Tellus (1996), 48B, 690–702
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TELLUS
ISSN 0280–6509

Seasonal variations of residence time and upper atmospheric contribution of aerosols studied with Pb-210, Bi-210, Po-210 and Be-7

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(Manuscript received 19 June 1995; in final form 3 April 1996)

ABSTRACT

All daily precipitation samples amounting to 196 in total were collected during the period from January 1990 to June 1991 at a station in Hakodate, Japan, and precisely analyzed for radon daughters, Pb-210, Bi-210 and Po-210. The samples collected after July 1990 were also determined for Be-7. The mean concentration and deposition rate of Pb-210 in 1990 were 0.20 Bq/l as a mean and 288 $Bq/m^2/yr$, respectively, and they were a few times greater in winter due to the northwest winter monsoon. The total mean activity ratios with 95% confidence limits were 0.467 ± 0.024 and 0.0624 ± 0.0133 for Bi-210/Pb-210 and Po-210/Pb-210, respectively. The residence time of aerosols calculated from the Po-210/Pb-210 ratio was always longer than that calculated from the Bi-210/Pb-210 ratio. The difference is chiefly due to old aerosols derived from the upper atmosphere (probably the stratosphere), of which contribution for Pb-210 has been estimated to be 6% as a mean, or 12% in spring and 2% in early winter. Subtracting the upper atmospheric component, we have obtained the residence time of tropospheric aerosols carrying Pb-210 to be 4.8 ± 0.3 days for a whole year or 5.9 ± 0.7 days and 3.6 ± 0.5 days for the periods from February to April and from May to June, respectively. The deposition rate of Be-7 was correlated well with the upper atmospheric component of Pb-210. The larger deposition rate of Be-7 suggests that the study area is extremely effective for the removal of aerosols of stratospheric origin.

1. Introduction

The Japanese Islands are well located at the east end of the Asian Continent for the study of removal of continental aerosol especially transported by the northwest monsoon in winter. Turekian et al. (1977) and Tsunogai et al. (1985) have reported the concentration of Pb-210 in surface air as well as its deposition flux decreasing exponentially toward the east from the Asian coast in the Pacific region. The Japanese Islands are covered by the continental Siberian air mass in winter and by the marine Ogasawara air mass in summer. Therefore, we expect there may be some seasonal differences in the concentration, flux and residence time of atmospheric components due to difference in their source material. In this study, we observed radon daughters, Pb-210 (half-life 22.3 years), Bi-210 (5.01 days) and Po-210 (138 days), which have been used as tracers of contin-

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ental aerosols, collecting meteoric precipitation samples every day for one and a half years at a station in Hakodate, Hokkaido in the northern part of Japan.

Many investigators have tried to estimate the mean atmospheric residence time of aerosols using the radon daughters (Burton and Stewart, 1960; Fry and Menon, 1962; Peirson et al., 1966; Anand and Rangarajan, 1990) and found the residence time obtained from the Po-210/Pb-210 ratio is longer than that from the Bi-210/Pb-210 ratio. The longer residence time obtained from the Po-210/Pb-210 ratio has been attributed to the addition of soil particles (Poet et al., 1972; Moore et al., 1973) or of upper atmospheric aerosols (Gavini et al., 1974; Tsunogai and Fukuda, 1974) or of volcanic dust (Lambert et al., 1979) to the tropospheric aerosols. These previous studies, however, are insufficient to pinpoint to one particular reason, due to data scanty in number and due to large analytical errors for Bi-210. In this study, we developed a precise determination method for Pb-210, Bi-210 and Po-210 (Tokieda et al., 1994) and applied it to the samples obtained in this study. Furthermore, we measured a cosmogenic nuclide, Be-7 which gave us information on aerosols from the upper atmosphere.

2. Sampling and analytical method

Precipitation samples were collected with a box type collector, which was 60 cm high and 90×90 cm^2 in open area, and set on the flat roof (10 m above the ground) of a building at the Hakodate campus of Hokkaido University (41°48'N, 140°44'E), in Hakodate, Hokkaido (Fig. 1). In order to avoid the admixture of resuspended snow and soil particles from the nearby earth's surface, the collector was set on a rack of 1 m high. A polyethylene bag was installed inside the collector, which was replaced once a day at usually 9 o'clock. When the precipitation sample was found in the bag, the water was sampled and analyzed even for a small volume of samples. All the precipitation samples amounting to 196 were collected during 535 days from 12 January in 1990 to 30 June in 1991.

By using the analytical method for Pb-210, Bi-210 and Po-210 of Tokieda et al. (1994), these three nuclides were separated from each other within 10 hours after the sampling. The quick separation made precise the determination of Bi-210. The mean relative counting errors (1 σ values) were 2.1% for Pb-210, 4.8% for Bi-210 and 6.0% for Po-210. Since July 1990, the concentration of Be-7 was simultaneously determined for the same samples. The analytical procedure for these four radionuclides especially Be-7 is briefly described below.

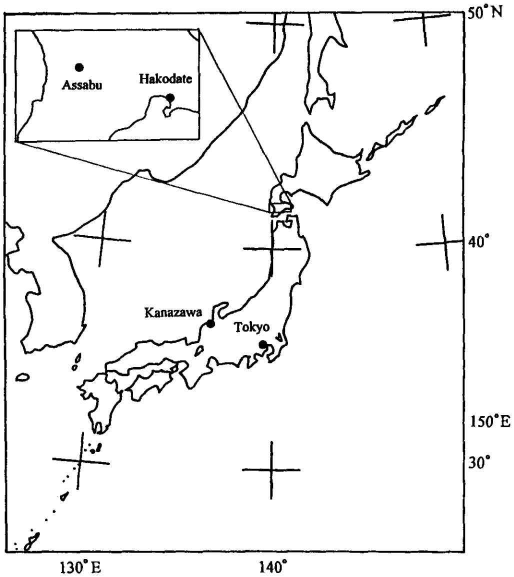


Fig. 1. Map showing the sampling station, Hakodate, and the reference stations, Assabu (Fukuda and Tsunogai 1975) and Kanazawa and Tokyo (Tsunogai et al. 1985).

