Measurements of short-lived cosmogenic nuclides in rain samples

● Measurements of Short-lived Cosmogenic Nuclides in Rain Samples

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Abstract

Extremely low activity levels of cosmic ray induced nuclides have been measured in freshly precipitated rainwater by quick chemical separation coupled with ultra low background gamma spectrometry. The nuclides detected were ^{38}S (half-life = 2.83 h)-^{38}Cl (37.2 m), ^{39}Cl (55.6 m), ^{18}F (109.7 m), ^{24}Na (14.96 h), ^{28}Mg (20.9 h), ^{7}Be (53.3 d) and ^{22}Na(2.602 y). Number of atoms in rain water were evaluated to be ranging from 400~1900 L^{-1} for ^{39}Cl (n=6, mean; 1200), 30~1500 L^{-1} for ^{24}Na (n=16, mean; 520), 80~600 L^{-1} for ^{28}Mg (n=13, mean; 260), $1x10^{6}$~$4x10^{7}$ L^{-1} for ^{7}Be (n=16, mean; $7x10^{6}$) and $2x10^{3}$~$1x10^{5}$ L^{-1} for ^{22}Na (n=9, mean; $2x10^{4}$), Number of atoms and their ratios were found to depend highly on weather conditions

1. Introduction

More than 20 cosmic ray produced (CP) nuclides have been discovered mainly in rainwater [1~11]. Among these nuclides, activity levels, (in other words production rate) of short-lived CP nuclides are extremely low, therefore, have not been used as radioactive tracers in hydrological and/or geochemical studies due mainly to the difficulty in measurements. By the use of ultra low-background Ge detector at Ogoya Underground Laboratory (OUL)[12, 13] and quick chemical procedures, we succeeded to detect many of the short-lived CP nuclides, i.e. ^{38}S (half-life = 2.83 h)-^{38}Cl (37.2 m), ^{39}Cl (55.6 m), ^{18}F (109.7 m), ^{24}Na (14.96 h) and ^{28}Mg (20.9 h) together with ^{7}Be (53.3 d) and ^{22}Na (2.602 y) in freshly precipitated rain water. In this paper, we describe detection techniques and some of the preliminary results obtained by recent experiments.

2. Experimental

2.1 Sampling of rain water

Since the activity levels of short-lived CP nuclides in rainwater are extremely low, large

volume of rainwater must be collected in a short-lime. At first, we tried to collect by using 4~6 plastic buckets placed on the roof of our laboratory (LLRL). This method, however, could collect only 10 L of rainwater within 30 min even in heavy rain. After various trials, we got an idea to use part of roof (29 m^2) as catchment area of rainwater and collect rainwater through downpipe. By this method, we succeeded to collect sufficient amount of rainwater without difficulty because 2 mm of precipitation correspond to 58 L of rainwater. A 50 L of rainwater could be collected within 5 min during heavy rain and 30 min during ordinary rain.

2.2 Quick chemical separation of CP nuclides

The second problem is to develop quick chemistry to separate quantitatively the CP nuclides within short time preferably 30 min. This problem was solved by the use of cation- and anion-ion exchange resin, POWDEX®-PCH and POWDEX®-PAO, which are widely used at nuclear power plants for chemical treatment of liquid waste contaminated with artificial radionuclides. By the preliminary experiments, it was confirmed that 2 g of cation and anion exchange resin per 10 L of rainwater was enough to concentrate quantitatively the CP nuclides. A 10 g aliquot of POWDEX®-PCH and POWDEX®-PAO were added to 50L of rain sample and stirred for ~10 min with propeller-type stirrer. The ion exchange resins absorbing the CP nuclides were settled on a bucket bottom and collected on a filter paper. Total time required for the chemical separation was 30~40 min in the case of 50 L sample. By this procedure, sample volume could be reduced to about 1/1000 of original one. Gamma ray counting source was prepared simply by packing the half-dried ion exchange resin to in a required geometry polyethylene bag.

2.3 Gamma ray measurements of the CP nuclides

Since the half-lives of CP nuclides are very short and their activity levels are extremely low, it is essential to start gamma ray measurements as quickly as possible by using ultra low background Ge detectors. It takes ~ 30 min drive by car to transport the sample from our laboratory (LLRL) to Ogoya underground laboratory, which is located at 21 km from LLRL. Counting of the CP nuclides could be started 70 ~ 75 min after the end of rainwater sampling.

