Pb-210, Bi-210 and Po-210 in meteoric precipitation and the residence time of tropospheric aerosol

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Abstract—The concentrations of ^{210}Pb , ^{210}Bi and ^{210}Po in rain and snow were determined. Extremely high concentrations of ^{210}Pb , e.g. 190dpm/l, were observed for snow produced from convective clouds by the north-west monsoon on 3 Feb. 1972, at Assabu, Hokkaido. The high concentration is due to the active convection in the continental air mass which contains much radon and has rather low absolute humidity. The $^{210}\text{Bi}/^{210}\text{Pb}$ ratio was higher in rain in summer than in snow in winter. By using the two models, CRM (continuous removal model) and SRM (sudden removal model), the residence times of aerosols in the atmosphere are calculated from the activity ratios, $^{210}\text{Bi}/^{210}\text{Pb}$ and $^{210}\text{Po}/^{210}\text{Pb}$. The residence time from the $^{210}\text{Bi}/^{210}\text{Pb}$ ratio is apt to be shorter for the snow by the winter monsoon. This is due to the addition of aged aerosols which may come from the lower stratosphere to the troposphere. By resolving the simultaneous equations, the percentage of aged aerosols and the true residence time of tropospheric aerosols are estimated to be 1 to 5% and 2 to 7 days for the snow produced by the winter monsoon. The residence time, however, varies widely owing to the origin and history of the air mass.

INTRODUCTION

The residence time or lifetime of an aerosol is a good measure of its stability in the atmosphere. Radioactive nuclides are a useful means for the determination of the residence time, because their quantities decrease with time and most of these nuclides adhere to neighboring aerosol immediately after their production. As reviewed by JUNGE (1963), these nuclides are classified into three groups according to their origins, which are cosmic-ray produced nuclides such as ^7Be and ^{32}P , fission products, and natural radon- and thoron-decay products. However, the residence time of tropospheric aerosols obtained from the nuclides of the former two origins, 30 ~ 40 days, is longer than that from radon daughters. This is mainly due to the difference in vertical distributions of aerosols containing these nuclides.

The vertical distribution of a radon daughter, ^{210}Pb , in the atmosphere (e.g. MOORE *et al.*, 1973) is similar to that of whole aerosols (e.g. JUNGE, 1963), because radon exhales from the earth's surface and decays within a comparatively short time.

As the residence time of tropospheric aerosols, therefore, the time estimated from radon daughters seems to be more appropriate than that estimated from cosmic ray produced nuclides or fission products. The half lives and chief radiation characteristics of radon and its decay products are shown in Table 1. Using the $^{210}\text{Po}/^{210}\text{Pb}$ ratio in rain or aerosols, LEHMANN and SITTKUS (1959), and BURTON and STEWART (1960) determined the residence time of tropospheric aerosols to be about 20 days. On the other hand a residence time less than 7 days was obtained from the $^{210}\text{Bi}/^{210}\text{Pb}$ ratio by FRY and MENON (1962), POET *et al.* (1972) and MOORE *et al.* (1973). The discrepancy in the residence time between the $^{210}\text{Po}/^{210}\text{Pb}$ and the $^{210}\text{Bi}/^{210}\text{Pb}$ methods may be due to the following three causes. 1) The contribution of stratospheric aerosols, in which $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios are expected to be near unity, while that of tropospheric aerosols is commonly less than 0.1; 2) The contribution of soil dust where the radioactive equilibrium is substantially reached among ^{210}Pb , ^{210}Bi and ^{210}Po ; 3) The contribution of nuclear fission debris which may contain ^{210}Po .

