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Daily variation of aerosols of marine and continental origin in the surface air over a small island, Okushiri, in the Japan Sea

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ABSTRACT

Daily aerosol samples were collected for 6 months from 25 November 1984 at Okushiri Island in the Japan Sea. The samples were analyzed for aluminum (an indicator of mineral aerosol) and sodium (an indicator of sea salt aerosol), and the following results and conclusions were obtained. The atmospheric concentration of aluminum was higher in spring than winter, while the concentration of sodium in winter decreased toward spring. The concentration of sodium was positively correlated with westerly wind force but the concentration of aluminum was not correlated with the wind force. According to a harmonic analysis of the observed data, the concentration of aluminum showed a 4-day periodicity, but we could not find any remarkable periodicity for sodium. These results suggest that the mineral aerosols in the surface air over the Japan Sea were transported across a long distance, probably from the arid regions of the Asian continent with continental air masses changing its direction gradually, and that a large amount of sea salt aerosol is produced in the Japan Sea during the winter monsoon and transported directly to the coastal area along the Japan Sea.

1. Introduction

The aeolian transport of terrigenous materials to the Pacific ocean has been observed indirectly by studies of lead-210 (Tsunogai and Nozaki, 1971; Nozaki and Tsunogai, 1973; Nozaki et al., 1976), by particulate aluminum (Uematsu et al., 1985a) in the surface water and by quartz in the deep-sea sediments (Rex and Goldberg, 1958), among others. There are also some reports on the observation of atmospheric soil particles in the surface air (Duce et al., 1980; Rahn et al., 1981) and above the boundary layer (Darzi and Winchester, 1982; Parrington et al., 1983) over the Pacific ocean, which may be transported from arid regions in the Asian continent.

Recently, US and Japanese scientists performed a cooperative network observation for a period of 2 years in order to clarify the transport process quantitatively (Uematsu et al., 1983, 1985b; Tsunogai et al., 1985). They collected weekly aerosol samples and found that spring peaks of atmospheric mineral dust are simultaneous at all the stations in the North Pacific and are accompanied by *Kosa* episodes. Tsunogai and Kondo (1982) have previously found a large daily variation of atmospheric concentration of aluminum on board the R/V *Hakuho Maru* (30°N, 161°E), and it seems to be difficult to discuss the transport process in detail with weekly aerosol samples.

We have therefore carried out daily aerosol sampling for 6 months during the winter monsoon at the Okushiri Island in the Japan Sea and performed chemical analyses to clarify the transport process of aerosols of marine and continental origin.

2. Sampling and analytical method

Two sampling systems were set on the roof of a building, the Kamuiwaki public resting place on the west coast of Okushiri Island (42°09′N,

