Pb-210 in precipitation in Japan and its implication for the transport of continental aerosols across the ocean

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ABSTRACT

Total precipitation including rain or snow and dry fallout was collected once a month from February 1970 to March 1972 at five sampling stations in Hokkaido located from the coast of the Sea of Japan to inland. The monthly deposition rate of Pb-210 was determined together with other chemical constituents. The results are summarized as follows: (1) The deposition rate and the concentration of Pb-210 are remarkably high in winter. (2) A linear relation between the deposition rate of Pb-210 and the amount of rainfall is found in both summer and winter respectively. (3) As compared with the production rate of Pb-210, namely the exhalation rate of Rn-222 from the continent, the deposition rate of Pb-210 to the unit area at the coast of the Sea of Japan in winter is 2–4 times the exhalation rate of Rn-222 from the continent. (4) The budget calculation of Pb-210 in the atmosphere shows that about 40 % of aerosols transported from the continent are caught by snow and deposited over the Sea of Japan and the Japan Islands in winter. Therefore, it is concluded that the Japan Islands play an important role as a collector of the continental aerosols in winter.

1. Introduction

Rex & Goldberg (1958) observed a large amount of quartz, unweathered material, in the deep sea sediments collected at the middle latitudes of the North Pacific and pointed out the importance of transport of continental substances via the atmosphere. Recently Tsunogai & Nozaki (1971) and Nozaki & Tsunogai (1973*a*) concluded that Pb-210 in the surface water of the Pacific is originated from the continent via the atmosphere on the basis of the latitudinal variation of Pb-210 which is similar to that predicted by Lambert & Nezami (1965) and its vertical distribution in the ocean.

It is not so easy to collect directly continental aerosols over the ocean. Prospero & Bonatti (1969) collected some samples of the dust (>2 μ size) using the mesh technique on board over the eastern Equatorial Pacific, and Parkin et al. (1970) collected those in the same way over the North Atlantic. Tsunogai et al. (1972), however, could not find continental aerosols in the maritime air by filtering the air with membrane filter (0.1 μ pore size and 90 mm diameter) on board over the Pacific.

The Japan Islands lying at the middle latitudes may have some role on the transport of continental aerosols. The air mass transported from the continent by the north-west monsoon gains lots of moisture over the Sea of Japan, resulting in heavy snow fall in the western coastal area of Japan in winter. Some studies have been made mainly from the physical aspects of this subject by Ninomiya (1964), Asai (1965), Isono et al. (1966), etc.

Recently Ishizuka (1972) found that most of the solid particles in snow on the coast of the Hokuriku district in winter comprizes soil particles originated from the arid or semi-arid regions of North China or Mongolia. There are, however, no quantitative studies on the aerosols transported from the continent.

Some radioactive elements such as Pb-210 (half-life, 21.4 years) seem to be useful for this study. Pb-210 in the atmosphere is a daughter of Rn-222 (half-life, 3.8 days) exhaled from the earth's surface. According to Chamberlain &

