Article

Variation in the Atmospheric Concentrations of ^{214}Pb with Local Meteorological Conditions at the Observation Site —Observation at Kawasaki, Japan—

Nanako MATSUZAWA[†], Yuya KOIKE^{*,††} and Jun SATO^{**,*}

Graduate School of Environmental and Information Sciences, Yokohama National University
79-7 Tokiwadai, Hodogaya-ku, Yokohama-shi, Kanagawa Pref. 240-8501, Japan

*Radioisotope Center, The University of Tokyo

2-11-16 Yayoi, Bunkyo-ku, Tokyo 113-0032, Japan

**Department of Applied Chemistry, School of Science and Technology, Meiji University
1-1-1 Higashi-mita, Tama-ku, Kawasaki-shi, Kanagawa Pref. 214-8571, Japan

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Every four hours variations in atmospheric ^{214}Pb concentrations were observed at Kawasaki, Japan. The monthly averaged concentration ranged from 1.3 to 4.1 Bq/m³. The variation patterns within 24 hours of the atmospheric concentration of short-lived ^{214}Pb (half-life : 26.8 min) is similar to that of ^{212}Pb (half-life : 10.6 h). The variation in atmospheric concentrations of ^{214}Pb was observed to depend strongly on the local meteorological conditions in a short period. High concentrations of ^{214}Pb tend to appear on the days with westerly wind from inland area, low wind velocity and without precipitation.

Key Words : atmospheric concentration of lead-214, Kawasaki, Pacific side area of Japan, every four hour variation, local meteorological condition

1. Introduction

Atmospheric ^{214}Pb (half-life : 26.8 m) and ^{212}Pb (half-life : 10.6 h) are the progenies of ^{222}Rn (half-life : 3.82 d) and ^{220}Rn (half-life : 55.6 s), respectively, which are emitted from the ground surface into the atmosphere, and exist in the atmosphere attached to aerosol particles.

Lead-214 has been used to observe the atmospheric diffusion of ^{222}Rn ¹⁾ and the exhalation of ^{222}Rn from the ground²⁾. Lead-212, the longest-lived decay product of ^{220}Rn , has also

been used as a tracer to estimate the degree of vertical mixing of the surface air³⁾. Concentrations of the atmospheric ^{212}Pb have been investigated world-widely^{3)–16)}, and observations of the atmospheric ^{222}Rn and its short-lived daughter products, ^{218}Po , ^{214}Pb and ^{214}Bi , have appeared in some literatures^{5), 8), 15)}.

As the main source of the atmospheric ^{222}Rn and ^{220}Rn is the ground surface, the concentrations of ^{214}Pb and ^{212}Pb within the surface air will reflect local meteorological conditions together with the geology of the observing locality and the neighboring area. Actually, it has been reported that the atmospheric concentration of ^{214}Pb and ^{212}Pb could vary by the local rainfall, wind velocity and wind direction^{1), 16)}. Ho and Measday¹³⁾ pointed out that the ^{212}Pb

[†] Present address : Nippon Oil Corporation, Chidoricho, Naka-ku, Yokohama-shi, Kanagawa Pref. 231-0815, Japan.

^{††} To whom all correspondences should be addressed.

concentration was reduced by heavy rainfall, high wind velocity and low temperatures, and that the concentration was also dependent on the wind direction, while atmospheric inversions have little impact. The atmospheric ^{212}Pb concentrations at Yokosuka ($35^{\circ}20'\text{N}$, $139^{\circ}35'\text{E}$), located on Tokyo Bay, were observed to be around 20 Bq/m^3 and remained at low level throughout the year⁴⁾, which was the lowest level observed previously at Kawasaki ($35^{\circ}36'\text{N}$, $139^{\circ}31'\text{E}$), an adjacent city of Tokyo¹⁷⁾.

The atmospheric concentrations of ^{214}Pb at Yokosuka⁴⁾ ranged from 1.1 to 2.8 Bq/m^3 , showing a slight seasonal variation of “one-peak” pattern: high in winter with low precipitation. The average atmospheric concentration of ^{214}Pb was observed to be 1.84 Bq/m^3 at Tokushima¹⁵⁾ in the period from April 2002 to January 2003, showing a slight seasonal variation of “one-peak” pattern: also low in summer and high from autumn to winter.

The present report deals with every four hours variation in the atmospheric concentration of ^{214}Pb observed at Kawasaki located in the Pacific side area of Japan with varying meteorological conditions by confirming the every four hours observation with ^{212}Pb carried out previously at the same observing locality¹⁷⁾. The discussion will be extended to the contribution of the local meteorological conditions to the atmospheric concentrations of the two lead isotopes, ^{214}Pb and ^{212}Pb , having significantly different half-lives.