Table 1. *Half-lives and chief radiation characteristics of radon and its daughters*

	Half-life	Decay modes and energies (MeV)		
Rn-222	3.82 d	α	5.490	
Po-218	3.05 m	α	6.002	
Pb-214	26.8 m	β	0.65, 0.73	0.053 to 0.777
Bi-214	19.7 m	β	0.4 to 3.26	0.61 to 3.07
Po-214	164 μ s	α	7.687	
Pb-210	20.4 y	β	0.015, 0.061	
Bi-210	5.01 d	β	1.16	
Po-210	138 d	α	5.305	
Pb-206	Stable			

The degree of contribution of the last cause to ^{210}Po was proved by BEASLEY (1969) to be negligibly small. POET *et al.* (1972) and MOORE *et al.* (1973) determined ^{222}Rn , ^{210}Pb , ^{210}Bi , ^{210}Po and ^{90}Sr concentrations in rain and in air at various altitudes up to 16 km over the continental United States and they concluded that most of tropospheric ^{210}Po is of continental surface origin. MARENCO and FONTAN (1972), on the other hand, reported that the contribution of ^{210}Po and ^{210}Pb from continental surface dusts was negligibly small.

REX and GOLDBERG (1958) found high quartz contents of deep-sea sediments from mid-latitudes of the North Pacific and attributed the source of the quartz to air-borne dust from the continent. TSUNOGAI and NOZAKI (1971) and NOZAKI and TSUNOGAI (1973a,b) found nearly the same latitudinal variation of air-borne ^{210}Pb in the surface water of the vast area of the Pacific Ocean as that of quartz. These facts suggest that the residence time of the continental aerosol is long enough to transport air-borne ^{210}Pb evenly to the same latitudinal belt of the Pacific, that is to say, aerosols of stratospheric or upper tropospheric origin comprise substantial part of

^{210}Pb as well as ^{210}Po in the maritime precipitation.

In the present investigation, we studied the relation among these three nuclides in the snow in Hokkaido where the surface was covered by snow in winter and the snow prevented surface dusts from being blown up.

Sampling and analytical methods

Rain and snow samples were collected at Hakodate and Assabu (Fig.1). The sampler for rain and snow at Hakodate was made of polyethylene plates supported with a wooden frame, of which the dimension was 530×145 cm. Snow samples at Assabu were collected on a polyethylene sheet, which was spread on the ground, the rim of which being fixed on a frame of snow pile.

Fig.1. Sampling locations (X) situated in the southern part of Hokkaido Island. Contour lines indicate 0, 100, 500 and 1,000m above the sea level.

The scheme for the simultaneous determinations of ^{210}Pb , ^{210}Bi and ^{210}Po is shown in Fig.2. The detailed procedure will be reported elsewhere. The α activities of ^{210}Po were measured by a 2π gas flow type proportional counter (Aloka, JDC-311) and the β activities of ^{210}Bi by a 2π gas flow type GM counter with shielding anticoincidence counters (Aloka, LBC-20). The counting error was less than 10% of each radioactivity. The overall yield for lead or bismuth was 50 to 90%. The coprecipitation efficiency of polonium or lead with calcium and the chemical yield of polonium were assumed to be the same as those of bismuth. These assumptions were valid in the laboratory experiments where the tracers of ^{210}Pb , ^{210}Bi and ^{210}Po were added to water samples of volumes up to 30ℓ).

Fig.2. The schematic diagram for the determination of ^{210}Pb , ^{210}Bi and ^{210}Po in precipitation.

RESULTS AND DISCUSSION

Concentrations of ^{210}Pb and the ratios of $^{210}\text{Bi}/^{210}\text{Pb}$ and $^{210}\text{Po}/^{210}\text{Pb}$ in precipitation

The results of ^{210}Pb determinations in the precipitation at Hakodate are given in Table 2. The concentration of ^{210}Pb is higher in the snow in winter than in the rain in other seasons. Although the numbers of the observed data are rather small, the medians, 36dpm/l in the snow and 8dpm/l in the rain, are nearly equal to those calculated from the monthly deposition rates of ^{210}Pb at Hakodate and Assabu observed by FUKUDA and TSUNOGAI (1974). The seasonal variation is chiefly due to the origin of air masses supplying water vapor and aerosols, that is, the high concentration in the snow is caused by the continental air mass.