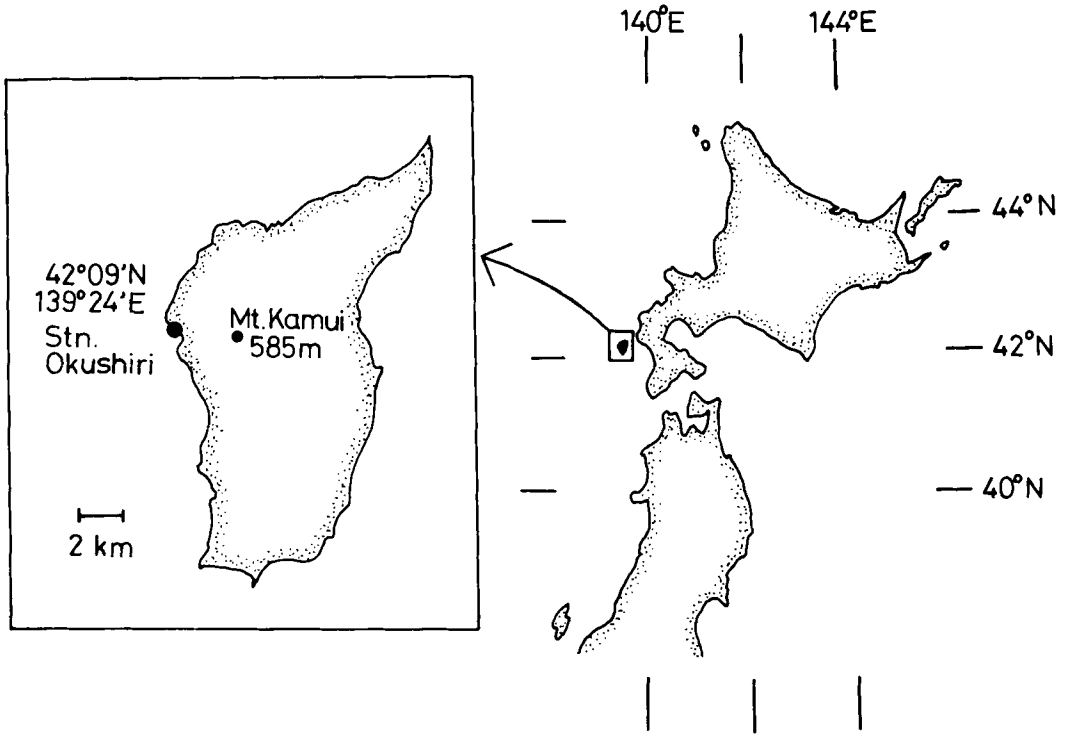

Fig. 1. Sampling stations at Okushiri Island (solid circles).

139°24′E). The samplers were about 6 m above the ground, facing directly into the westerlies from the Asian continent (Fig. 1). This station is the same as that used by Tsunogai et al. (1985). The ground is about 2 m above mean sea level and open to the sea in the 160°–340° sector; the distance to the shore line is 5 m.

The sampling apparatus which was similar to that used by Tsunogai et al. (1985) and duplicate aerosol samples were collected on Whatman no. 41 filters (20 × 25 cm) by sucking air continuously with a flow rate of 900–1100 l min^{-1} without a wind sensor. The filters were changed once a day at 7.30 a.m. The sampling continued for 187 days, from 25 November 1984 to 30 May 1985. On 25 December we failed to change the filters because of a terrible blizzard. The total number of samples is therefore 186, including one 2-day sample. A quarter of each sample filter was used in this study. Another quarter was stored for future use and the remainder (one half of the filter) was used for heavy metal analysis by Professor K. A. Rahn at the University of Rhode Island, USA. Another complete set of filters was used for the determination of lead-210.

A sample filter was dissolved with a mixture of hydrofluoric acid, nitric acid and perchloric acid in a sealed teflon vessel (Noriki et al., 1980). Aluminum and sodium were determined by atomic absorption methods. Aluminum and sodium are contained almost exclusively in mineral and sea-salt particles, respectively.

3. Results and discussion

3.1. Daily variation of atmospheric concentration of aluminum and sodium

The observed atmospheric concentration of aluminum and sodium are illustrated in Fig. 2, together with the daily mean wind direction and speed which were measured at Mt. Kamui (585 m MSL, about 3 km east of the Okushiri station in Fig. 1) from 1 December. Their monthly,

