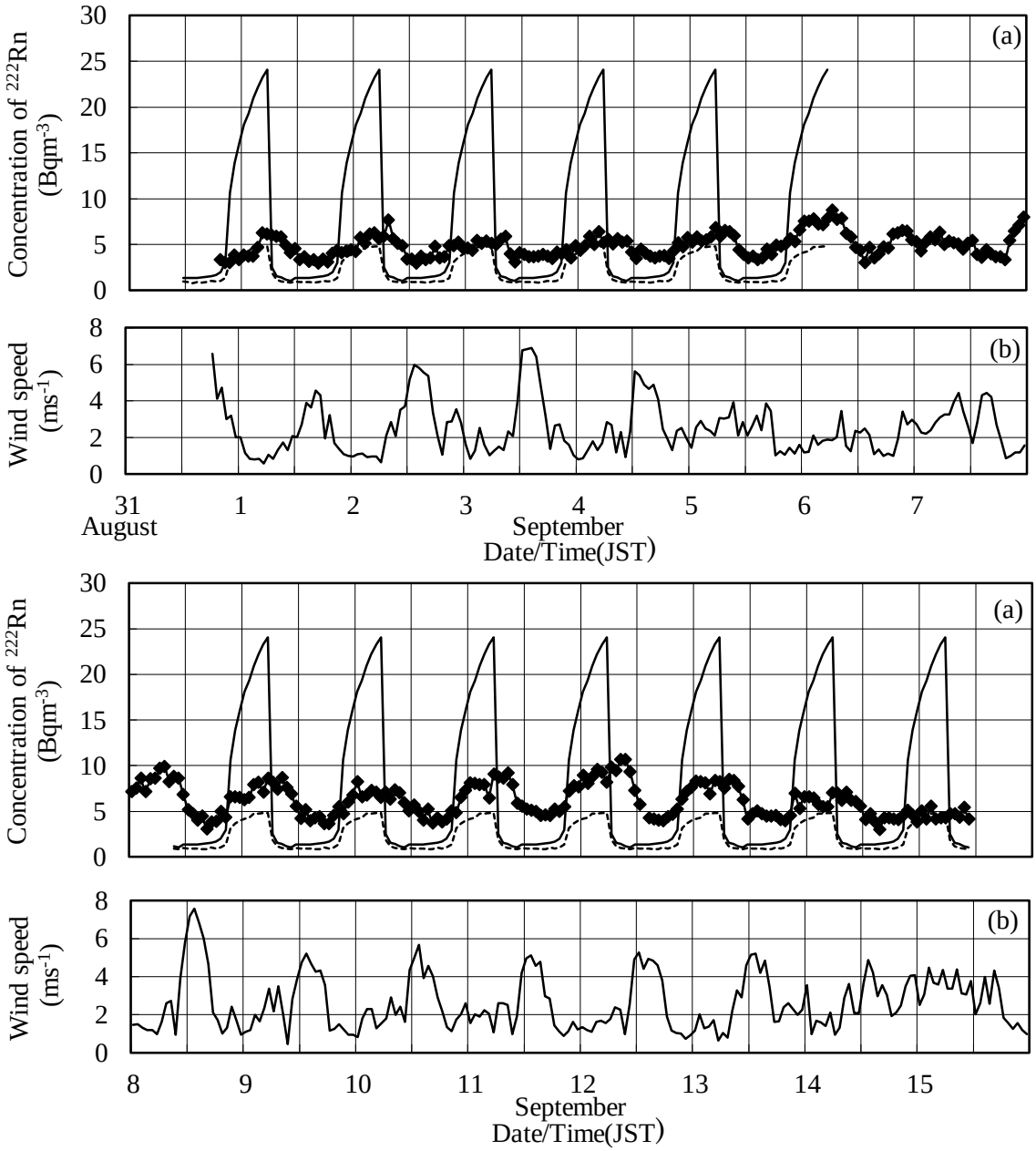

Figure3. (a) Time variations of the concentrations of ^{222}Rn at the K-site. —◆—: Measured concentration of ^{222}Rn, ————: Concentration of ^{222}Rn calculated using the method of Beck and Gogolak [9] for the cases ranging from neutral stability in the daytime (NNN) to strong stability at night (IWN), --------: Concentration of ^{222}Rn calculated using the method of Beck and Gogolak [9] for the cases ranging from strong instability in the daytime (SSN) to weak stability at night (WNN). (b) Time variation of wind speed measured with an ultrasonic anemometer at the K-site. The readings of the wind represent 1-hour averages taken during the same intervals as those of the concentrations of ^{222}Rn.

exception of a single one-hour period, and was larger than that at the U-site (see Fig. 1) [1]. Echoes from the Doppler sodar revealed the existence of surface-based inversion layers extending to heights between 100 and 200 m all nights for the observation periods by the Doppler sodar, that is, during the nights of 3–5 and 8–10 September 1998. It was cloudy during the night of 31 August – 1 September and the wind was weak from a little before dawn. It was again cloudy and the wind was weak during the night of 5–6 September, and the atmospheric conditions were close to neutral stability both of these nights. It was clear during the nights of 1–5 and 8–14 September, and weak mixing due to a land breeze was stronger at the K-site than at the U-site, while the surface based inversion, which existed at the base of the land breeze was weaker at the K-site than at the U-site. Therefore, the atmospheric conditions at the K-site were closer to neutral stability during these nights than those at U-site. During the night of 14–15 September, it was cloudy and the wind was strong, resulting in close-to-neutral stability. Consequently, at night, the concentrations of ^{222}Rn at the K-site much more nearly approximated the concentrations calculated using the method of Beck and Gogolak [9] for the case of weak stability (WNN) than those at the U-site.

At the K-site, although there were cases of horizontal circulation of the land breeze with low frequency, the wind directions of the land breeze were predominant between NNE and ENE during the nights of the intensive observation period, as shown in Fig. 2. Although dilution due to the circulation of the land breeze and its return flow affects the concentration of ^{222}Rn near the surface, the variation of the concentration of ^{222}Rn near the surface is considered to be mainly due to the vertical turbulent diffusion at the K-site, as at the U-site [1].