The activity ratios of ^{210}Bi and ^{210}Po to ^{210}Pb are also shown in Table 2. Although

Table 2. Concentrations of Pb-210, activity ratios of Bi-210 and Po-210 to Pb-210, and calculated residence times, T_{CE} , T_{CF} , T_{SE} and T_{SF} at Hakodate

Date				Precipitation	Pb-210	Bi-210** Pb-210	Po-210** Pb-210	T_{CE}	T_{CF}	T_{SE}	T_{SF}
				mm	dpm/l	$\times 10^2$	$\times 10^2$	day	day	day	day
<i>Rain</i>											
1970	May	27,	1	3.4	20	—	1.0 ± 0.1	—	5	—	5
			2	4.5	13.0	—	1.8 ± 0.1	—	7	—	8
	June	26		10.0	34	80 ± 2	1.5 ± 0.1	29	6	18	7
		27		7.0	16.5	72 ± 3	1.9 ± 0.1	19	8	13	9
	July	31		7.0	8.1	—	2.2 ± 0.2	—	8	—	9
	Aug.	4,	1	4.6	2.1	90 ± 10	7.6 ± 0.7	>29	22	>18	22
			2	3.1	18.8	81 ± 4	3.0 ± 0.2	30	11	19	11
			3	3.7	5.1	48 ± 6	3.4 ± 0.3	7	11	5	12
1971	Sept.	19,	1	6.6	5.4	114 ± 9	1.2 ± 0.2	>30	6	>30	6
			2	5.3	5.5	55 ± 3	1.2 ± 0.2	9	6	7	6
	Sept.	28,	1	4.4	7.9	113 ± 6	1.0 ± 0.2	>30	5	>30	5
			2	6.0	5.5	82 ± 4	0.9 ± 0.2	33	5	20	5
Median					8	81	1.5	30	6	19	7
<i>Snow</i>											
1971	Jan.	30		3.0	22	—	1.7 ± 0.1	—	7	7	7
	Feb.	1		4.5	26	—	1.4 ± 0.1	—	6	7	6
		2		1.5	39	63 ± 13	1.3 ± 0.1	12	6	9	6
		4*		0.9	36	41 ± 5	1.4 ± 0.2	5	6	4	6
	Feb.	9,	1	2.4	45	56 ± 2	1.7 ± 0.1	9	7	7	7
			2	5.4/2	11.0	60 ± 5	3.3 ± 0.3	11	11	8	12
			2'	5.4/2	11.1	68 ± 5	4.0 ± 0.3	15	13	11	13
	Feb.	15,	1	2.0	42	77 ± 2	1.6 ± 0.2	22	7	15	7
			2	1.1	22	99 ± 5	3.3 ± 0.3	>30	11	>30	12
			3	1.2	6.1	121 ± 9	6.7 ± 0.9	>30	20	>30	20
	Nov.	29		3.0	92	56 ± 3	1.0 ± 0.1	9	5	7	5
		20		7.5	41	56 ± 3	1.2 ± 0.1	9	6	7	6
	Dec.	3,	1	5.0	15.2	78 ± 5	5.6 ± 0.6	24	17	16	17
			2	2.1	56	71 ± 3	3.6 ± 0.3	17	12	12	12
			3	5.7	117	60 ± 1	1.4 ± 0.1	10	6	8	7
	Dec.	6		4.0	42	75 ± 3	2.4 ± 0.2	20	9	14	9
	Dec.	7,	1	3.5	97	46 ± 2	2.4 ± 0.2	6	9	5	9
			2	5.6	35	61 ± 2	1.7 ± 0.1	11	7	8	8
			3	1.1	34	97 ± 6	3.6 ± 0.4	>30	12	>30	12
Median					36	63	2.4	12	9	9	9

* Collected at Assabu.