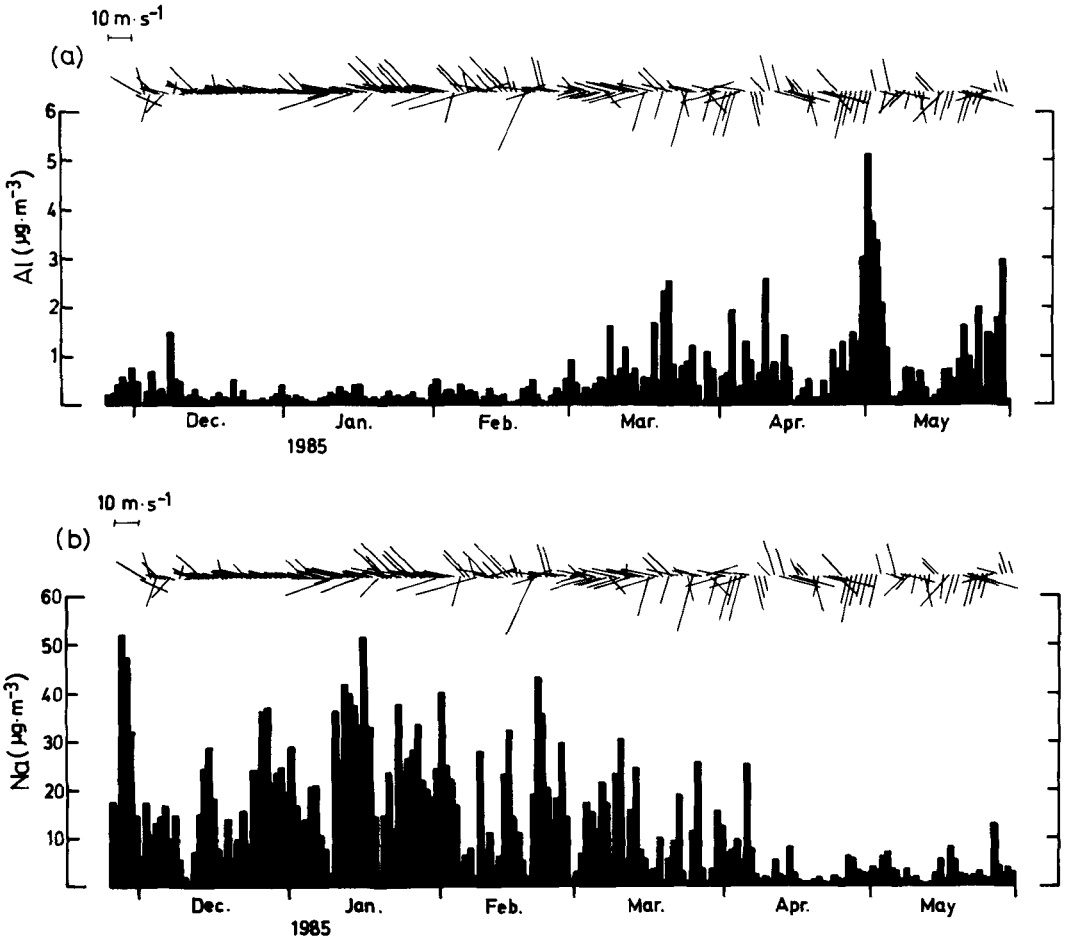

Fig. 2. Atmospheric concentration of (a) Al and (b) Na at Stn. Okushiri with wind direction and wind speed which were measured at Mt. Kamui.

trimonthly and total arithmetic mean concentrations with standard deviations ($\pm 1\ \sigma$) are given in Table 1.

As shown in Fig. 2a, the aluminum concentration in spring (1 March–30 May 1985) is higher than in winter (25 November 1984–28 February 1985). Both the monthly mean concentration and the relative standard deviation of aluminum are at a minimum ($0.20 \pm 0.09\ \mu g\ m^{-3}$) in January and maximum ($1.2 \pm 1.2\ \mu g\ m^{-3}$) in May (Table 1). The largest concentration of aluminum in the daily samples ($5.13\ \mu g\ m^{-3}$) was observed on 1–2 May, about one order of magnitude larger than overall mean concentration. This result is consistent with the conclusion of

Table 1. *Average concentrations of aluminum and sodium with their standard deviations*

	Al	Na ($\mu g\ m^{-3}$)
Nov., 1984	0.47 ± 0.19	30 ± 15
Dec., 1984	0.30 ± 0.27	16 ± 9.0
Jan., 1985	0.20 ± 0.09	24 ± 12
Feb., 1985	0.23 ± 0.14	17 ± 12
Mar., 1985	0.77 ± 0.58	11 ± 8.2
Apr., 1985	0.85 ± 0.69	4.1 ± 4.7
May, 1985	1.2 ± 1.2	3.4 ± 2.6
Nov., 1984–Feb., 1985	0.26 ± 0.20	19 ± 12
Mar., 1985–May, 1985	0.94 ± 0.88	6.2 ± 6.6
Total	0.59 ± 0.72	13 ± 12

Uematsu et al. (1983) and Tsunogai et al. (1985), that the mineral dust in the surface air over the North Pacific is chiefly transported from the arid region in the Asian continent in spring. However, the maximum daily concentration of aluminum obtained in this study is only about one third of the maximum observed previously on 16–23 April 1983 at Okushiri, 16.3 μg m^{-3} (Tsunogai et al., 1985). This suggests that the contribution of the Kosa episodes is highly variable from year to year in Japan, as observed by Matsumoto (1983, 1985).