Asphalt and water inhibit the exhalation of ^{222}Rn from the ground. However, taking a level of weak stability into consideration, it is likely that the apron, runway, ditch and waterway had little or no influence on the concentration of ^{222}Rn in the case of the land breeze.

4.2. MORNING CALM (TRANSITION PERIOD FROM A LAND BREEZE TO A SEA BREEZE)

We find from Figure 3a that the measured concentrations of ^{222}Rn started decreasing at 0800-1000 JST and reached a low level after about 1100 JST. Fig. 4 shows typical time variations of wind speed and standard deviation (σ_w) of vertical wind speed observed with the Doppler sodar at the K-site for the periods of 3–5 and 8–10 September 1998. The land breeze weakened around sunrise (about 0550 JST), the morning calm appeared and then changed to the sea breeze at 1015–1115 JST (1015 JST in Fig. 4a). During both periods where the observation with the Doppler sodar was carried out, variations of the vertical wind speed at night were small and σ_w was less than 0.3 ms^{-1}, as represented by Fig. 4b. Regions where σ_w was above 0.3 ms^{-1} developed rapidly in the upper air after sunrise. We take this value of σ_w to be a suitable indication of the depth of the convective mixing layer. On this basis, the depth of the convective mixing layer reached about 700 m at 0900–1030 JST (1030 JST in Fig. 4b) during both these periods. However, it is conjectured that thermal updrafts developed very easily at the location of the Doppler sodar, based on the fact that horizontal winds were very weak near the surface (0–200 m) during the morning calm after sunrise as shown in Fig. 3b and 4a, and that the runway and apron are constructed of asphalt, in contrast to bare fields and pasture around the Air-Station. The Doppler sodar may have detected the mixing layer developing more strongly due to this phenomenon. Therefore, it is believed that the relatively calm meteorological conditions during the transition period from land breeze to sea breeze are responsible for the observation that the concentration of ^{222}Rn decreased more slowly at the K-site than at the A-site (see Fig. 1) [3], despite development of the mixing layer after sunrise, as in the case of the U-site [1].