Stable Pb, Bi and Be and Po-208 were added to the samples collected as carriers and yield tracers for Pb-210, Bi-210, Be-7 and Po-210. The Po, Bi and Pb isotopes were determined by electrodepositing in turn from the same sample solution as described in Tokieda et al. (1994). After oxidizing the metals with HNO_3 and H_2O_2, the remaining solution was changed to a medium of 8 N HCl and passed through a cation exchange column (Bio-Rad AG 1X8) to remove Fe^{3+}. The eluate was changed to a 0.5 N oxalic acid solution, and passed through an anion exchange column (Dowex 50W-X12) to collect Be isotopes. The volume of solution containing Be isotopes was reduced to about 2 ml by evaporation on a hot plate and the solution was transferred to a test tube of 5 ml. Its gamma activity from Be-7 was counted with a well type NaI (Tl) scintillation detector connected to a single channel pulse height analyzer (Aloka Model TCD-601). After counting the gamma activity, the solution was diluted by one millionth and the chemical yield of Be-7 was determined by measuring its Be concentration using an atomic absorption spectrophotometer (Hitachi Z-8000). The scintillation detector had 15.7% of counting efficiency and 35 cpm of background. A mean chemical yield was 63% and a mean counting error was 24% for Be-7.

We checked the uncertainty due to the sampling by collecting 3 times duplicate samples with different two collectors. The concentrations and fluxes of Pb-210, Bi-210, Po-210 and Be-7 in the duplicate samples agreed within the differences expected from the counting statistics.

3. Results

3.1. *Variation of concentrations and fluxes of Pb-210 and Be-7*

The city, Hakodate, is located at the northern part of Japan, but at the southern most of Hokkaido Island. In winter the area is usually under the continental influence of Siberian air mass. According to the observation by Hakodate Marine Observatory, the season usually begins in November and ends in March, bringing snow. In spring, it is generally calm due to moderately high pressures coming fairly periodically from the Asian Continent. The weather continues until July even during the rainy season "Tsuyu" in the mainland of Japan, due to the influence of the Okhotsk high pressure extending to the northern Japan. After ending the short summer under the Ogasawara air mass, autumn comes, of which weather is somewhat similar to spring.

Fig. 2 shows the daily concentration of Pb-210, and the Bi-210/Pb-210 and Po-210/Pb-210 ratios. Their monthly mean values are listed in Table 1 together with the monthly deposition rate (flux) of Pb-210. Although we recognize some interannual differences in the monthly values between 1990 and 1991, the monthly mean values computed from all the data are used in the following discussion. Reflecting the above weather cycle, the concentration and flux of Pb-210 vary seasonally. According to the seasonal variation, we can summarize the results by dividing a year into the following 3 seasons:

(1) A season with low concentration and low flux (April–June);

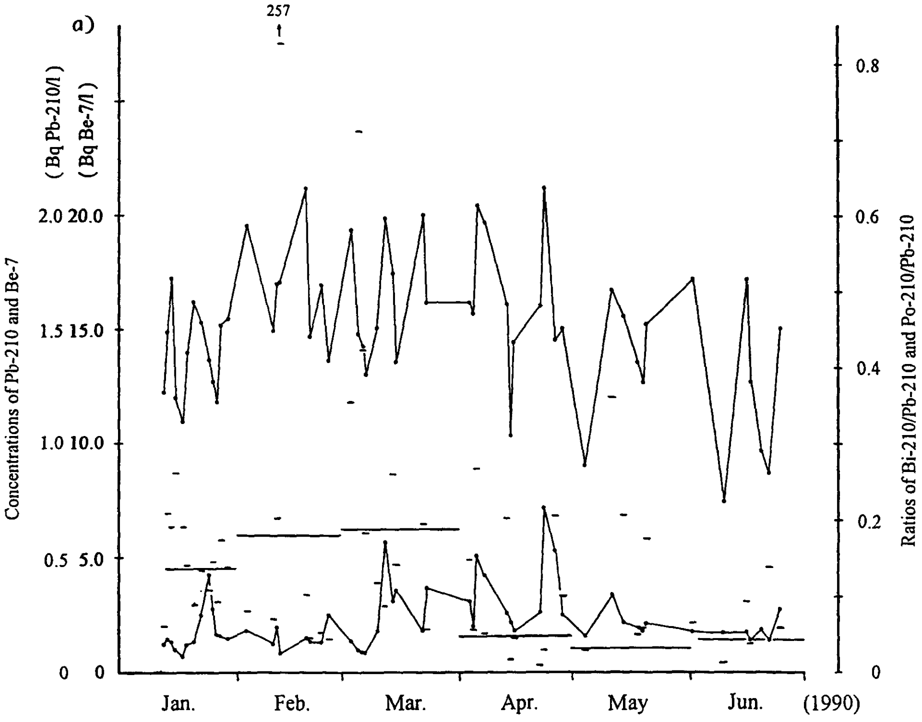
(2) A season with low concentration and high flux (July–October);