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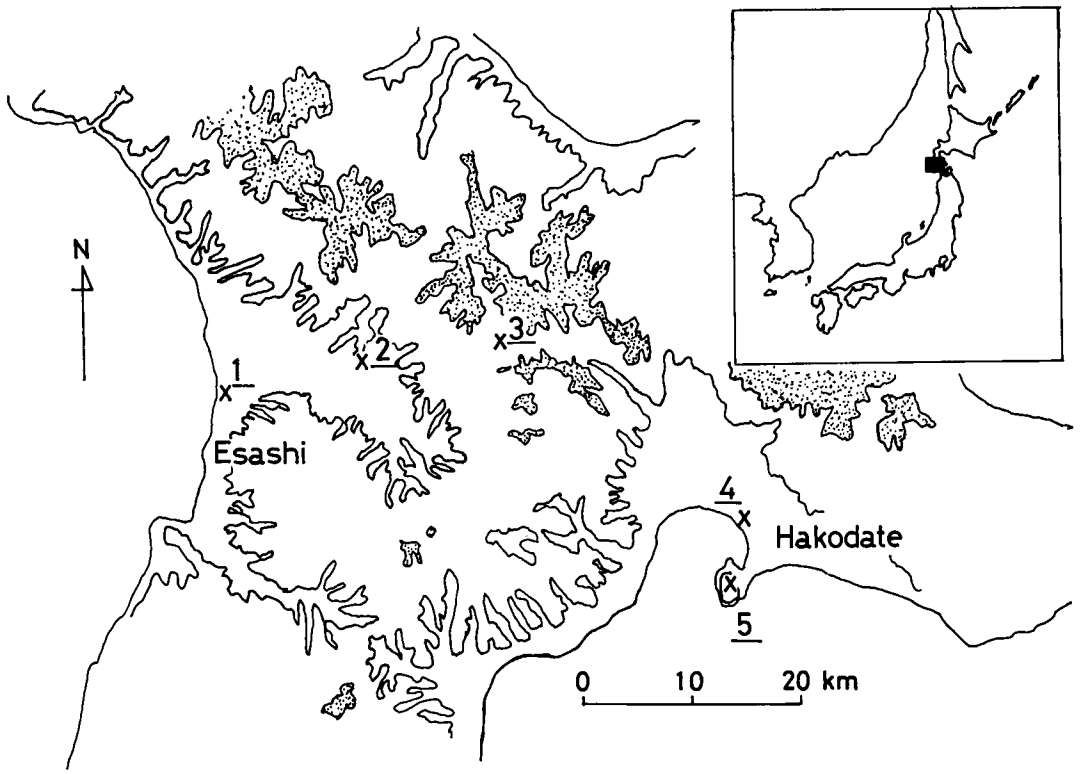

Fig. 1. Sampling stations in the south-western Hokkaido. Contour lines of 0, 100 and 500 m above the sea level are shown.

Dyson (1956) and Lassen & Rau (1960), radioactive nuclides produced in the atmosphere, especially metallic elements having low vapor pressure, adhere to surrounding particles as soon as they are born. These nuclides in aerosols, then, are transported by wind and removed chiefly by precipitation. Vertical profiles of continental aerosols are rather similar to those of Rn-222 (see Junge, 1963). Even though the concentration of Pb-210 in stratospheric aerosols is fairly high (Feely & Seitz, 1970), the contribution of the stratospheric aerosols to the total aerosol deposition is small owing to its long residence time in the stratosphere, say one year (Katsuragi, 1965). Tsunogai & Fukuda (1974) estimated the fraction of Pb-210 of the stratospheric origin in the total deposition to be less than 5%, using Pb-210, Bi-210 and Po-210 in snow produced from the north-west monsoon in winter in Japan. Therefore it is possible to use Pb-210 as a tracer of continental aerosols in the troposphere. Some studies have already been carried out (e.g. Lambert et al., 1966; Feely & Seitz, 1970; Persson, 1970; Peirson, 1971).

Now we tried to study quantitatively the transport of aerosols from the continent to the Sea of Japan, Japan Islands and the Pacific. We collected total precipitation including rain or snow and dry fallout and determined the monthly deposition rate of Pb-210 at five stations from the coast of the Sea of Japan to the inside of the Hokkaido Island.

2. Sampling stations and equipment

Five sampling stations (Fig. 1) were placed between Esashi & Hakodate in Hokkaido, and total precipitation including dry fallout was collected once a month from February 1970 till March 1972.

As shown in Fig. 2, a big polyethylene bag was supported by a tall drum can settled on the plain ground, and the bottom of the bag

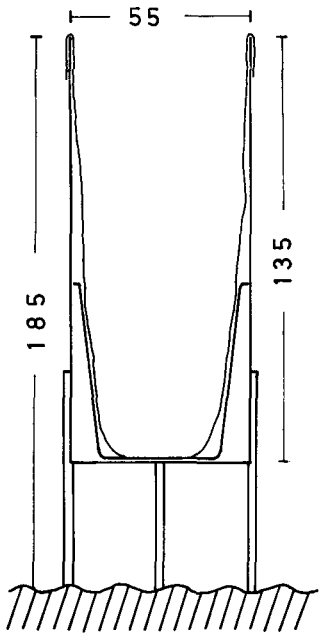

Fig. 2. Sampling equipment. Figures indicate dimensions in cm.

was protected with a polyethylene bucket (60 l). The bucket and the bag were replaced every month. Surface area of the equipment was 0.237 m^2. We have found no instance of narrowing of the area with snow. A sample of the few tens of liters was usually collected every month.