4.3. DAYTIME

The measured concentrations of ^{222}Rn were larger than the calculated ones in the daytime when the sea breeze was dominant after the morning calm (see Fig. 3a). This tendency is similar to that at the A-site which is located about 22 km south of the coast of the Sea of Japan and forms a basin at a ridge of the Chuhgoku mountains [3] and at the U-site which is located about 12 km north of the coast of the

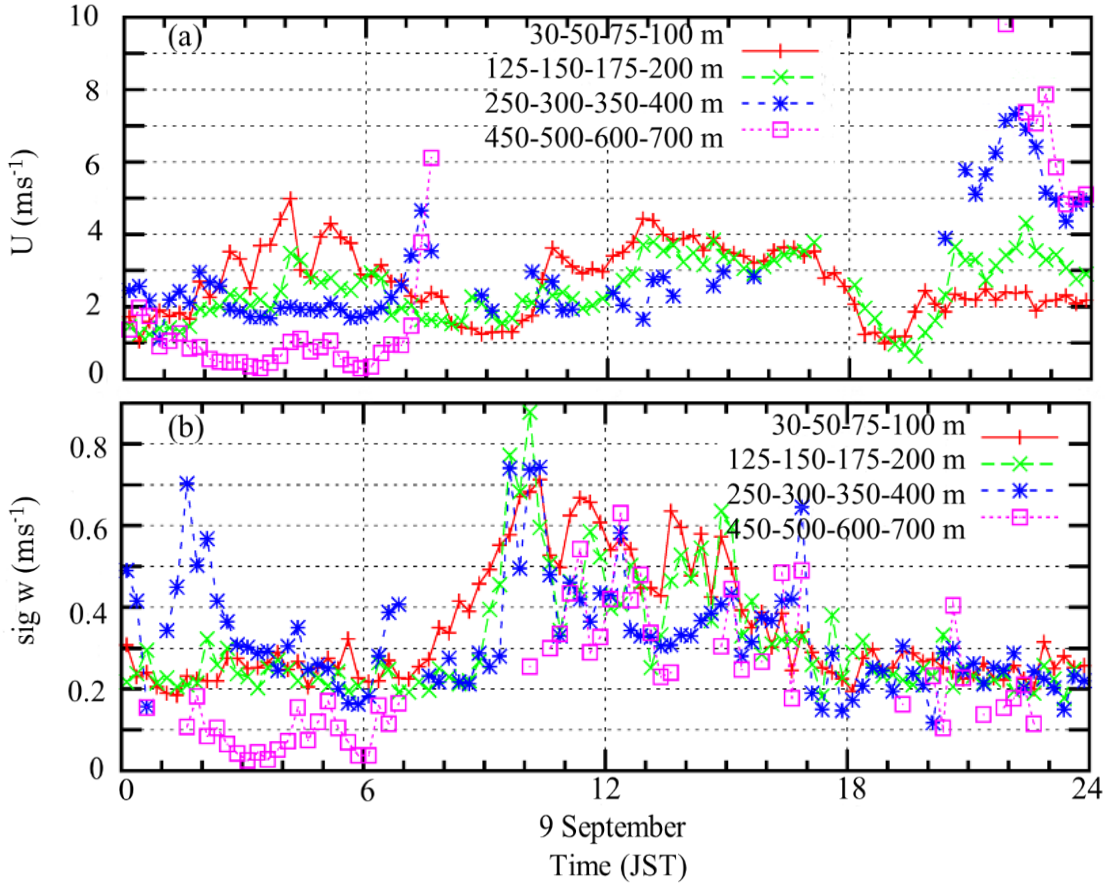

Figure 4. Time variations of (a) wind speed and (b) standard deviation of vertical wind speed obtained with a Doppler sodar at the K-site on 9 September 1998. 30-50-75-100 m : Average value of measurements at 30, 50, 75 and 100 m. 125-150-175-200 m : Average value of measurements at 125, 150, 175 and 200 m. 250-300-350-400 m : Average value of measurements at 250, 300, 350 and 400 m. 450-500-600-700 m : Average value of measurements at 450, 500, 600 and 700 m.