Table 1 shows gamma ray peaks used for the determination of the CP nuclides. As known

from Table 1, most of the CP nuclide emits high-energy gamma rays with high abundance. Two large volume coaxial type Ge detectors with 91 and 93.5 % of detection efficiencies relative to a 7.6 cmϕ x 7.6 cm NaI (Tl) detector were used for the CP nuclides analysis. Gamma ray spectra were recorded at 20 ~ 30 min intervals during first 2-4 hours to determine short-lived CP nuclides such as ^{38}Cl and ^{39}Cl and also to reduce the interference of natural short-lived ^{214}Pb (27.0 m) and ^{214}Bi (19.7 m), which are progenies of airborne ^{222}Rn. After short time of measurements, a few hours of measurements were continued 1 ~ 2 days (mostly 1 day) for ^{24}Na and ^{28}Mg. Fig. 1 shows examples of gamma ray spectrum obtained for rain samples collected on Sep. 29, 2004. Gamma ray peaks derived from ^{38}Cl (supported by ^{38}S), ^{39}Cl, ^{18}F, ^{24}Na, ^{28}Mg, ^{7}Be and ^{22}Na could surely be detected although peak areas were extremely low.

2.4 Calculation of atom numbers of the CP nuclide

Since half-lives of most of the CP nuclide are short compared with counting time, activity of all of the CP nuclides even for ^{7}Be and ^{22}Na, was calculated by integral method. Peak area of the nuclide with half-life of $T_{1/2}$ for infinite measurement time was calculated by following equation.

$$I_{nfinity} = A_{obs}/(1 - \exp(-\ln(2)\, t/T_{1/2}),$$

where $A_{infinity}$ and $A_{obs,}$ are counts by infinite measurement and the one obtained by counting time of t, respectively. Validity of this method is shown in Fig. 2.

3. Results and Discussion

First rain water measurement was performed in last summer (June, 2004). Since then more than 30 measurements have been performed as of March, 2005. Number of atoms was calculated based on the integral count method described above. Here, specific activities of the CP nuclide during the 10~30 min of sampling were assumed to be constant. In this paper, we show preliminary results of the first 16 measurements performed until September, 2004.

3.1 Number of atoms of the CP nuclides per litter of rainwater

Table 2 summarizes the number of atoms of ^{24}Na, ^{28}Mg, ^{39}Cl, ^{38}S, ^{7}Be and ^{22}Na per litter of rain water and Fig. 3 show ratios of atom numbers of the CP nuclides relative to the most

abundant ^{7}Be. The values not given in the table are not reliable ones due to poor counting statistics. The errors are only of counting statistics(1 σ) . As known from Table 2, atom numbers of short-lived CP nuclides are only order of $10^{2} \sim 10^{3}$ atoms L^{-1} and vary largely rain by rain.

If horizontal and vertical movement of air-mass occur in similar time scale of half-lives of one of the short-lived CP nuclides, very useful information will be obtained by the measurement of CP nuclides in rain. The CP nuclides are also useful to investigate scavenging of airborne particles, natural and CP nuclides and also pollutants because these materials are attached on the surface of rain drop or incorporated inside of the rain drop and effectively scavenged onto ground surface. Measurements of the isotopic ratios such as ^{38}Cl/^{39}Cl and ^{28}Mg/^{24}Na is most promising to analyze such phenomena not only because half-lives of these pair are similar but also because these nuclides can be detected for most of rain samples.

Conclusion

Activity levels of the CP nuclides with half-lives shorter than 1 day could be detected by quick chemistry coupled with ultra low background gamma spectrometry. Number of atom of the CP nuclides in rain water were measured to be ranging from 30 ~ 1500 L^{-1} for ^{24}Na (n=16, mean; 520), 80 ~ 600 L^{-1} for ^{28}Mg (n=13, mean; 260), 400 ~ 1900 L^{-1} for ^{39}Cl (n=6, mean; 1200), $1x10^{6} \sim 4x10^{7}$ $L^{-1}$for ^{7}Be (n=16, mean; $7x10^{6}$) and $2x10^{3} \sim 1x10^{5}$ L^{-1} for ^{22}Na (n=9, mean; $2x10^{4}$). This study may be first application of short-lived CP nuclides will open new application fields in hydrology, meteorology, chemistry and geochemistry of atmosphere.

References

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Table and Figure Captions.

Table 1. Nuclear properties of the CP nuclides in rain samples measured in this study

Table 2. Number of atoms of the CP nuclides in unit volume of rain samples..

