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High Resolution Simultaneous Measurements of Airborne Radionuclides at Sub-regional Sampling Points by Ultra Low Background Gamma Spectrometry

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INTRODUCTION

In our environment, there exist many kinds of radionuclides which have half-lives ranging from sub-millisecond to longer than millions of years. Therefore, measurements of suitable airborne radionuclides provide useful information on time scale of processes in the atmosphere.

Airborne radionuclides attached on aerosol particles have been used as valuable tracers to investigate movement of air mass, removal processes of airborne contaminants and their residence times and so on. In the present work, sampling of high volume of air (10^2-10^4 m^3) coupled with non-destructive gamma spectrometry have been performed to investigate above mentioned process based on the measurements of terrestrial ^{210}Pb ($T_{1/2}$=22.3 a) and cosmogenic ^{7}Be ($T_{1/2}$=53.3 d) by regular sampling of one to several day(s) and by extremely short sampling of 2 to several hours. Only a sparse data can be obtained with relatively long sampling time since variation of airborne radionuclides is expected to occur very sensitively to meteorological changes (~several hours) such as passage of cold front, typhoon and so on. Hence, it is important to make monitoring of short-lived radionuclides such as ^{214}Pb ($T_{1/2}$=26.8 min) and ^{212}Pb ($T_{1/2}$=10.6 hrs) with sampling periods of several hours together with ^{210}Pb and ^{7}Be. In addition, samplings were made simultaneously at multiple points to obtain spatial information on airborne materials. This kind of investigation has not been performed so far because of the difficulty of measurements due to low radioactivity of long-lived radionuclides and also to the rapid decay of short-lived ones. The difficulties of measurements were overcome by using 11 extremely low-background Ge detectors installed in Ogoya Underground Laboratory (OUL) of Low Level Radioactivity Laboratory, Kanazawa University.

In this study, the results of high resolution temporal variations of airborne ^{212}Pb along with ^{210}Pb and ^{7}Be observed at three monitoring points in Ishikawa Prefecture of Japan in conjunction with the results from daily monitoring are presented.

MATERIALS AND METHODS

Activity levels of airborne radionuclides were monitored at three points viz, 1) Low Level Radioactivity Laboratory (LLRL, 40 m above mean sea level), 2) Shishiku Plateau (640 m) located about 8 km east of LLRL to investigate vertical distribution of radionuclides, and 3) Hegura Island (10 m) lying about 50 km north away from Noto Peninsula in the Japan Sea (about 180 km north of LLRL) to evaluate the influence of Asian continent or of mainland of Japan. Location map of monitoring points is shown in Figure 1.

Radionuclides attached to airborne particles were collected on silica fiber filter (Advantec QR-100, 25.4 cm x 20.3 cm) by using high volume air sampler (SIBATA HV-1000F) operated at flow rate of 800-900 L min^{-1}. Short (2-6 hrs) and long (1-7 day(s)) sampling were aimed to investigate diurnal and seasonal variation of airborne radionuclides, respectively. Gamma ray counting sources

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(35 mm in diameter) were prepared from 3/4th portion of each filter samples using hydraulic press immediately after the sampling to measure short-lived ^{212}Pb. Gamma ray measurements of ^{212}Pb was performed for 4-12 hours by Ge detectors at LLRL for the samples collected at LLRL and at OUL for the samples collected at Shishiku and Hegura Island mainly because of low activity level of radionuclides which further lowered due to radioactive decay during transportation. After the measurement of ^{212}Pb, all samples were re-measured for long-lived ^{210}Pb and ^{7}Be for 2-4 days.

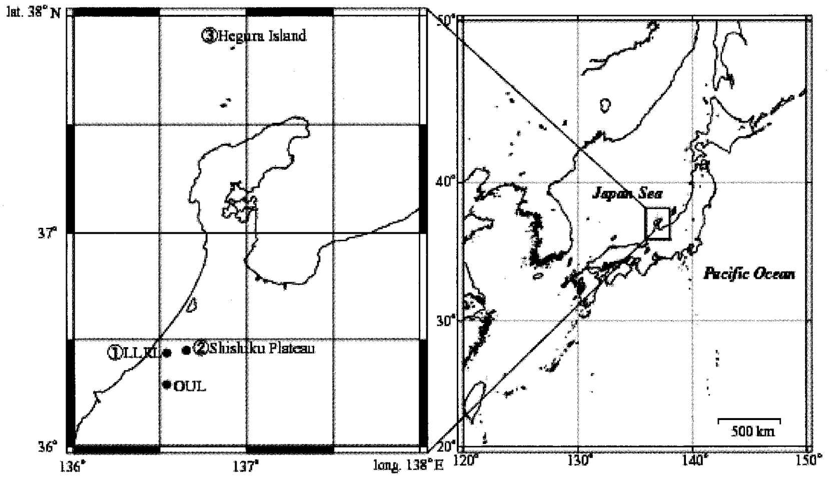

Figure 1 Location map of monitoring points and Ogoya Underground Laboratory.

RESULTS AND DISCUSSIONS

Activity levels and variation patterns of long-lived ^{210}Pb and ^{7}Be were found to be similar independent of the sampling points and radon-like diurnal variation was not observed. It is noteworthy that a rapid decrease both of ^{210}Pb and ^{7}Be activities was observed during the time just before the approach of cold front and passing over. Such decrease can be explained by scavenging of airborne particles from the atmosphere due to wash-out process and/or by intrusion of the air mass originating from low latitude ocean area containing lower ^{210}Pb and ^{7}Be. In addition, temporal increases of ^{210}Pb/^{7}Be ratios during the passage of cold front or typhoon were observed. These increases might be explained by passing of air mass with low ^{7}Be originated from lower latitude and by restraint of the inflow of air mass from upper atmosphere containing abundant ^{7}Be due to the updraft. This kind of rapid variation could not be observed by longer intervals of sampling made before.

Activity levels of short-lived ^{212}Pb at Shishiku Plateau (0.003-0.038 Bq m^{-3}) and Hegura Island (0.004-0.052 Bq m^{-3}) were measured to be nearly the same level, which were only approximately 1/4th of the level at LLRL (0.011-0.205 Bq m^{-3}). The lower activity at Hegura Island may be explained by lower production of ^{220}Rn, which is precursor of ^{212}Pb, released from only ~0.75 km^2 of Hegura Island and the lower activity at Shishiku Plateau might result from radioactive decay during transfer from near sea level to 640 m of height. Decrease of ^{212}Pb activity was observed when the cold front passed over just like those of ^{210}Pb and ^{7}Be. We consider that such lowering was caused by scavenging of airborne particles from the atmosphere by rainfall and/or dilution by air mass containing lower amount of airborne particles coming from ocean area. It is noteworthy that strong negative correlation was observed between ^{212}Pb concentration and wind velocity. This fact shows that short-lived ^{212}Pb can be used as a useful tracer to investigate transport of air mass in sub-regional area.

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