** Errors are 1σ values calculated from the counting errors.

the activity ratios of ^{210}Bi to ^{210}Pb are near unity for some samples, most of the ratios fall in the range from 0.5 to 0.8, which are nearly comparable to or slightly larger than those in precipitation observed by PoET *et al.* (1972). For the snow the median of $^{210}\text{Bi}/^{210}\text{Pb}$ ratio is smaller than that for the rain, but the median of $^{210}\text{Po}/^{210}\text{Pb}$ ratio for the snow is somewhat larger than that for the rain. This fact is discussed later on the basis of the concept of residence time. The concentration of ^{210}Pb is

apt to decrease with the course of rainfall, although the ratio of $^{210}\text{Bi}/^{210}\text{Pb}$ increases. This may be due to the fact that the washout of younger aerosols in the lower atmosphere contributes more seriously to the earlier precipitation. This fact is not so obvious for the snowfall from convective clouds.

In Table 3 the results observed at Assabu are shown. The samples were collected during the period 1 ~ 3 Feb. 1972. The surface weather maps at 09JST, 2 Feb. and 09JST, 3 Feb. are shown in Fig.3. The low was moving toward the north-east over the Japan Sea and the cold front passed through in the evening at about 17JST, 2 Feb. After the passing of the cold front, it was a typical weather in winter in Japan; The north-west monsoon blew and snow fell from the cold sky at Assabu. In Fig.4, a marked contrast can be found for the concentrations of chloride and ^{210}Pb in the snow before and after the passing of the cold front. The variation may be chiefly due to the difference between the air masses. Thus the air mass from the continent contains much radon and its daughters, and contains much sea salt introduced by the strong wind traversing over the Japan Sea (TSUNOGAI, 1974).

Table 3. Concentrations of Pb-210, activity ratios of Bi-210 and Po-210 to Pb-210 and calculated residence times, T_{CE} , T_{CF} , T_C , T_{SE} , T_{SF} and T_S and the contribution of the upper atmosphere, A_{DU}/A_D for snow collected at Assabu on Feb. 1 to 3, 1972

No.	Precipitation mm	Pb-210 dpm/l	Bi-210*	Po-210*	CRM				SRM			
			Pb-210 $\times 10^2$	Pb-210 $\times 10^2$	T_{CE} day	T_{CF} day	T_C day	A_{DU}/A_D %	T_{SE} day	T_{SF} day	T_S day	A_{DU}/A_D %
1 Feb.1,22:40 -Feb.2, 02:00	0.83	40	41 ± 1	3.9 ± 0.3	5.0	13	4.5	4.8	3.2	13	2.6	6.5
2 Feb.2, 02:00-03:00	0.76	12	68 ± 10	17 ± 1	16	49	5.2	25	11	47	3.7	30
3 Feb.2, 07:00-08:30	1.33	13	73 ± 11	10 ± 1	20	30	17	8.2	14	30	12	14
4 Feb.2, 10:50-12:00	2.83	6.2	90 ± 8	7.8 ± 0.8	64 (>32)	23	-	-	35 (>19)	23	-	-
5 Feb.2, 12:00-13:00	2.80	3.5	72 ± 11	3.8 ± 0.6	19	13	-	-	13	13	-	-
6 Feb.2, 13:00-15:00	1.92	8.4	86 ± 15	8.2 ± 1.2	43 (>17)	24	-	-	25 (>12)	24	-	-
7 Feb.2, 15:20-16:00	0.72	17	54 ± 11	10 ± 1	8.6	30	6.4	15	5.7	30	3.6	19
8 Feb.2, 19:45-22:40	0.86	23	48 ± 6	8.1 ± 0.8	6.6	24	5.1	11	4.1	24	2.9	15
9 Feb.2,22:40 -Feb.2, 04:40	0.47	133	49 ± 3	3.7 ± 0.3	6.9	12	6.5	3.5	4.4	13	3.9	5.9
10 Feb.3, 04:40-09:50	0.62	190	39 ± 1	1.6 ± 0.2	4.7	6.2	4.6	0.7	3.0	6.2	2.9	1.7
11 Feb.3, 10:50-11:25	0.52	141	31 ± 1	1.5 ± 0.1	3.3	6.4	3.1	1.6	1.9	6.5	1.7	2.4
12 Feb.3, 11:25-11:50	0.63	107	28 ± 1	1.1 ± 0.1	2.8	5.4	2.7	1.2	1.6	5.4	1.4	1.8
13 Feb.3, 11:50-13:00	0.88	138	47 ± 2	3.3 ± 0.2	6.5	11	6.0	3.2	4.1	12	3.7	5.2
14 Feb.3, 13:00-20:00	0.23	188	53 ± 2	2.1 ± 0.2	8.0	8.1	8.0	0.2	5.2	8.2	4.9	2.3