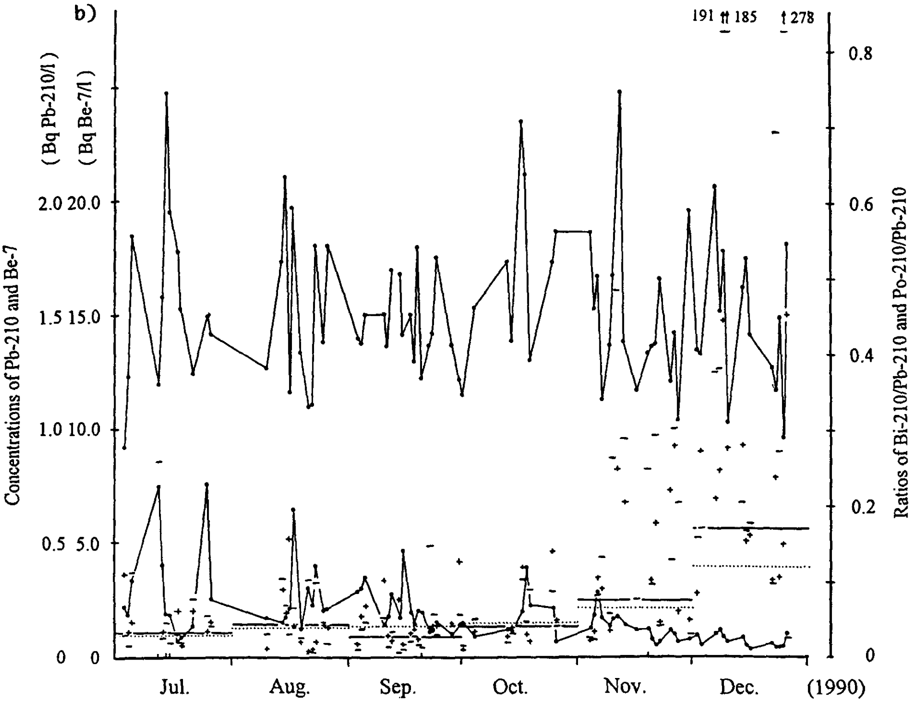
(3) A season with high concentration and high flux (November–March).

The seasonally mean Pb-210 concentration and its flux are listed together with the rainfall record in Table 2. The seasonal variation are induced by the change in air masses over Hokkaido that we have described above. In winter, the concentration of Pb-210 was high and its deposition rate was also higher as well as the precipitation rate. The high concentration was amplified by the cold and convective Siberian air mass of continental origin of which absolute humidity is low relative to the air mass of marine origin. In summer and autumn the low concentration was caused by the marine warm air mass containing relatively low concentration of Pb-210 and the high flux was due to the high precipitation rate, while the fairly low precipitation rate causing the low concentration and low flux of Pb-210 in spring.

In Fig. 3 the monthly fluxes of Pb-210 are compared with those obtained at Assabu, Hokkaido (Fukuda and Tsunogai, 1975), Tokyo and Kanazawa (Tsunogai et al., 1985). The monthly variation of flux of Pb-210 at Tokyo in the Pacific coast shows a summer peak, while that at Kanazawa in the Japan Sea coast of central Japan shows only a winter peak. The monthly variation at Hakodate gives two maxima, one in summer to autumn and the other in winter, as well as that at Assabu, although the winter peak is higher at Assabu nearer the Japan Sea coast. This feature may indicate that Hakodate is located at the transition zone between the Pacific and Japan Sea sides.

The snow sample collected on 13 February 1990 contained 4.23 Bq/l of Pb-210, which was one of the highest concentrations of Pb-210 ever reported. This extremely high concentration may not be explained only by the effective washout processes, because aerosols are always almost completely removed from the vertically developed convective air mass. According to the weather maps, a low pressure was found in the Japan Sea on 11 February 1990 and moved to the Okhotsk


a)
257
Concentrations of Pb-210 and Be-7
(Bq Pb-210/l)
(Bq Be-7/l)
0 0
0.5 5.0
1.0 10.0
1.5 15.0
2.0 20.0
Ratios of Bi-210/Pb-210 and Po-210/Pb-210
0
0.2
0.4
0.6
0.8
Jan.
Feb.
Mar.
Apr.
May
Jun.
(1990)


b)
191
185
278
Concentrations of Pb-210 and Be-7
(Bq Pb-210/l)
(Bq Be-7/l)
0 0
0.5 5.0
1.0 10.0
1.5 15.0
2.0 20.0
Ratios of Bi-210/Pb-210 and Po-210/Pb-210
0
0.2
0.4
0.6
0.8
Jul.
Aug.
Sep.
Oct.
Nov.
Dec.
(1990)