The concentration of sodium in the atmosphere varied differently from that of aluminum, showing higher values in winter than in spring, as also reported by Toba (1966) and Erickson et al. (1986) (Fig. 2b). The maximum monthly mean of the concentration of sodium was 30 ± 15 μg m^{-3} in November and the minimum was 3.4 ± 2.6 μg m^{-3} in May (Table 1). This result agrees with the inference made by Tsunogai et al. (1972), Tsunogai (1975) and Tsunogai and Shinagawa (1977), that a large amount of sea salt particles is transported to the coast of the Japan Sea in winter due to the strong north-west monsoon.

3.2. *Correlation between atmospheric concentration of aluminum or sodium, and velocity of westerly winds*

We have examined the relationship between the concentration of aluminum or sodium in the atmosphere and the velocity of the westerly wind. To express the intensity of westerly wind, we define the wind factor (W_f) by the following expression:

$$W_f = W_s \cos|W_d - 285|, \quad (1)$$

where W_s is the wind speed in m s^{-1} and W_d is the wind direction in degrees. We set 285° as a standard for the wind direction, because this angle is typical of the winter monsoon and is the center of the sector open to the sea at the station.

We find no correlation between the concentration of aluminum and the wind factor, in either winter or spring (Fig. 3a). This may be explained by the fact that the atmospheric mineral dust originates in the remote Asian continent and is transported to the station through various pathways, changing gradually with the direction of wind.

On the other hand, as shown in Fig. 3b, the concentration of sodium is well correlated with the wind factor. The correlation is expressed by the following equation obtained by a least squares method:

$$Na = 4.04 \exp(0.09 W_f), \quad (2)$$

where Na is the concentration of sodium in the atmosphere (μg m^{-3}). Tsunogai et al. (1972) have also found that the atmospheric concentration of sea salt on board the ship in the Pacific increases exponentially with the wind speed as follows:

$$S = 0.33 \exp(0.62 W_s), \quad (3)$$

where S is the sea salt concentration in the atmosphere (μg m^{-3}). Although the absolute con-

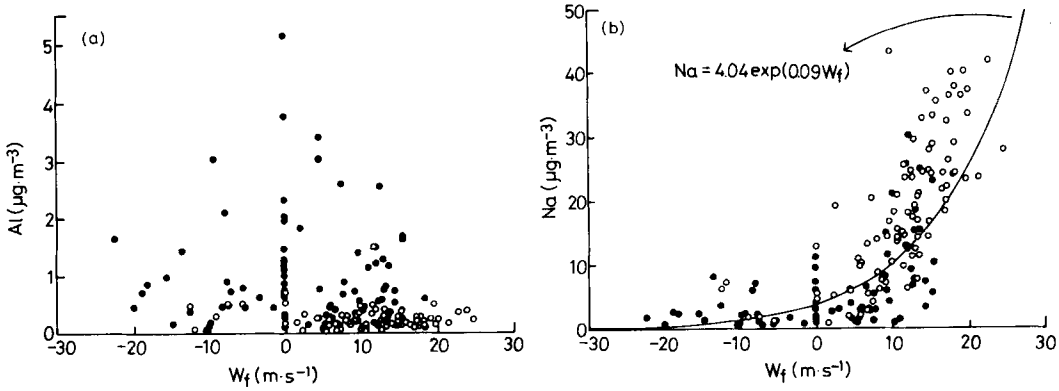

Fig. 3. Relationship between W_f (see text) and (a) Al or (b) Na. The solid circles refer to the spring season (March–May 1985) and the open circles to the winter season (Nov 1984–Feb. 1985). The curve in Fig. 3b is obtained by a least squares method.

centration depends on the local conditions, the wind dependency in (2) is much smaller than that of (3). This may be chiefly caused by sea salt aerosol transported by the wind from other directions, because $W_f = 0$ does not imply $W_s = 0$. Although the wind dependency is highly variable, depending on the sampling condition, it is certain that the production of sea salt particles is ultimately related to wind speed (Woodcock, 1953; Blanchard, 1963; Toba, 1965; Savoie and Prospero, 1977; McDonald et al., 1982) and a larger part of sea salt particles deposit near the source area (Tsunogai et al., 1972; Tsunogai, 1975; Tsunogai and Shinagawa, 1977), and that the strong north-west monsoon in winter brings most of the annual sea salt deposition in the coastal area along the Japan Sea.