* Errors are 1σ values calculated from the counting errors.

At Assabu, the maximum concentration of ^{210}Pb observed in the snow is 190 dpm/l which is the highest among the concentrations ever reported. The mean concentration of ^{210}Pb is 31 dpm/l for the precipitation in winter (Nov.~March), which has been obtained from the two-year observation by FUKUDA and TSUNOGAI (1974). These concentrations are a few to a few ten times higher than the mean value for the world precipitation, 5 dpm/l estimated by BURTON and STEWART (1960). MIYAKE and KUBOTA (1966) also observed high concentrations of ^{210}Pb in the snow by the winter monsoon over Shishiku, Ishikawa Pref., the middle part of Japan, of which maximum was 164 dpm/l for the snow precipitated on 3 Feb. 1966, although the concentration varied widely from sample to sample depending on the meteorological conditions. Radon daughter nuclides seem to be useful for the study of snowfall in Japan.

The residence time of tropospheric aerosols

If the distribution of radon in the troposphere is the same as that of aerosols and radon daughters of metal elements adhere to neighboring aerosols as soon as they are produced from gaseous radon, the residence time of tropospheric aerosols can be estimated by using radon daughters in the air or precipitation. However, modeling or some assumptions are needed for the practical estimation of the residence time. We consider the following two models.

1) Continuous removal model (abbreviated as CRM)

This model has been used by many researchers (e.g. BURTON and STEWART, 1960; JUNGE, 1963), in which the production and removal of atmospheric aerosols are assumed

Fig.3. Surface weather maps at 09JST, 2 Feb. and 09JST, 3 Feb. 1972.

Fig.4. Concentrations of chloride (Cl) in ppm and ^{210}Pb in dpm/l for samples collected at Assabu on 2-3 Feb. 1972.

to be constant and the removal process is assumed to be the first order with respect to the concentration. The time change of numbers (N) of atoms of radon daughters (^{210}Pb abbreviated as D, ^{210}Bi as E and ^{210}Po as F) in aerosols is expressed as follows,

$$dN_D/dt = R - (\lambda_D + \lambda)N_D \quad (1)$$

$$dN_E/dt = \lambda_D N_D - (\lambda_E + \lambda)N_E \quad (2)$$

$$dN_F/dt = \lambda_E N_E - (\lambda_F + \lambda)N_F \quad (3)$$

where R is the production rate of ^{210}Pb , that is, the decay rate of ^{222}Rn and λ is the removal rate of aerosols or the decay constants of radon daughters. Thus the reciprocal of $\lambda(1/\lambda = T)$ is the residence time of aerosols or the mean-life of radon daughters. At a steady state, the solutions of above equations are written by using radio-activity (A) as follows,

$$A_E/A_D = T/(T + T_E) \quad (4)$$

$$A_F/A_D = T^2/(T + T_E)(T + T_F) \quad (5)$$

This model is applicable only to a mean value obtained by sufficiently long observations in a sufficiently vast area. In actual precipitations, T from equation (4) does not always agree with that from equation (5) owing to some causes which are discussed later. Thus the residence times calculated from the activity ratios, A_E/A_D and A_F/A_D , by using CRM are designated as T_{CE} and T_{CF} , respectively, in the following discussion.