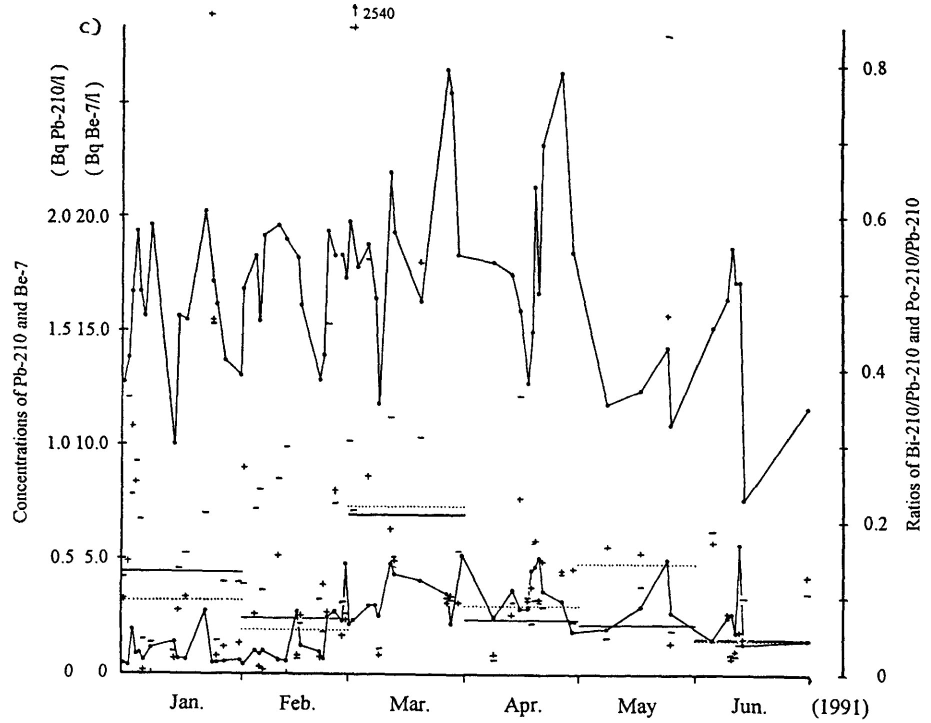


Fig. 2. Daily variations of concentrations of Pb-210 and Be-7 and ratios of Bi-210/Pb-210 and Po-210/Pb-210 during January 1990 and June 1990 (a), July 1990 and December 1990 (b) and January 1991 and June 1991 (c). The ratios of Bi-210/Pb-210 and Po-210/Pb-210 are shown with solid circles (●) and open circles (○), respectively. Concentrations of Pb-210 and Be-7 are shown with minus (−) and plus (+) signs, respectively. Solid and dashed horizontal lines refer to monthly mean concentrations of Pb-210 and Be-7, respectively.

Sea on 12 February becoming an extremely strong one there. Tsunogai and Shinagawa (1977) have also reported a high Pb-210 concentration (4.18 Bq/l) at Shishiku, Ishikawa in February 1966, which was derived together with much "Kosa", the Chinese desert dust sometimes observed in spring in Japan, and sulfate from the Asian continent under the similar weather condition. The high Pb-210 concentration observed would be amplified by the cold air mass of low absolute humidity which relatively concentrated Pb-210. During the winter monsoon season, the strong northerly or northwesterly wind brings much amount of continental aerosols from the Asian continent to Japan. This induces the high deposition rate of Pb-210 in winter in the region facing the Japan Sea, while the Pacific coast is extremely dry and has little rainfall in winter.

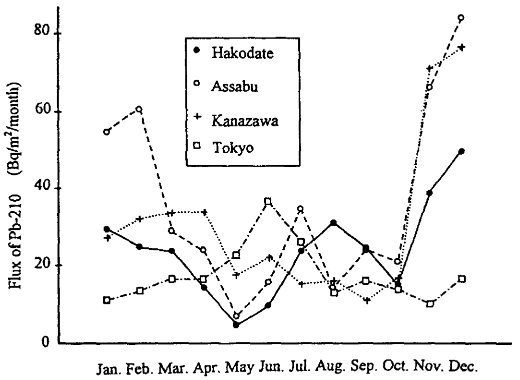


Fig. 3. Monthly deposition rates of Pb-210 obtained at various stations in Japan, Hakodate (this study), Assabu (Fukuda and Tsunogai 1975), Kanazawa (Tsunogai et al. 1985) and Tokyo (Tsunogai et al. 1985).

Table 1. *Monthly mean concentrations and deposition rates of Pb-210 and Be-7 and ratios of Bi-210/Pb-210 and Po-210/Pb-210*

Month	Rainfall (mm/month)	Number of samples (Samples/days)	Pb-210		Be-7		Bi-210/Pb-210**	Po-210/Pb-210**
			(Bq/l)*	(Bq/m^2/month)	(Bq/l)*	(Bq/m^2/month)		
(1990)								
Jan.	70	13/20	0.47	32.4	—	—	0.417±0.057	0.0515±0.0287
Feb.	48	8/28	0.60	28.8	—	—	0.513±0.068	0.0477±0.0159
Mar.	32	10/31	0.70	22.4	—	—	0.492±0.082	0.0715±0.0478
Apr.	99	11/30	0.16	15.5	—	—	0.492±0.093	0.1047±0.0508
May	27	6/31	0.24	6.7	—	—	0.412±0.082	0.0652±0.0190
Jun.	88	7/30	0.16	13.7	—	—	0.377±0.122	0.0540±0.0132
Jul.	232	12/31	0.10	23.8	1.2	278	0.470±0.124	0.0881±0.0686
Aug.	220	11/31	0.14	31.2	1.3	280	0.460±0.110	0.0710±0.0487
Sep.	281	17/30	0.09	24.5	1.3	377	0.439±0.052	0.0615±0.0297
Oct.	117	9/31	0.13	15.4	1.5	171	0.506±0.117	0.0512±0.0300
Nov.	158	17/30	0.25	39.3	2.2	347	0.452±0.107	0.0375±0.0174
Dec.	88	14/31	0.57	50.5	8.7	761	0.440±0.094	0.0207±0.0078
(1991)								
Jan.	60	15/31	0.34	20.5	3.2	193	0.472±0.084	0.0276±0.0198
Feb.	84	14/28	0.24	20.5	1.9	160	0.522±0.061	0.0480±0.0378
Mar.	34	11/31	0.70	23.6	7.4	250	0.580±0.125	0.1016±0.0313
Apr.	59	10/30	0.24	14.3	3.0	176	0.556±0.122	0.1036±0.0293
May	14	4/31	0.21	2.9	4.7	65	0.371±0.043	0.0937±0.0392
Jun.	38	7/30	0.15	6.0	1.5	58	0.449±0.109	0.0721±0.0457

* The mean concentrations are obtained by dividing the total deposition by the total rainfall.