3. Analytical method

After adding a carrier solution of bismuth to the sample, Pb-210 and Bi-210 are coprecipitated as carbonates and hydroxides with ammonium hydroxide and sodium carbonate. The precipitate is dissolved in hydrochloric or nitric acid solution. A small aliquot of the solution is used for the measurements of bismuth to determine coprecipitation efficiency and lead from the air pollution. After adding a carrier solution of lead free from Pb-210 to the remaining solution, hydroxides are precipitated at pH 7 with ammonium hydroxide. The hydroxides are again dissolved with hydrochloric acid. Bismuth hydroxide is reprecipitated at pH 4.5 with ammonium hydroxide. The precipitate is dissolved in hydrochloric acid solution and polonium in the solution is removed by the spontaneous electro-deposition method of Nozaki & Tsunagai (1973*b*). By adding ammonium hydroxide to the solution bismuth oxychloride precipitated at pH 2.5 and the precipitate is mounted on a small circular membrane filter. The β-activity of Bi-210 is counted by using a 2 π gas flow type GM counter with shielding anticoincidence counters and the content of bismuth is also determined.

On the other hand, all the filtrates and washings are combined and the concentrations of lead and bismuth are determined by using an aliquot. A carrier of bismuth is again added to the solution. After allowing to stand for more than 2 months Bi-210 produced from Pb-210 is separated and its β-activity is determined as stated above.

The concentration of bismuth is determined colorimetrically with thiourea (Sandell, 1965) and that of lead is determined with dithizone (Sandell, 1965) or an atomic absorption method. The overall yields for Pb-210 or Bi-210 was 50 to 90%. When a large amount of iron is contained in the sample, the iron is eliminated by the extraction with methyl isobutyl ketone prior to the separation of bismuth from lead.

4. Results and discussion

Fig. 3 shows the monthly deposition rate of Pb-210 (nCi/m^2·month) observed at four sampling stations and the amount of rainfall (mm) observed at the Esashi Meteorological Station (Station 1), the Uzura agricultural weather station (Station 2) and the Hakodate Marine Observatory (Station 4). At Station 3 on the summit of Mt. Hakodate, the sampling often failed because of the strong wind which blew away the snow. The dashed lines in Fig. 3 show the presumed deposition rates which are obtained by multiplying the mean concentration of Pb-210 at neighboring stations by the observed amount of rainfall at the particular station. This is due to the fact that the concentration of Pb-210 does not vary so widely from station to station.

(*a*) *Variation of the monthly deposition rate of Pb-210*

A remarkable peak among the monthly deposition rate of Pb-210 is found in winter as shown in Fig. 3 at first sight. To make this fact