3.3. *Harmonic analysis of atmospheric concentration of aluminum, sodium, and wind factor*

We have examined periodicity in the daily variations of aluminum and sodium concentrations and wind factors with the aid of harmonic analysis (Bendat and Piersol, 1971; Hino, 1977). The data obtained in this study can be expressed as a function of days and expanded in a Fourier series, which gives the dominant period as that having a maximum amplitude.

If we divide the total interval, $0 \leqslant x \leqslant 2T$, into k equal fractions, x_0, x_1, x_2, ..., x_i, ..., x_k ($x_0 = 0$, $x_k = 2T$) and assign the value of $f(x_i)$ to each fraction, the function, $F(x)$, is expressed by:

$$F(x) = a_0 + \sum_{n=1}^{2T} \{a_n \cos(n\pi x T^{-1}) + b_n \sin(n\pi x T^{-1})\}. \quad (4)$$

In eq. (4), a_0, a_n and b_n are the Fourier coefficients and

$$a_0 = k^{-1} \sum_{i=1}^{k} f(x_i), \quad (4.1)$$

$$a_n = 2k^{-1} \sum_{i=1}^{k} f(x_i) \cos(n\pi x_i T^{-1}), \quad (4.2)$$

$$b_n = 2k^{-1} \sum_{i=1}^{k} f(x_i) \sin(n\pi x_i T^{-1}), \quad (4.3)$$

where x_i is the x_ith day from the start of observations, and $f(x_i)$ is the observed value for the x_ith day in $\mu g\ m^{-3}$. After dividing the observation period into 2 seasons, i.e., the winter season (25 November 1984–28 February 1985, $2T = 96$, for aluminum and sodium and 1 December 1984–28 February 1985, $2T = 90$, for the wind factor), and the spring season (1 March–30 May 1985, $2T = 91$), we calculate a_0, a_n and b_n values for aluminum, sodium, and the wind factor.

Fig. 4 shows the power spectrum, the sum of squares of the Fourier coefficients, $a_n^2 + b_n^2$, plotted against the period (number of days up to 30 days).

The power spectrum of aluminum does not show a definite trend in winter, but the curve has a remarkable maximum at $n = 4$ in spring (Fig. 4a). This means that variation of the atmospheric concentration of mineral dust does not vary periodically in winter, but the concentration shows maxima about every 4 days with a high probability in spring. This periodicity coincides with the period of air mass exchange in the spring season in the mid-latitudes of eastern Asia, showing that the mineral dust in the maritime air over the North Pacific originates over the Asian continent. Furthermore, since the maximum value varies widely from case to case, the amount of Kosa transported to the Pacific Ocean is expected to vary widely from year to year depending on the scale and number of Kosa episodes (Tsunogai et al., 1985). To quantitatively explain the maximum value, we need detailed information on the scale of dust storms in the Asian continent and the meteorological conditions which control the transport process.

The power spectrum for sodium in the winter season has a maximum at $n = 10$. However, since the curve varies widely, we cannot safely conclude that the variation of sea-salt concentration has a clear periodicity (Fig. 4b). In spring, a maximum value of the curve can be seen at $n = 1$, which may indicate that the sea-salt concentration in spring varies within a one-day period and that the periodicity cannot be found from daily samples.

Fig. 4c is the power spectrum for the wind factor. The spectra for both the seasons are similar to that of sodium in the spring season. That is to say, the daily variation of the wind factor is not periodic or a noisy phenomenon; the same holds for the sodium concentration in the spring season. Since the daily samples are insuf-