2. Experimental

Collection of aerosol samples was made from March 2003 to February 2004 on a terrace, 10 m above the ground surface, of a building of Meiji University, 60 m above sea level, Kawasaki, on glass fiber filters (Advantec, GB-

100R) using a high volume air sampler (Sibata, HVC-500) at an aspirating speed of $0.6\text{ m}^3/\text{min}$. Every collection lasted for four hours. Each filter was cut out to make four disks of 38 mm diameter and pressurized to prepare the source of 3.2 mm thick, and packed into an acrylic canister having a window of a thin Mylar film (Chemplex, $3.6\text{ }\mu\text{m}$ thick) for the non-destructive γ -ray measurement. The portion recovered for the measurements was 58% of the effective surface area of the filter. Radioactivity of ^{214}Pb and ^{212}Pb was determined by the 351.9-keV and 238.6-keV γ -rays, respectively, with an HPGe spectrometer.

3. Results and Discussion

3.1 Seasonal variation of atmospheric ^{214}Pb concentrations

Figure 1 shows the monthly average of every four hours concentrations of atmospheric ^{214}Pb at Kawasaki from March 2003 to February 2004. Concentration of atmospheric ^{214}Pb increased from autumn to winter, and decreased in summer. The monthly averaged concentration of ^{214}Pb at the present observation site ranges from 1.3 to 4.1 Bq/m^3 and the seasonal variation shows “one-peak” variation pattern which is similar to that of ^{212}Pb ¹⁷⁾.

Fig. 1 Monthly average of every four hours concentrations of atmospheric ^{214}Pb at Kawasaki from March 2003 to February 2004.

The variation patterns of the monthly averaged atmospheric ^{222}Rn concentrations were observed at Niigata, Chiba, Saga, Miyagi, Hiroshima, Ehime, Hokkaido, Aichi, Shimane, Akita, Osaka and Kagoshima, all over the Japanese islands, from March 1986 to February 1987¹⁸⁾. Concentrations of atmospheric ^{222}Rn increased generally from autumn to winter, and decreased in summer, the variation pattern being similar to that of ^{214}Pb observed in the present study. Monthly averages of concentrations of atmospheric ^{222}Rn in the Pacific side of Japan were lower than those in the inland area of Japan.

3.2 Variation within 24 hours

Figure 2 shows the variation in the every four hours concentration of atmospheric ^{214}Pb at Kawasaki from March 2003 to February 2004. The variation patterns within 24 hours of the atmospheric concentration of short-lived

^{214}Pb are similar to that of ^{212}Pb concentration : maxima appeared in the every early morning (0:00–8:00) due to the stable atmosphere caused by the nocturnal inversion layer, and minima in the every late afternoon (12:00–16:00) due to the diurnal mixing of the atmosphere, which has previously been suggested¹⁷⁾.

The observation of the parent ^{222}Rn at Komae (35° 37' N, 139° 35' E), adjacent city of the present site, also showed that high concentrations appeared in the early morning¹⁹⁾. These observations are reflected in that the mixing in the surface air is generally active in the afternoon and the surface air is stable at night, and abrupt alternation takes place at sunrise and at sunset⁵⁾.

3.3 Distribution of ^{214}Pb concentrations

Figure 3 shows the histograms of ^{214}Pb and ^{212}Pb concentrations for the 921 every four hours observations at the present observation

Fig. 2 Every four hours concentrations of atmospheric ^{212}Pb and ^{214}Pb at Kawasaki from March 2003 to February 2004. The range for each month on the abscissa represents the number of days for observation.

Fig. 3 Logarithmic distribution of atmospheric concentrations of ^{214}Pb (■) and ^{212}Pb (□).

site of the limited area, which forms log-normal distribution.

3.4 Appearance of lower concentration

Figure 4 shows typical variations of atmospheric concentrations of ^{214}Pb and ^{212}Pb with precipitation in May 2003. Concentrations of ^{214}Pb and ^{212}Pb decreased with precipitation. The low concentration observed in summer (Fig. 1) may be due to a number of rainy days together with large amounts of precipitation.

Figure 5 shows correlation between atmos-

Fig. 4 Typical variations of atmospheric concentrations of ^{214}Pb (●) and ^{212}Pb (○) with precipitation in May 2003.

Fig. 5 Correlation between atmospheric ^{214}Pb concentrations and wind velocity.

Fig. 6 Sampling location at Kawasaki.

pheric ^{214}Pb concentrations and wind velocity. The concentrations tended to decrease on the day of higher wind velocity: high concentration of ^{214}Pb in the lower layer of surface air may be diluted with the air of low concentration of ^{214}Pb at higher layer by possible turbulence.

Figure 6 shows the present sampling location at Kawasaki. When the wind blew from the oceanic area, the atmospheric concentrations tended to decrease.