** The standard deviations of daily values are attached to the mean values.

Table 2. *Seasonally averaged concentrations and deposition rates of Pb-210 and Be-7 and Bi-210/Pb-210 and Po-210/Pb-210*

Season	Rainfall (mm/month)	Number of samples (samples/days)	Pb-210		Be-7		Bi-210/Pb-210**	Po-210/Pb-210**
			(Bq/l)*	(Bq/m^2/month)	(Bq/l)*	(Bq/m^2/month)		
Spring (Apr.-Jun.)	54.3	45/182	0.18	9.9	0.92	49.8	0.406 ± 0.119	0.0852 ± 0.041
Summer-Autumn (Jul.-Oct.)	212.2	49/123	0.11	23.7	1.30	276.5	0.463 ± 0.099	0.0683 ± 0.047
Winter (Nov.-Mar.)	72.5	102/230	0.41	29.8	2.95	213.9	0.481 ± 0.098	0.0480 ± 0.036
Total*** (annual)	115.3	141/365	0.20	22.7	2.25	260.0	0.479 ± 0.106	0.0595 ± 0.044

* The mean concentrations are obtained by dividing the total deposition by the total rainfall.

** The standard deviations of daily values are attached to the mean values.

*** Annual values are calculated using the data during July 1990 and June 1991.

Israël (1951) and Wilkening and Clements (1975) estimated Rn-222 flux from the earth's surface to the atmosphere to be 0.70–0.75 atom/cm^2/s, being equivalent to 220–230 Bq/m^2/yr of the Pb-210 production rate in the continental atmosphere. Although our deposition rate of Pb-210 does not include the dry deposition during 230 days of no rain in 1990, which seems to be less than 20% of the wet deposition (Tsunogai, 1975; Matsunami et al., 1979; Fuller and Hammond, 1983), the annual Pb-210 flux in 1990 was 306 Bq/m^2/yr. Fukuda and Tsunogai (1975) have obtained similar values of 250–470 Bq/m^2/yr in this district. The Japanese Islands, therefore, are an effective sink of Pb-210 or of aerosols of continental origin, in particular in winter.

In Table 1, the monthly mean fluxes and concentrations of Be-7 in precipitation at the Hakodate station are also shown. The seasonal variation of concentration of Be-7 was similar to that of Pb-210 having a different source function, indicating that their concentrations were greatly affected by the precipitation rate, namely the dilution with water condensed. The higher concentration of Be-7 was observed during winter and spring. The one in winter may be caused by low water vapor pressure in cold air and the other in spring agrees with the spring peak of material of stratospheric origin such as ozone. However, the monthly deposition flux of Be-7 was larger even in summer as well as winter (Table 2), which would be due to larger precipitation rate in summer.

3.2. Atmospheric residence times of aerosol from the ratios of Bi-210/Pb-210 and Po-210/Pb-210

In order to calculate the atmospheric residence time of aerosol bearing Pb-210, Sporadic Removal Model (SRM) proposed by Tsunogai and Fukuda (1974) was used because it seemed to be more appropriate for the status of removal of continental aerosols in winter monsoon in the Japan Islands than Continuous Removal Model (CRM) used by Burton and Stewart (1960) postulating a steady state removal of aerosols from the air column. In the SRM it is assumed that aerosols in the air column are accumulated with a constant rate before being removed completely by the wet deposition. The Pb-210 produced from Rn-222 in the continental air mass across the Japan Sea is effectively removed in the convective cloud forming snow over the Japanese Islands.

According to Tsunogai and Fukuda (1974), the residence times (in days) calculated from the activity ratios of Bi-210/Pb-210 (T_E) and the Po-210/Pb-210 (T_F) are given by

$$\frac{\text{Bi-210}}{\text{Pb-210}}=\frac{1}{1-\exp(-2T_E/t_1)}\times\left\{1-\frac{\exp(-2T_E/t_1)}{1-t_2/t_1}-\frac{\exp(-2T_E/t_2)}{1-t_1/t_2}\right\}, \tag{1}$$

$$\frac{\text{Po-210}}{\text{Pb-210}}=\frac{1}{1-\exp(-2T_F/t_1)}\times\left\{1-\frac{\exp(-2T_F/t_1)}{(1-t_2/t_1)(1-t_3/t_1)}-\frac{\exp(-2T_F/t_2)}{(1-t_1/t_2)(1-t_3/t_2)}-\frac{\exp(-2T_F/t_3)}{(1-t_1/t_3)(1-t_2/t_3)}\right\}, \tag{2}$$

where t_1, t_2 and t_3 are the half-life of Pb-210, Bi-210 and Po-210 of 22.3×365, 5.01 and 138 days. The differences in the calculated residence times between the two models, SRM and CRM, are usually small and their means are 12% for T_E and 21% for T_F.

The apparent monthly mean residence times of T_E and T_F (Fig. 4) range from 3.7 to 8.5 days and from 9 to 29 days, respectively. A total mean values of 196 samples with 95% confidence limits were 5.6 ± 0.3 days and 19 ± 1 days, although the standard deviations of all the values were 2.4 and 10 days, respectively, for T_E and T_F. The mean residence times were similar to those previously reported ones (Fry and Menon, 1962; Moore et al., 1973; Gavini et al., 1974). The T_F values were always longer than the T_E values without exception.

None appreciable seasonal variation in the residence time was found for T_E, where the residence times were 5.5 ± 2.7, 5.4 ± 2.2 and 5.7 ± 2.3 days in spring, summer and autumn, and winter, respectively. The seasonal mean values were 25 ± 3, 20 ± 3 and 15 ± 2 days in spring, summer to autumn, and winter, respectively, for those from the ratio of Po-210/Pb-210. The seasonal variation was more pronounced and showed a difference from that derived from the ratio of Bi-210/Pb-210.