2) Sudden removal model (abbreviated as SRM)

The residence times obtained from CRM are not always applicable when residence times of individual precipitations, especially those of the snow produced by the north-west monsoon in winter in Japan, are compared, because the radon daughters are steadily formed in the continental air mass and suddenly removed from the air mass over Japan. Therefore, we assume another model where aerosols in the air are accumulated with a constant rate during the time between two subsequent precipitation events, after formerly accumulated aerosols are almost perfectly removed from the air by the former precipitation. In this case, the mean residence time (T) of aerosols is equal to a half of the time from the beginning of the accumulation of aerosols to the last precipitation. Thus the numbers of radon daughters produced in the atmosphere are obtained by integrating the following equations from $t = 0$ to $t = 2T$;

$$dN_D/dt = R - \lambda_D N_D \quad (6)$$

$$dN_E/dt = \lambda_D N_D - \lambda_E N_E \quad (7)$$

$$dN_F/dt = \lambda_E N_E - \lambda_F N_F \quad (8)$$

The notations are the same as in the case of CRM. By using activity ratios, the solutions of above equations are expressed as

$$A_E/A_D = \frac{1}{1 - e^{-2T/T_D}} \left(1 - \frac{e^{-2T/T_D}}{1 - T_E/T_D} - \frac{e^{-2T/T_E}}{1 - T_D/T_E} \right) \quad (9)$$

$$A_F/A_D = \frac{1}{1 - e^{-2T/T_D}} \left\{ 1 - \frac{e^{-2T/T_D}}{(1 - T_E/T_D)(1 - T_F/T_D)} - \frac{e^{-2T/T_E}}{(1 - T_D/T_E)(1 - T_F/T_E)} - \frac{e^{-2T/T_F}}{(1 - T_D/T_F)(1 - T_E/T_F)} \right\} \quad (10)$$

In a similar manner as for CRM, the residence times calculated from A_E/A_D and A_F/A_D are designated as T_{SE} and T_{SF} , respectively. Even though the removal of aerosols by each rainfall was not so perfect, the residence time would have some meanings similar to that in CRM for individual rain.

In Fig.5, the residence times (T_{CF} , T_{CE} , T_{SE} and T_{SF}) are shown as the function of the activity ratio, A_E/A_D or A_F/A_D . The calculated residence times for rains and snows are also given in Tables 2 and 3. As these values have considerable errors especially for large T_{CE} or T_{SE} values, it is not so meaningful to discuss individual values.

The residence time of aerosols in rains calculated from A_F/A_D , T_{CF} or T_{SF} , is apt to be shorter than that from A_E/A_D , T_{CE} or T_{SE} . This may be due to the analytical error for determinations of ^{210}Po in the rain, which results from the adsorption of ^{210}Po in solution to a sampler of polyethylene when the rain water flew rather slowly on the polyethylene plates. The snow, on the other hand, was melted after the sampling. Thus the residence time calculated from A_F/A_D for the rain is not so reliable, although T_{CE} or T_{SE} for the rain at Hakodate is certainly larger than one week. As shown in Table 2, the residence time from A_F/A_D for

Fig.5. The relation between the activity ratio of ^{210}Bi to ^{210}Pb (A_E/A_D) or ^{210}Po to ^{210}Pb (A_F/A_D) and the residence time (T).

the snow is nearly equal to that from A_E/A_D especially in SRM. The difference between the residence times from A_E/A_D and A_F/A_D is discussed in the next section by using data for the snow obtained at Assabu, which are more precise because of the high concentrations of these nuclides and the rather rapid separation of ^{210}Bi from ^{210}Pb after the sampling.