Inland Sea and is in the east of Okayama Plain [1]. Considering the wind from the Sea of Japan or the Inland Sea having lower exhalation rate of ^{222}Rn than land, the measured concentrations of ^{222}Rn are expected to be smaller than the calculated ones. However, in fact, it was the other way about. Since mixing is large in the daytime, the measured concentrations of ^{222}Rn contain considerably more ^{222}Rn exhaled at areas more than 80 km away from the experimental site [11]. This is the cause of the difference between the concentrations calculated and measured. Unfortunately, we can not apply a three-dimensional (3D) flow and dispersion model taking distant sources from Japan and east Asia into account, such as Nagoya City [11], to the K-, A- and U-sites.

5. Air pollutants, and land and sea breezes

In Fig. 5, we show time variations of the concentrations of NO, NO_2 and SO_2 at observatories 1, 2, 3 and 4 for the period from 7 to 13 September. We find that there are increases in the concentrations of NO and NO_2 at observatory 1 at 0700 or 0800 JST and at 1700 or 1800 JST on weekdays (7–11 September) due to heavy traffic during the morning and evening rush hours, respectively. Noticeable increases also appeared in the concentrations of NO and NO_2 at observatory 2 on weekdays due to activity at Kasaoka City including traffic conditions and due to the morning and evening calm. It is also noted that the concentrations of NO and NO_2 increased at observatory 3 at 0800–0900 JST on weekdays due to the morning rush hour. Since these increases are matters extraneous to advection, we eliminate them from the following analyses.