4. Discussion

4.1. Difference in the residence time of aerosols between the Bi-210/Pb-210 and Po-210/Pb-210 methods

The fact that the atmospheric residence time of aerosols obtained from the Po-210/Pb-210 ratio (T_F) is longer than that from Bi-210/Pb-210 ratio (T_E) has already been noted by some authors (Poet et al., 1972; Marenco and Fontan, 1972; Moore et al., 1973; Gavini et al., 1974; Tsunogai and Fukuda, 1974). This difference can be explained by the addition of some "old" Pb-210 having a high Po-210/Pb-210 ratio to the lower tropospheric aerosols, because it makes T_F long more than T_E. As a candidate of the material containing "old" Pb-210 and extra Po-210, Poet et al. (1972) considered soil dust from the earth's surface and Tsunogai and Fukuda (1974) suggested the upper atmospheric aerosols. Lambert et al. (1979) stated the contribution of Po derived from volcanoes, because they observed volcanic gas having 10–100 of the Po-210/Pb-210 ratio. Hussain et al. (1991) reported Po-210 emanated from the sea by biological activity. A possibility of Po-210 derived from volcanic gas would be ignored, because a large volcanic eruption was not recorded in the world and also in Japan in 1990 when we started the observation. A possibility of Po-210 emanated from the sea would not be significant, because the concentration of Pb-210 is only a few tenths Bq/100L in seawater and Po-210 is quickly removed by forming sinking particles in Funka Bay near Hakodate (Tanaka et al., 1983).

Even though the difference between T_E and T_F was not small in winter when the earth's surface was mostly covered by snow, Pb-210 from soil particles blown up from the nearby earth's surface may somewhat contribute to the difference. If Pb-210 concentration in soil dust is 0.67 Bq/g or 0.27 Bq/g, which is a maximum or a mean concentration of Pb-210 in the surface soil around Hakodate (Table 3), the contribution of Pb-210 from soil dust turns out to be 1.4 Bq/m^2/yr or 0.57 Bq/m^2/yr, corresponding to 0.5% or 0.2% of total fallout, since the residue in precipitation

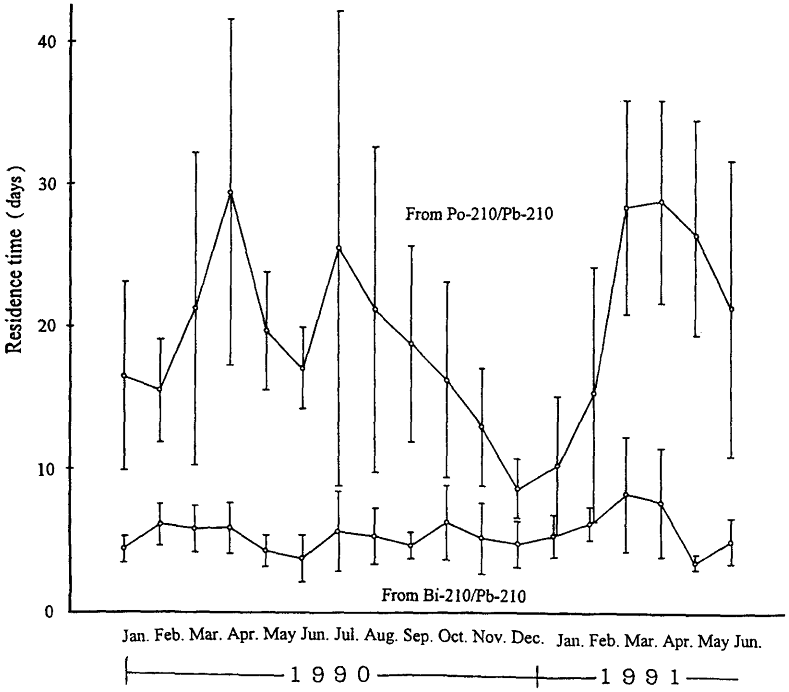


Fig. 4. Monthly mean residence times of aerosols calculated from the Bi-210/Pb-210 and Po-210/Pb-210 ratios with the Sporadic Removal Model (SRM) of Tsunogai and Fukuda (1974). The standard deviations of daily values are attached to the mean values.

Table 3. *Concentration of Pb-210 in surface soils*

Location		Pb-210 (Bq/g)	Refs.
(Region near Hakodate)			
Hakodate Campus,	a)	0.140±0.007	(1)
Hokkaido Univ.	b)	0.192±0.007	(1)
	c)	0.313±0.007	(1)
Mt. Hakodate	a)	0.310±0.007	(1)
	b)	0.345±0.007	(1)
Hakodate Park		0.605±0.012	(2)
Ichinowatari, Ohno	a)	0.085±0.003	(1)
	b)	0.178±0.005	(1)
	c)	0.190±0.005	(1)
(Others)			
Sanpoji Temp, Shiga pref.		0.151±0.007	(2)
Fuchu, Tokyo		0.16±0.007	(2)
South France		0.01	(3)
Moscow		0.11~0.30	(4)

(1) This study.
(2) Kurata and Tsunogai (1986).
(3) Marenco and Fontan (1972).
(4) Vilenskiy (1979).

samples was 2.15 g/m^2/yr. This value is comparable to that estimated by Joshi (1968), 0.5% using U-238, and indicates that its contribution to airborne Pb-210 is not significant. The contribution to airborne Po-210, however, is not negligibly small, because the mean Po-210/Pb-210 was 0.0590, and we correct the effect in the following calculation.

4.2. Estimate of upper atmospheric component

The contribution of the upper atmospheric Pb-210 is calculated if we can assume the followings.

(1) Radon daughters in the precipitation samples come from the following three sources; lower and upper atmospheric aerosols, and soil particles from the nearby earth's surface. The distinction between the two layers is somewhat arbitrary, but we regard them as the troposphere and the stratosphere, respectively, although

Pb-210 of stratospheric origin is transported for a long distance after entering the troposphere.