Supply of aerosols having a longer residence time to tropospheric aerosols in precipitation

As shown in Table 3, the residence time from A_F/A_D , T_{CF} or T_{SF} is somewhat larger than that from A_E/A_D , T_{CF} or T_{SE} for snow samples collected at Assabu. Similar results were obtained for aerosols in the air and precipitation over the continental U.S.A. by POET *et al.* (1972) who attributed the cause to ^{210}Po from the surface soil. At Assabu, however, the ground surface was covered by snow and the contamination of ^{210}Pb in equilibrium with ^{226}Ra in the surface material to precipitation seems to be negligibly small as discussed by MARENCO and FONTAN (1972, 1973). The reported concentrations of ^{210}Pb in the surface soil are 0.3 pCi/g (MARENCO and FONTAN, 1972) and $3 \sim 8$ pCi/g (VILENSKIY, 1970). The snow at Assabu contained more than 50 pCi/l of ^{210}Pb and less than 10 mg/l of soil dust which was derived from the continent. Thus, the surface material contributes only by 10% for ^{210}Po or 0.3% at most for ^{210}Pb to the snow produced from the north-west monsoon in winter in Japan. Accordingly, we tentatively assume that the excess ^{210}Po in snow comes from the lower stratosphere or the upper troposphere. If we can estimate or assume the residence time of aerosols from the upper atmosphere, the contribution of aerosols from the upper atmosphere and the true residence time of aerosols in the troposphere or in the lower atmosphere are calculated as the solutions of the following simultaneous equations. By putting $A_E/A_D = X$ and $A_F/A_D = Y$ and by using suffixes L and U for components of the lower atmosphere and the upper atmosphere, respectively, we have

$$A_{DU} = A_{EU}/X_U = (A_E - A_{EL})/X_U = (A_E - A_{DL}X_L)/X_U \quad (11)$$

$$A_{DU} = A_{FU}/Y_U = (A_F - A_{FL})/Y_U = (A_F - A_{DL}Y_L)/Y_U \quad (12)$$

$$A_{DU} = A_D - A_{DL} \quad (13)$$

$$X_L = f(T) \quad (14)$$

$$Y_L = g(T) \quad (15)$$

where $f(T)$ is the function of T defined by equation (4) or (9), and $g(T)$ is defined by equation (5) or (10).

The values of X_U and Y_U are computed by assuming the residence time of aerosols of the upper atmosphere to be one year on the basis of the estimation by KATSURAGI (1965). Being calculated from the above simultaneous equations, the residence time of aerosols of the lower atmosphere (tropospheric aerosol), T_C in CRM or T_S in SRM, and the percentage of aerosols of the upper atmosphere (aerosol of the lower stratosphere), A_{DU}/A_D , are shown in Table 3.

For the snow during the north-west monsoon (samples No.9 to 14 in Table 3), the residence time of tropospheric aerosols is 2 to 7 days, which is close to that calculated from the activity ratio, A_E/A_D . The activity ratio, A_F/A_D has been used chiefly for the estimation of the residence time of tropospheric aerosols, but the residence time is not so reliable because the ratio, A_F/A_D is so small that T_{CF} or T_{SF} is largely affected by the contamination of relatively small amounts of material having the ratio near unity. Although the residence time of aerosols in the north-west monsoon over Japan is short in accordance with the conclusion of POET *et al.* (1972), the residence time calculated from radon daughters varies widely reflecting the origin and history of the air mass. Therefore, the residence time may be one of the promising means of studying meteorological problems.

The proportion of aerosols of the lower stratospheric origin is 1 to 5% for snow produced by the north-west monsoon on 3 Feb. 1972 at Assabu. The amount of these aerosols precipitated, however, is not small, because the concentration of ^{210}Pb is fairly large. The percentage of aged aerosols is larger for the snow produced by the warm front on 2 Feb. Their absolute amount in the snow, however, is not large. Based on only these data, we cannot conclude whether aged aerosols come from the lower stratosphere or the maritime air mass of the North Pacific. Even though the contribution of aged aerosols is small, it can be easily estimated by precisely determining ^{210}Pb , ^{210}Bi and ^{210}Po in precipitation. It is our future problem to determine the contribution of stratospheric aerosols and to relate it with other chemical constituents such as nitrogen and sulfur compounds.

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