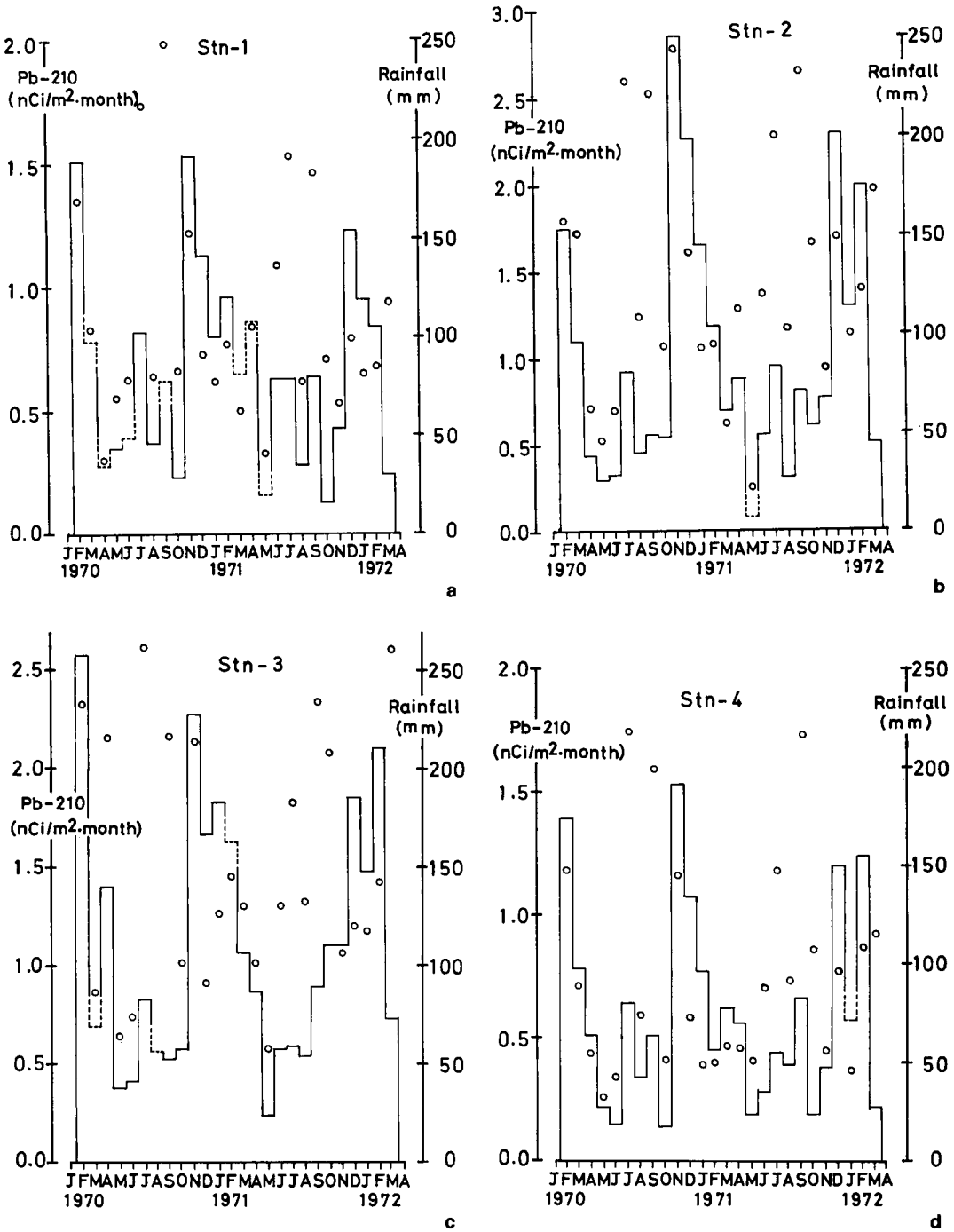

Fig. 3. Monthly deposition rate of Pb-210 and amount of rainfall at station 1 (Fig. 3*a*), 2 (Fig. 3*b*), 3 (Fig. 3*c*) and 4 (Fig. 3*d*). The bar graphs show the deposition rates in nCi/m²·month and open circles show amounts of rainfall in mm/month.

Table 1. *Rainfall in mm, and deposition, monthly deposition rate and mean concentration of Pb-210 in nCi/m², nCi/m²·month and pCi/l, respectively, at the four stations in the south-western part of Hokkaido*

	Summer (May to Aug.)		Winter (Nov. to Feb.)		Whole year (Mar. to Feb.)	
	1970	1971	1970	1971	1970	1971
Station 1						
mm	446	445	416	331	1 325	1 216
nCi/m²	1.93	1.70	4.42	3.45	8.26	7.43
nCi/m²·month	0.48	0.43	1.10	0.86	0.69	0.62
pCi/l	4.3	3.9	10.6	10.4	6.2	6.1
Station 2						
mm	456	444	573	453	1 557	1 443
nCi/m²	2.01	1.93	7.96	6.37	12.61	11.33
nCi/m²·month	0.50	0.48	1.99	1.59	1.05	0.94
pCi/l	4.4	4.3	13.9	14.1	8.1	7.8
Station 3						
mm	539[a]	502[a]	576[a]	487[a]	1 733[a]	1 661[a]
nCi/m²	2.19	1.95	7.40	6.52	12.78	12.38
nCi/m²·month	0.55	0.49	1.85	1.63	1.06	1.03
pCi/l	4.0	3.9	12.8	13.4	7.4	7.4
Station 4						
mm	365	377	316	307	1 075	1 124
nCi/m²	1.35	1.30	3.82	3.39	7.11	6.72
nCi/m²·month	0.34	0.33	0.95	0.85	0.59	0.56
pCi/l	3.7	3.4	12.1	11.0	6.6	6.0

[a] Calculated by adding mean evaporation amount obtained at other stations to the amount of water in the sampler.

more clearly, the deposition rate of Pb-210, the amount of rainfall and the calculated concentration of Pb-210 in precipitation are summarized in Table 1.