(2) Pb-210, Bi-210 and Po-210 in soil particles are under a radioactive equilibrium and the soil particles contain 0.27 Bq/g of Pb-210. The uncertainty derived from this assumption is not larger than 10% as described above.

(3) The residence time of the upper atmospheric aerosols component of Pb-210 is 1 year (Katsuragi, 1965; Reiter, 1975). The uncertainty derived from variation in its residence time is also not greater than 10%, because both the ratios of Bi-210/Pb-210 and Po-210/Pb-210 are not highly variable for aerosol particles older than 1 year.

The proportion of the upper atmospheric contribution to the total Pb-210 flux calculated was larger in April and July 1990 or in May 1991 (Fig. 5). The spring maximum observed for the upper atmospheric contribution of Pb-210 coincides with the spring peak of the transport of upper atmospheric ozone to the surface air (Hayakari et al. 1989). However, the peak in July is not concordant with the seasonal variation of tropospheric ozone of stratospheric origin. Gavini et al. (1976) reported a high Pb-212/Pb-210 ratio during an intense storm, and attributed this to the active turbulent mixing within the thunderstorm. Since some of our samples in July were derived from thunderstorms, a peak of the upper atmospheric contribution of Pb-210 observed in July may be originated in the active vertical convection reaching the lower stratosphere in summertime.

Tsunogai (1971) has applied the Rn-222 and Pb-210 system to the SO_2 and sulfate system and estimated the lifetime of SO_2 in the atmosphere, which is 1.4 days or 1/4 of the lifetime of Rn-222 of 5.5 days. Here we have obtained the contribution from the upper atmosphere of 6% for Pb-210. Although the quantitative estimate is not easy, we can expect that the upper atmospheric contribution is smaller than 6% for sulfate emitted as SO_2. On the other hand, if the lifetime of its gaseous progenitor such as DMS (dimethyl sulfide) from the marine environment would be longer than 5.5 days, the contribution could be larger than 6%. Future simultaneous observation of atmospheric radon daughters and sulfur species will provide valuable information on the behavior of sulfur compounds in the atmosphere.

4.3. Correlation between Be-7 and upper atmospheric Pb-210

The Pb-210 flux derived from the upper atmosphere and its ratio to the total Pb-210 flux have been estimated to be 17.8 Bq/m^2/yr. and 5.7%, respectively, and they are considerably larger than those derived from soil particles of the earth's

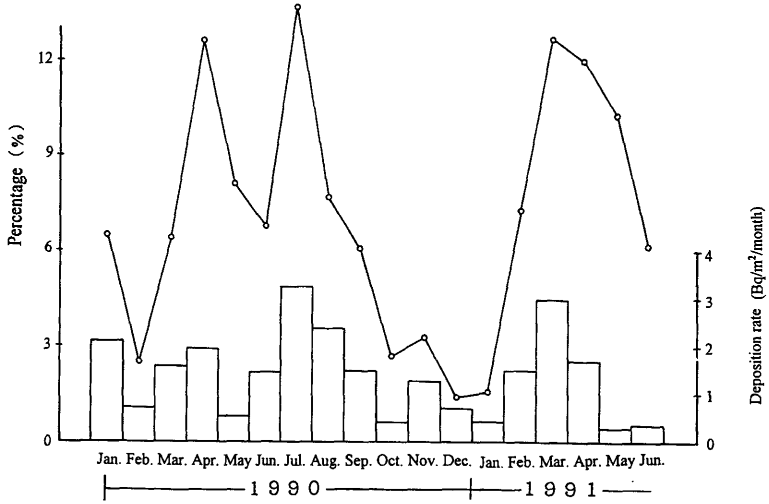


Fig. 5. Monthly mean contribution of the upper atmospheric Pb-210 in percentage to the total deposition (line graph) and monthly deposition rate of Pb-210 (bar graph).

surface (0.2%). Consequently, only a half of Po-210 deposited on the earth's surface is produced in the troposphere. The monthly Be-7 fluxes are plotted against those of the upper atmospheric Pb-210 in Fig. 6. They seems to be classified into two groups, one is those observed in February to August and the other is those in September to January. There are good correlations for both groups. The mean activity ratios of the Be-7 flux to the upper atmospheric Pb-210 were 0.0030 and 0.010 during the periods from September to January and from February to August, respectively, with an annual mean of 0.0061. We do not know the reason why the two different regression lines exist, but it may be related to change in the air masses. Leifer et al. (1984) has reported the stratospheric concentrations of Pb-210 and Be-7 are 0.042 and 6.7 Bq/100 SCM, respectively, at altitude of 13.7 km between 40°N and 45°N. This gives the Pb-210/Be-7 ratio of 0.0064, which is just the same as the mean value in this study. The coincidence also supports our conclusion that the difference between T_E and T_F is due to the added upper atmospheric aerosols.

According to Dutkiewicz and Husain (1985), about 25% of Be-7 deposited on the earth's surface has come from the stratosphere, which corresponds to the deposition rate of 680 Bq/m^2/yr obtained in this study. When the mean flux from the stratospheric can be obtained, if the concentration of stratospheric Be-7 is 6.7 Bq/100SCM and its residence time is 1 year as given above, which is 160 Bq/m^2/yr or the flux observed at Hakodate. Thus, the study area is extremely effective for the removal of aerosols of stratospheric origin. This seems to be due to the location conditions of study area, Hakodate, of much rain and in the mid-latitudes under the gap of the tropopause.