3.5 Appearance of higher concentration

Table 1 lists the varying meteorological conditions under which high concentration of ^{214}Pb was observed. High concentrations tended to

appear on the day without precipitation, with low wind velocity, and with wind direction from inland area, and with stable atmosphere in the early morning. Combination of the four meteorological conditions makes high concentrations of ^{214}Pb . Table 2 lists the unusually high atmospheric ^{214}Pb concentration appeared

Table 1 Meteorological conditions for unusually high concentration

Meteorological conditions
1. Non-precipitation
2. Wind velocity of less than $3\text{ m}\cdot\text{s}^{-1}$
3. From 0 o'clock to 8 o'clock
4. Wind from inland area

Table 2 Unusually high atmospheric ^{214}Pb concentrations

Sampling period	Concentration [Bq · m ⁻³]	Precipitation [mm]	Wind velocity [m · s ⁻¹]	Wind direction
Mar. 16, 2003, 4:00 - 8:00	7.36	0.0	1.1	NW
Apr. 18, 2003, 0:00 - 4:00	7.60	0.0	0.9	W
Apr. 18, 2003, 4:00 - 8:00	7.25	0.0	0.3	W
Nov. 1, 2003, 0:00 - 4:00	7.86	0.0	1.2	NW
Nov. 1, 2003, 4:00 - 8:00	6.78	0.0	0.8	N
Nov. 2, 2003, 0:00 - 4:00	10.82	0.0	0.4	NW
Nov. 2, 2003, 4:00 - 8:00	11.04	0.0	1.0	NW
Nov. 3, 2003, 16:00 - 20:00	7.94	0.0	0.8	N
Nov. 3, 2003, 20:00 - 24:00	8.21	1.0	1.4	NW
Nov. 4, 2003, 0:00 - 4:00	7.05	0.0	0.9	W
Nov. 14, 2003, 4:00 - 8:00	7.31	0.0	1.3	NW
Nov. 19 2003, 0:00 - 4:00	9.05	0.0	0.8	N
Nov. 19 2003, 4:00 - 8:00	7.47	0.0	1.6	NW
Nov. 20, 2003, 4:00 - 8:00	7.02	0.5	1.9	N
Jan. 19, 2004, 20:00 - 24:00	8.75	0.0	1.1	W
Jan. 20, 2004, 0:00 - 4:00	9.50	0.0	1.6	NW
Jan. 20, 2004, 4:00 - 8:00	8.61	0.0	1.5	W
Feb. 13, 2004, 4:00 - 8:00	10.54	0.0	1.4	NW
Feb. 17, 2004, 4:00 - 8:00	8.44	0.0	1.5	N

outside of 2σ of the distribution of the whole observed data. The data showed that the appearance of high ^{214}Pb concentration depends on the combination of the four parameters in Table 1. The classified local meteorological conditions for higher concentrations of ^{214}Pb are illustrated in Fig. 7.

3.6 Atmospheric behavior of ^{214}Pb and ^{212}Pb

Similar atmospheric behavior of short-lived ^{214}Pb to that of ^{212}Pb was observed. Considering

the half-lives of the parent nuclides of ^{214}Pb and ^{212}Pb , collective half-lives are 3.8 days for ^{214}Pb in the ^{222}Rn – ^{214}Pb combination, and 0.44 days for ^{212}Pb in the ^{220}Rn – ^{212}Pb combination (Fig. 8). Because both half-lives of ^{222}Rn – ^{214}Pb and ^{220}Rn – ^{212}Pb are in day order, concentrations of ^{214}Pb and ^{212}Pb may show similar behavior to each other.

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Fig. 7 Contribution of the local meteorological conditions to the unusually high concentrations (Table 2). (1) 0:00–4:00; (2) 4:00–8:00; (3) 16:00–20:00; (4) 20:00–24:00

Fig. 8 Combination of half-lives for the collective half-lives of ^{222}Rn – ^{214}Pb and ^{220}Rn – ^{212}Pb pairs.

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要 旨

**観測地点の局所的気象条件による大気中の ^{214}Pb 濃度の変動
—川崎市における観測—**

松澤菜々子[†], 小池裕也*, 佐藤 純**

横浜国立大学大学院環境情報学府環境システム学専攻

240-8501 神奈川県横浜市保土ヶ谷区常盤台 79-7

*東京大学アイソトープ総合センター

113-0032 東京都文京区弥生 2-11-16

**明治大学理工学部応用化学科

214-8571 神奈川県川崎市多摩区東三田 1-1-1

大気中の ^{214}Pb 濃度を川崎市で4時間毎に観測した。観測された大気中 ^{214}Pb の月平均濃度は1.3～4.1 Bq/m³であった。短寿命核種である ^{214}Pb （半減期：26.8分）の大気中濃度の24時間変動パターンは、 ^{212}Pb （半減期：10.6時間）の濃度変動パターンと類似していた。短寿命の ^{214}Pb の大気中濃度変動は、短時間の局所的気象条件との間に強い関連性が観測された。大気中の ^{214}Pb 濃度は、内陸部からの西寄りの風が吹き、風速が小さく、降雨のない日に高くなる傾向があった。

[†]現所属：新日本石油株式会社, 231-0815 神奈川県横浜市中区千鳥町8番地