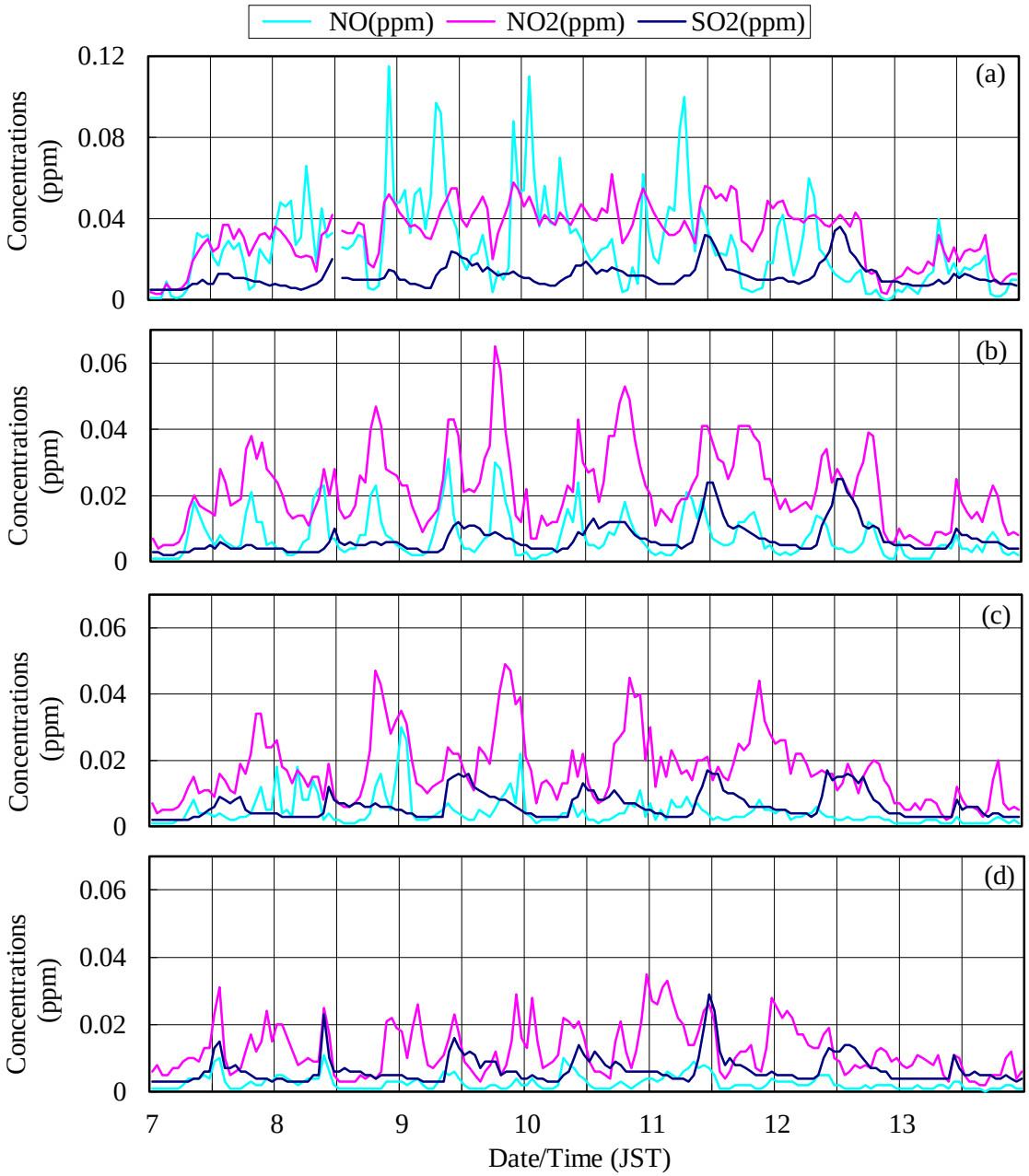

Figure 5. Time variations of the concentrations of NO, NO_2 and SO_2 at (a) observatory 1, (b) observatory 2, (c) observatory 3 and (d) observatory 4.

5.1. NIGHTTIME

Levels of the concentrations of NO and NO_2 increased substantially while levels of the concentration of SO_2 increased slightly at observatory 1 during the nights of 7–12 September, especially during the nights of 8–10 September. This indicates that Route No. 2 had heavy traffic during all of these nights, and that, although the surface-based inversion was rather weak here as mentioned in Section 4, NO, NO_2 and SO_2 emitted from motor vehicles accumulated in the surface-based inversion layer. The wind direction was NE–NNE at observatory 2 and the peaks of NO, NO_2 and SO_2 that appear in Fig. 5a do not appear in Fig. 5b except by coincidence during each of these nights. Therefore, we conclude that the concentrations of NO, NO_2 and SO_2 at observatory 2 were not affected by traffic on route No. 2 during these nights.

From Fig. 5c, we notice that significant peaks appeared in the concentrations of NO and NO_2 at observatory 3 during the nights of 7–13 September. The timing of these peaks was a little different from those at observatory 1. The wind direction was N–NE (see Fig. 2) except for 0300–0500 JST and

2400 JST on 10, and 0200 JST on 11 September at observatory 3 during the nights of 7–13 September. Observatory 3 was strongly affected by traffic on main local road route No. 3. The surface-based inversion was rather weak, and, as a consequence, it is likely that observatory 3 was only weakly affected by traffic on national road route No. 2. It is considered, from these observations, that the effect of route No. 3 concealed any influence of route No. 2.

On the other hand, it is noted, from Fig. 5a and 5d, that during the nights of 7–13 September, patterns of the concentrations of NO, NO_2 and SO_2 at observatory 4 are similar to those at observatory 1. Typical wind directions at the observatories at these nights are given in Fig. 2. In addition, the land breeze blew mainly along the waterway at the KAM-station as mentioned before. Therefore, it seems that the wind passing route No. 2 near the waterway reaches observatory 4.