Figure 1. Gamma ray spectrum of rain samples collected on Sep. 29, 2004 at Tatsunokuchi, Ishikawa Prefecture, Japan..

Figure 2. Calculation of peak counts of infinity measurement by integral method.

Figure 3. Atom numbers of the CP nuclides relative to most abundant ^{7}Be.

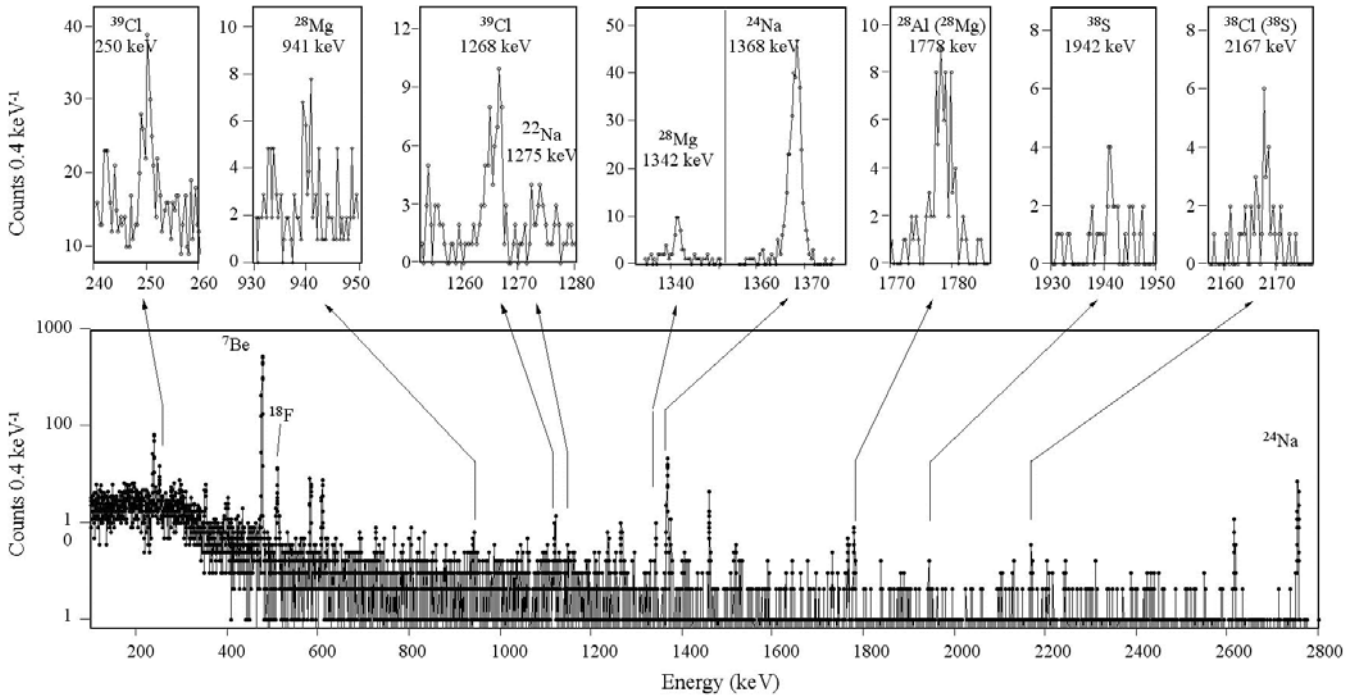

Fig. 1 Gamma ray spectrum of rain sample measured by ultra low-background Ge detector.