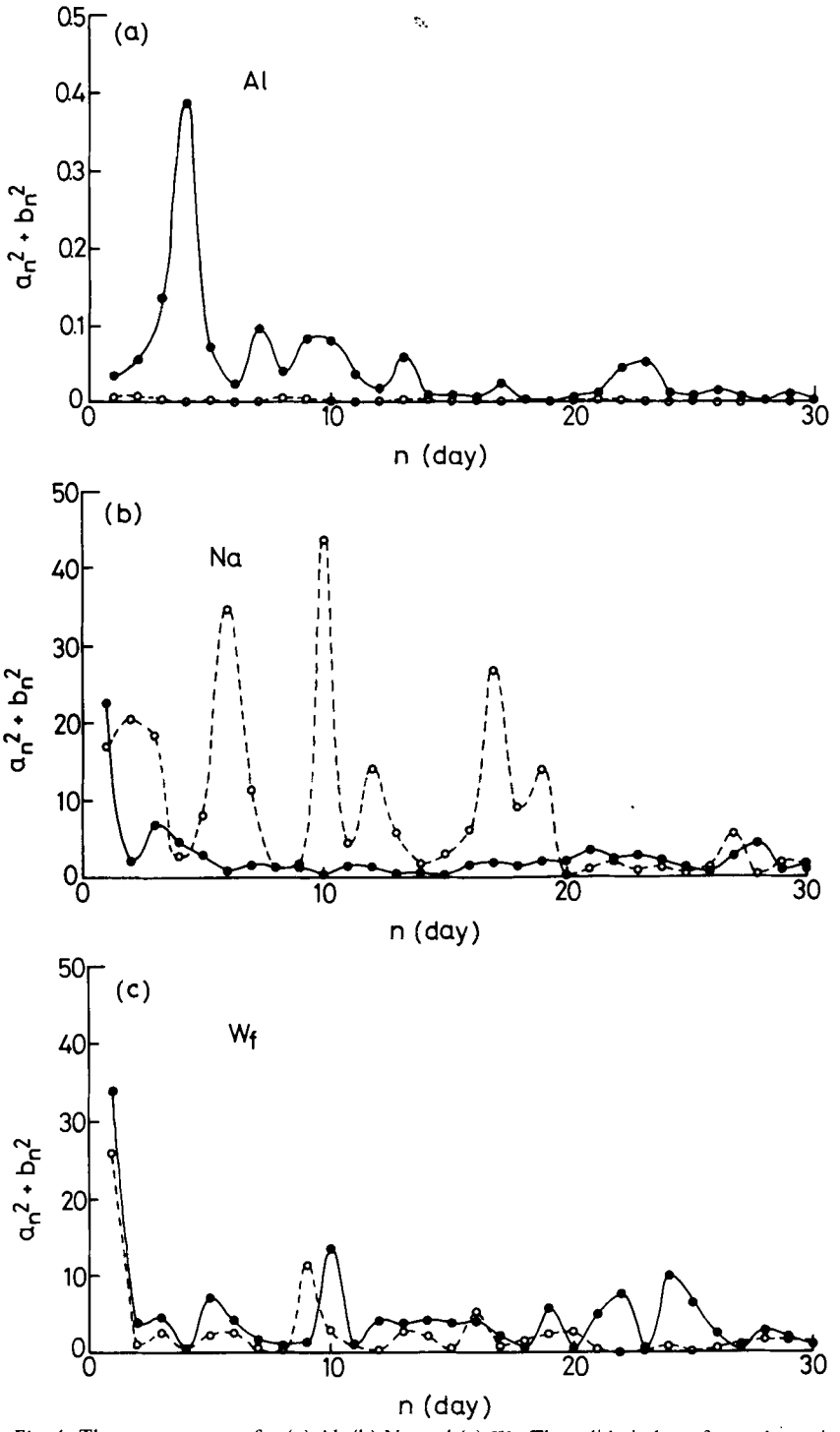

Fig. 4. The power spectra for (a) Al, (b) Na and (c) W_f. The solid circles refer to the spring season (March–May 1985) and the open circles refer to the winter season (Nov. 1984–Feb. 1985).

ficient to determine the periodicity of the concentration of sodium, and the wind force, we could find their periodicity by observing over shorter time intervals.

4. Conclusion

Our continuous measurements of the atmospheric concentrations of aluminum and sodium at Okushiri Island in the Japan Sea for 6 months during the winter monsoon lead to the following conclusions.

(1) The atmospheric concentration of mineral dust was higher in spring than in winter. The spring peaks of the atmospheric mineral dust occurred periodically about every 4 days in spring. These indicate that the mineral dust is blown up into the atmosphere by dust storms frequent in spring in the Asian continent and transported to the North Pacific ocean by the westerlies.

(2) The atmospheric concentration of sea salt was higher in winter than in spring. The concentration was strongly dependent on the wind speed, and does not show periodicity. These results suggest that a large amount of sea salt aerosol is produced in the Japan Sea during the winter monsoon and transported to the coastal area of Japan facing the Japan Sea.

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