5.2. MORNING CALM (TRANSITION PERIOD FROM LAND BREEZE TO SEA BREEZE)

It is noted, from Fig. 5d, that the concentration of SO_2 increased together with increases in the concentrations of NO and NO_2 as the wind shifted from NE immediately before 0900 JST to WNW at 1000 JST, and decreased together as the wind shifted from WNW at 1000 JST to SW at 1100 JST, on 8 September at observatory 4. The same phenomenon was observed at observatory 4 on 31 October 1998. When sea breezes were observed at observatory 3 during the period of 0900–1100 JST, the concentration of SO_2 often increased (on 5, 6, 8–12, 14 September). The concentrations of NO and NO_2 also increased significantly. Furthermore, when the wind direction was W–NW at observatory 4 during some hours including the morning calm, the concentrations of SO_2, NO and NO_2 often increased. Considering the wind directions, the peaks, which appeared in the concentrations of SO_2, NO and NO_2 at 1000 JST at observatory 4 and at 1100 at observatory 3 on 8 September, may have been due to Fukuyama Industrial Complex, ships on the waterway or lying in New Kasaoka Port.

5.3. DAYTIME

Comparing the time variations of concentration of SO_2 at observatory 1 and 2 (see Fig. 5a and 5b), we find that they have a similar pattern in the daytime. In addition, when peaks appeared simultaneously at observatories 1 and 2, the wind direction was SW – S at observatory 2 (see Fig. 2). Therefore, it seems that the peaks were mainly due to ships lying in a small port and on the waterway which lie between southwest and south from observatories 1 and 2.

From Fig. 2 and 5c, peaks which appeared at observatory 3 may have been mainly due to Fukuyama Industrial Complex which lies between southwest and south-southwest from observatory 3, and ships on wharfs of berths and on the waterway on the east of the complex.

When peaks appeared in the concentration of SO_2 at observatory 4 (see Fig. 5d), the wind direction was, for the most part, SSE. There is a valley between observatory 4 and a factory manufacturing sulfuric acid, as shown in Fig. 2. Therefore, it is thought that the peaks were due to the factory.

In the daytime, there existed cases in which the peaks in the concentration of SO_2 originating from factories and ships were accompanied by peaks appearing in the concentrations of NO and NO_2 such as those at observatory 3 at 1100 on 8 September, at observatory 4 at 1400 on 7 September, and at 1000 on 8 September. However, most peaks in the concentration of SO_2, except for these 3 peaks, were accompanied by peaks in the concentration of NO_2, but were not accompanied by peaks in the concentration of NO. This may be due to the fact that NO emitted by motor vehicles hid the peaks in the concentration of NO emitted by factories and ships, and due to the fact that NO decreased by the reaction of NO with oxidants. Two notable examples of these phenomena are the peaks in the concentrations of SO_2 and NO_2 at observatory 1 at 1100 JST on 9 September and 1400 JST on 12 September.

6. Conclusions

The concentrations of ^{222}Rn at night were lower than those at Akawase and Uchio reported in previous papers [1,3], and ranged from the midpoint of the concentrations calculated using the method of Beck and Gogolak [9] for the case of strong stability and for the case of weak stability to those for

the case of weak stability. Smaller increases of the concentration of ^{222}Rn at night and slow decreases of the concentration of ^{222}Rn during the morning calm, namely the transition period from land breeze to sea breeze could be also clearly explained by meteorological observations including Doppler sodar. In the daytime, the concentration of ^{222}Rn was almost the same as those at Akawase and Uchio.

At night, although atmospheric conditions were near weak stability, advection of exhaust fumes originating from motor vehicles was identified from analysis of time variations of concentrations of NO, NO_2 and SO_2, and wind directions around the experimental site. In the daytime including the morning calm, exhaust fumes originating from factories and ships were also identified by the analysis.

These results support a suggestion of a previous paper [1] that a set of atmospheric ^{222}Rn and air pollutants would be a useful indicator for the local nature of the atmospheric boundary layer.

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