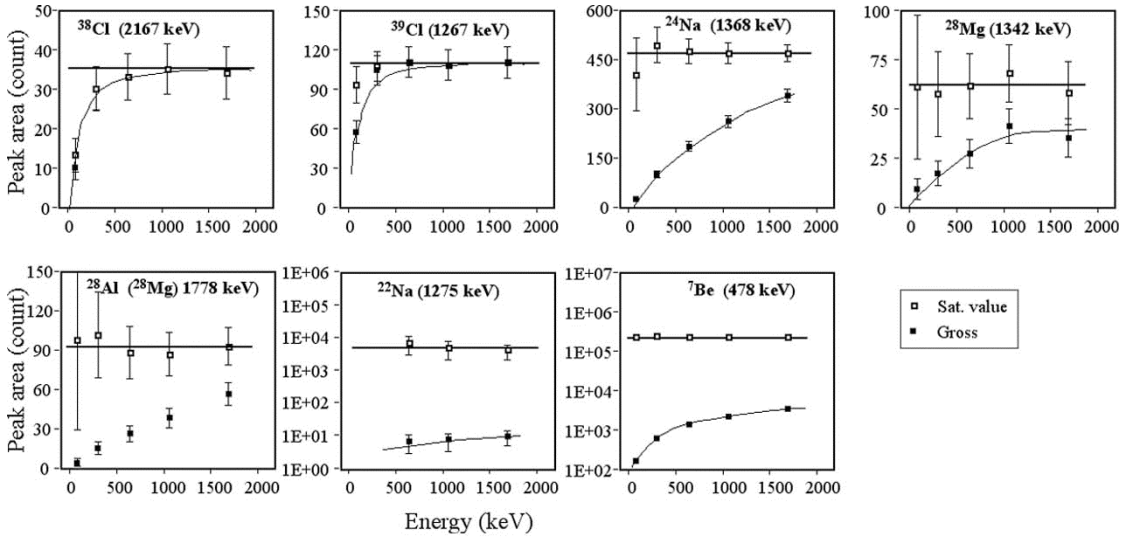
38Cl (2167 keV)
39Cl (1267 keV)
24Na (1368 keV)
28Mg (1342 keV)
28Al (28Mg) 1778 keV)
22Na (1275 keV)
7Be (478 keV)
Peak area (count)
Energy (keV)
Sat. value
Gross

Fig. 2

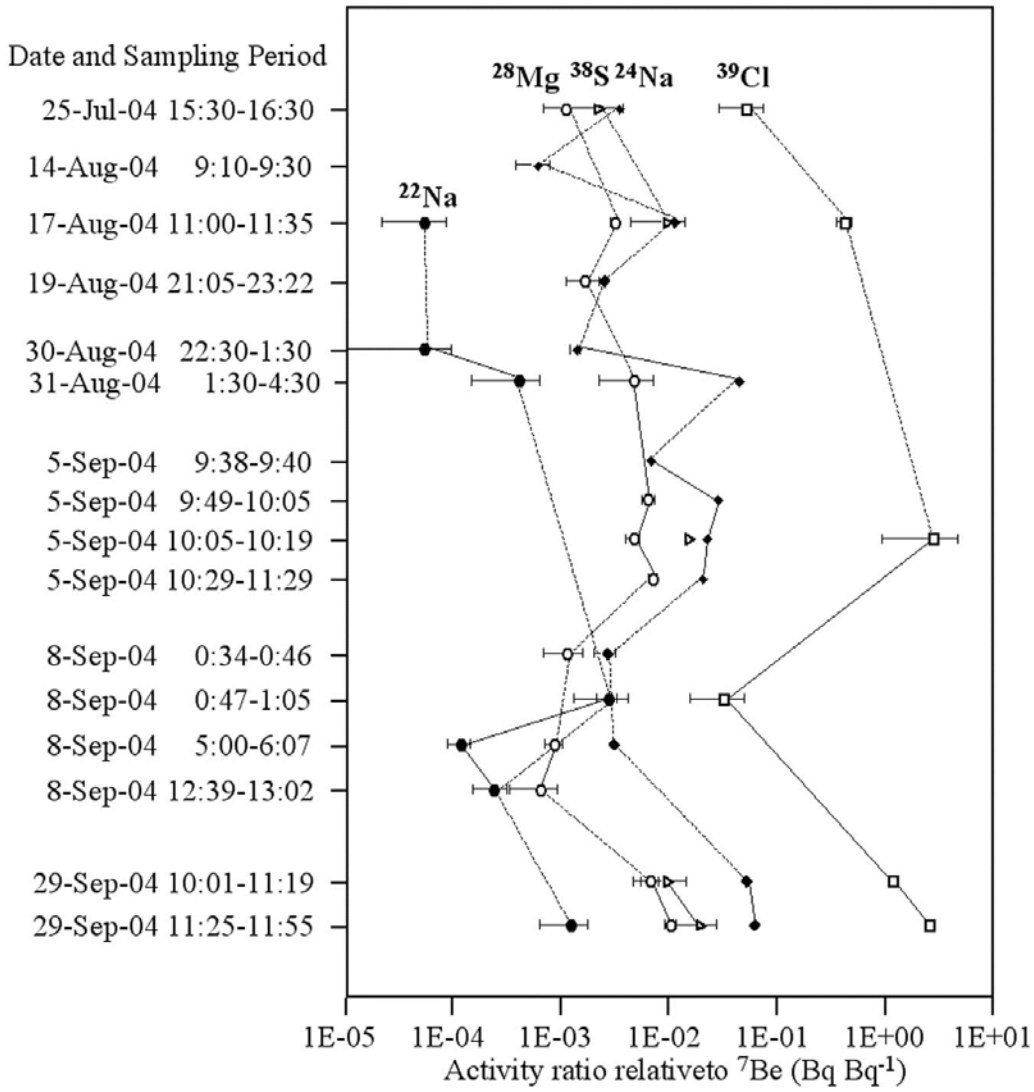
Date and Sampling Period
25-Jul-04 15:30-16:30
14-Aug-04 9:10-9:30
17-Aug-04 11:00-11:35
19-Aug-04 21:05-23:22
30-Aug-04 22:30-1:30
31-Aug-04 1:30-4:30
5-Sep-04 9:38-9:40
5-Sep-04 9:49-10:05
5-Sep-04 10:05-10:19
5-Sep-04 10:29-11:29
8-Sep-04 0:34-0:46
8-Sep-04 0:47-1:05
8-Sep-04 5:00-6:07
8-Sep-04 12:39-13:02
29-Sep-04 10:01-11:19
29-Sep-04 11:25-11:55
28Mg
38S
24Na
39Cl
22Na
1E-05
1E-04
1E-03
1E-02
1E-01
1E+00
1E+01
Activity ratio relativeto 7Be (Bq Bq-1)