In summer (May to August), the deposition rate of Pb-210 is about 0.5 nCi/m²·month regardless of the stations. The mean concentration of Pb-210 in summer rain is 4.0 pCi/l. Moreover, the annual variation of concentration in summer is hardly found in each station.

In winter (November to February), the deposition rate of Pb-210 is 0.9–2.0 nCi/m²·month depending on the stations. This variation may be caused by the amount of rainfall, because the difference in the concentrations among stations is rather small. The mean concentration of Pb-210 in snow is 12.3 pCi/l. The annual variation of the deposition of Pb-210 in winter is also due to the amount of rainfall. The higher concentration of Pb-210 in winter surely indicates that continental aerosols are directly transported over the Japan Islands by the winter north-west monsoon.

(b) Relation of the deposition rate of Pb-210 to the amount of rainfall

Fig. 4 shows the relation of the monthly deposition rate of Pb-210 to the amount of rainfall. We find a marked difference between summer and winter values. The following linear

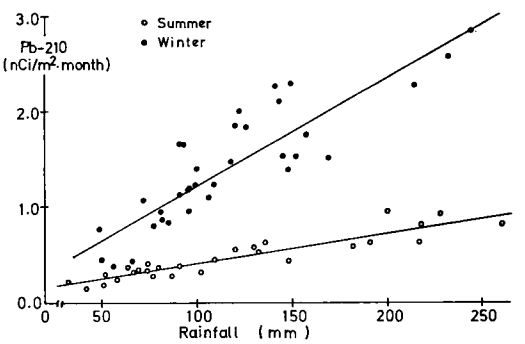

Fig. 4. Relation between the monthly deposition rate of Pb-210 in nCi/m²·month and the amount of rainfall in mm/month. Straight lines are obtained by the least squares' method in each season.

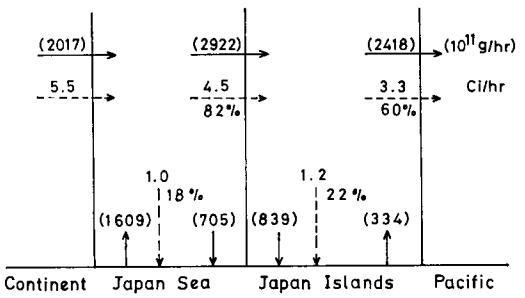

Fig. 5. Budget of Pb-210 and water-substance in the atmosphere over the Sea of Japan and the Japan Islands. Solid lines with arrow heads show the rates for water-substance in 10^{11} g/h and broken lines for Pb-210 in Ci/h.

relations are obtained by using the least square method;

$$y = 0.0031_6x + 0.098 \text{ in summer}$$

$$y = 0.0112_6x + 0.107 \text{ in winter}$$

where y is the deposition rate of Pb-210 and x is the amount of rainfall. The correlation coefficients are 0.92 in summer and 0.87 in winter. The gradient of the winter line is 3.5 times larger than that of the summer line.

The linear relation between the deposition rate of Pb-210 and the amount of rainfall indicates that the deposition rate of Pb-210 depends on the amount of rainfall in the same season, in other words, that Pb-210 in the atmosphere is removed not so much by washout, which is the process below the cloud of falling raindrops, as by rainout which is the process in the clouds. The high correlations observed separately in summer and winter reveal the relatively constant concentration of Pb-210 in each air mass and the difference in the gradients shows that the concentration of Pb-210 in the winter air mass is a few times larger than in the summer air mass.

(c) *Comparison of the deposition rate of Pb-210 with the exhalation rate of Rn-222*

The exhalation rate of Rn-222, a precursor of Pb-210 in the atmosphere, from the earth's surface to the atmosphere has been estimated to be 40×10^{-18} Ci/cm²·sec by Israël (1951). Accordingly the annual production rate of Pb-210 from Rn-222 in the atmosphere is 6 nCi/m²·yr or 0.5 nCi/m²·month.

The annual deposition rate of Pb-210 in Hokkaido (Table 1) is somewhat large as compared with the exhalation rate of Rn-222 from the same area of the continent. The deposition rate of Pb-210 in summer, about 0.5 nCi/m²·month, is nearly comparable to the production rate of Pb-210 in the continental atmosphere, while that in winter, 0.9–2.0 nCi/m²·month, is much higher. Therefore Pb-210 deposited to 1 m² of the coastal area of the Japan Islands in winter corresponds to the Pb-210 produced from 2 to 4 m² of the continent. This indicates that the Japan Islands are a good collector of continental aerosols. We discuss this fact more quantitatively in the next section.