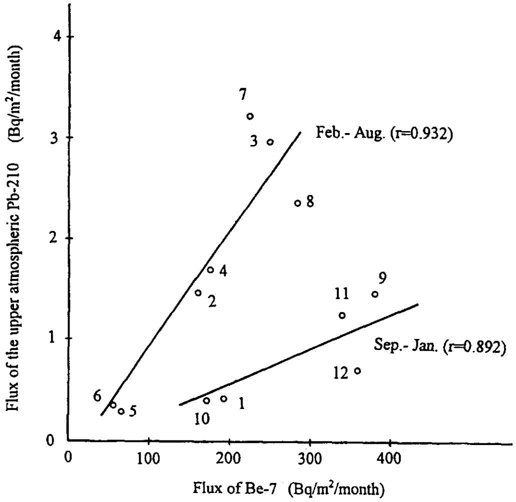


Fig. 6. Relationship between the upper atmospheric Pb-210 and Be-7 for the monthly mean deposition rates. The two regression lines are calculated separately for two seasons, February to August and from September to January.

4.4. Seasonal variation in residence time of tropospheric aerosols

Fig. 7 illustrates the monthly mean residence times of tropospheric aerosols, which were obtained by subtracting the contribution from surface soil and the upper atmospheric aerosols from the total fallout for radon daughters. They are not much different from the apparent residence times derived from the Bi-210/Pb-210 ratios (T_E), since both Bi-210 and Pb-210 are supplied mainly from the lower atmosphere. A weak seasonal variation is shown in Fig. 7, where longer values (5.9 ± 0.7 days) and shorter values (3.6 ± 0.5 days) are found during the periods from February to April and from May to June, respectively. The shorter ones in May and June seem to be due to marine air masses. In the marine air mass, old aerosols may be removed by fairly frequent precipitation events on the way to Hakodate or Japanese Islands. The total mean residence time with a 95% confidence limit was 4.8 ± 0.3 days. This value is slightly shorter than the atmospheric residence time of water vapor, indicating that aerosol carrying Pb-210 in the air column is effectively removed by a precipitation event.

5. Conclusions

We have obtained the following results from Pb-210, Bi-210, Po-210 and Be-7 in the daily precipitation samples at a station in Hakodate, Japan.

(1) The mean residence time of aerosols calculated from the Bi-210/Pb-210 ratio was 5.6 ± 0.3 days with no appreciable seasonal variation.

(2) That calculated from the Po-210/Pb-210 ratio was 19 ± 1 days and seasonal mean values

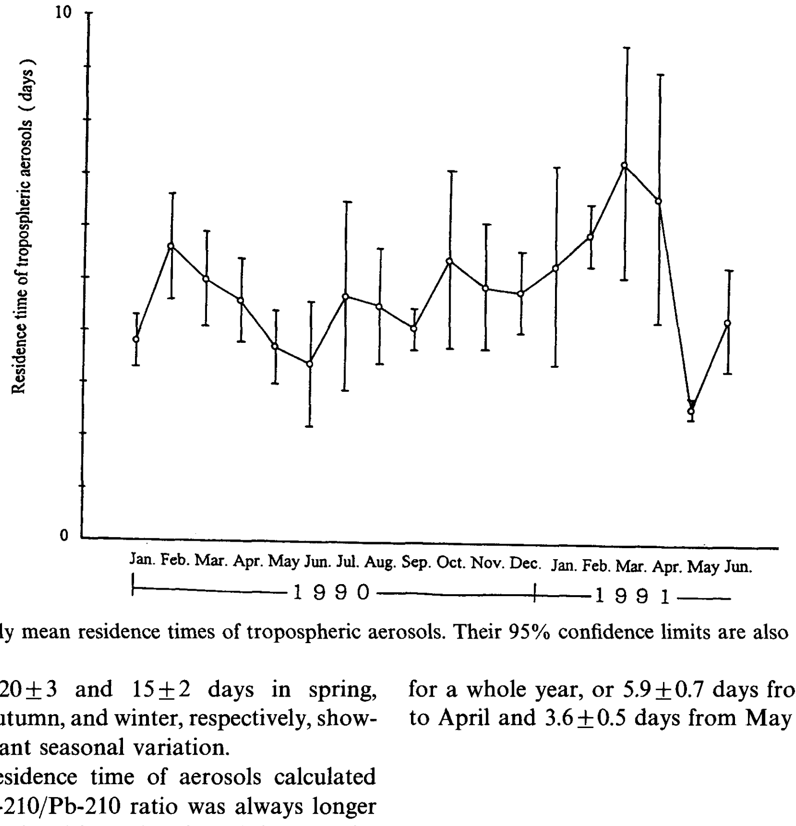


Fig. 7. Monthly mean residence times of tropospheric aerosols. Their 95% confidence limits are also shown.

were 25±3,20±3 and 15±2 days in spring, summer to autumn, and winter, respectively, showing a significant seasonal variation.

(3) The residence time of aerosols calculated from the Po-210/Pb-210 ratio was always longer than that calculated from the Bi-210/Pb-210 ratio. The difference is considered to be due to old aerosols derived from the upper atmosphere.

(4) The contribution of the upper atmospheric aerosols for Pb-210 was estimated to be 6% as a mean, or 12% in spring and 2% in early winter. The deposition rate of the upper atmospheric Pb-210 was well correlated with that of Be-7.

(5) The residence time of tropospheric aerosols carrying Pb-210 was estimated to be 4.8±0.3 days for a whole year, or 5.9±0.7 days from February to April and 3.6±0.5 days from May to June.

6. Acknowledgments

The authors wish to thank Prof. S. Noriki, Drs. N. Tanaka, S. Watanabe, H. Narita, K. Fukuda, T. Kurata and K. Yokota for the valuable discussion and their suggestion. The authors also thank Messrs. Y. Arashitani, T. Onodera, M. Kawasaki, S. Nishino, S. Hashimoto, N. Higashitani, S. Murata and Mrs Y. Kawamura for their help at the sampling and analysis.

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