Table 1. Nuclear properties of the CP nuclides in rain samples measured in this study

Nuclide	Half-liife	Gamma ray energy (keV)	Abundance (%)*	Remarks
^{39}Cl	55.6 m	1267.185	100	
^{38}S	179.3 m	1941.944	100?	
^{38}Cl	37.24 m	2167.405	42l4	daughter of ^{38}S
^{34}Cl	32.0 m	2127.492	100	
^{28}Mg	20.91 h	941.72	38.3	
		1342.25	52.6	
^{28}Al	2.241 m	1778.85	100	daughter of ^{28}Mg
^{24}Na	14.96 h	1368.63	100	
		2754.028	99.94	
^{22}Na	2.6019 y	1274.53	99.94	
^{18}F	109.77 m	511	100	
^{7}Be	53.29 d	477.59	10.52	

* Taken from Table of Isotopes 8th edition. No.1, 1996

Table 2. Activity levels of cosmic-ray produced nuclides measured for rain samples collected at Tatsunokuchi, Ishikawa Prefecture, Japan.

Sampling date	^{24}Na (1368 keV) (atom L $^{-1}$)	^{28}Mg (1342 keV) (atom L $^{-1}$)	^{39}Cl (1267 keV) (atom L $^{-1}$)	^{38}S (1942 keV) (atom L $^{-1}$)	^{7}Be (478 keV) (10^{4} atom L $^{-1}$)	^{22}Na (1275 keV) (10^{3} atom L $^{-1}$)
25-7-2004	± 20	217 ± 19	375 ± 158	56 ± 38	1080 ± 4	25 ± 9
14-8-2004	34 ±			-	381 ± 3	13 ± 6
17-8-2004	231 ± 15	101 ± 11	483 ± 66	38 ± 15	179 ± 2	2 ± 1
19-8-2004	223 ± 23	222 ± 66			727 ± 4	
31-8-2004 A	188 ± 19				1030 ± 6	13 ± 10
31-8-2004 B	459 ± 41	79 ± 36			105 ± 2	8 ± 4.9
5-9-2004 A	218 ± 32				275 ± 5	
5-9-2004 B	417 ± 30	140 ± 19	2790 ± 1799	64 ± 43	170 ± 3	
5-9-2004 C	843 ± 71	303 ± 43			476 ± 4	
5-9-2004 D	1079 ± 79	583 ± 55			763 ± 4	
7-9-2004	32 ± 17	110 ± 47			914 ± 4	19 ± 21
8-9-2004 A	249 ± 61	158 ± 57			763 ± 7	
8-9-2004 B	238 ± 49	343 ± 169	160 ± 80		702 ± 7	
8-9-2004 D	1420 ± 96	607 ± 95			378 ± 12	6.8 ± 8
30-9-2004 A	1471 ± 53	296 ± 49	1888 ± 154	58 ± 27	264 ± 4	
30-9-2004 B	716 ± 33	190 ± 21	1658 ± 145	52 ± 24	111 ± 2	27 ± 12