(d) *Estimate of the budget of continental aerosols in winter*

The budget of Pb-210 in the air coming from the continent across the Sea of Japan in winter is made on the basis of the Ninomiya's calculation (1964) for the water-substance budget. Ninomiya (1964) has used the data observed during the period of heavy snow storm in January over the Sea of Japan and the Japan Islands. His schematical results somewhat simplified are reproduced in Fig. 5.

The amount of the air mass transported from the continent to the Sea of Japan is calculated as the sum of the amount of the air mass from the earth's surface to 500 mb level height. The velocity of geostrophic wind (v) is calculated by the following equation,

$$v = \frac{g}{f} \cdot \frac{\partial z}{\partial n}$$

where g is the gravitational constant, f is the Coriolis' factor and $\partial z/\partial n$ is the geopotential gradient between Sapporo and Fukuoka. The transported amount of Pb-210 is obtained by multiplying the amount of the air mass by the content of Pb-210 in the air. As the content of Pb-210 in the continental air, we use the vertical profile of Pb-210 in the air of Jacobi & André (1963) who calculated the contents from those of Rn-222 measured over Ohio, USA by Wexler et al. (1956). The calculated vertical profile of Pb-210 is similar to that directly observed over the continental USA by Moore et al. (1973). The transported amount of Pb-210 from the continent to the atmosphere over the

Sea of Japan was calculated to be 5.5 Ci/h in the season of the north-west monsoon.

The amount of Pb-210 deposited over the Sea of Japan and the Japan Islands are calculated as the product of the amount of rainfall and the concentration of Pb-210 in precipitation. The mean concentration of Pb-210 in snow is calculated to be 13.7 pCi/l from the observed values in January at the four stations. The amounts of Pb-210 deposited over the Sea of Japan and the Japan Islands are estimated to be 1.0 and 1.2 Ci/h, respectively.

As summarized in Fig. 5, Pb-210 in the continental air mass falls on the Sea of Japan by 18% of the total transported amount, subsequently by 22% on the Japan Islands and its 60% fraction is passing to the Pacific. Thus, in the troposphere lower than the 500 mb level height where the horizontal divergence of vapor, liquid and solid water fluxes were negligibly small as discussed by Ninomiya (1964), 40% of aerosols transported from the continent may be collected by the snow over the Sea of Japan and the Japan Islands in winter. This proportion seems to be large as compared to the proportion of the coastal area of Japan along the Sea of Japan to the vast area of the Sea of Japan or the Pacific. When a dry and cold air mass is transported from the continent across the Sea of Japan, a great deal of moisture is supplied from the relatively warm Sea of Japan and the continental aerosols seem to be effectively removed from the air by the snow over the coastal area of the Sea of Japan.

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Pb-210 В ОСАДКАХ В ЯПОНИИ И СЛЕДСТВИЯ ИЗ ЭТОГО ДЛЯ ПРОБЛЕМЫ ПЕРЕНОСА КОНТИНЕНТАЛЬНОГО АЭРОЗОЛЯ ЧЕРЕЗ ОКЕАН

Раз в месяц с февраля 1970 г. по март 1972 г. на пяти станциях на о. Хоккайдо, расположенных в глубине от берега Японского моря, производился сбор полных месячных осадков в виде дождя или снега и сухих выпадений. Определялась среднемесячная скорость выпадения Pb-210, а также других химических веществ. Результаты суммируются следующим образом. 1) Скорости выпадения и концентрации Pb-210 очень высоки зимой. 2) Было найдено, что летом и зимой существует линейная связь между скоростью выпадения Pb-210 и количеством дождевых осадков. 3) По сравнению со скоростью порождения Pb-210, именно, со скоростью эксхаляции Rn-222 с континента, скорость выпадения Pb-210 на единицу площади на берегу Японского моря зимой от двух до четырех раз превосходит скорость эксхаляции Rn-222 с континента. 4) Расчеты баланса содержания Pb-210 в атмосфере показывают, что около 40 % аэрозолей, переносимых с континента, захватывается снегом и откладывается над Японским морем и Японскими островами зимой. Поэтому делается вывод, что Японские острова зимой играют важную роль в качестве коллектора континентального аэрозоля.