

2 学位論文概要

Studies on Measurements of Environmental Radioactivities Around a Nuclear Power Station (an excerpt)

Hisanobu Sakaue

Abstract

The thesis is composed of three chapters, of which the first chapter discusses about the position and significance of this study from a viewpoint of a local government. In Japan, it is an important duty of local governments to carry out continuous monitoring of radiation and radioactivity around nuclear power stations, in which estimated artificial dose should be reported to the public every year. Though recent levels of detected radioactivities in the monitoring are quite lower than those levels measured around 1970's, it is necessary to investigate their accurate distributions and behaviors as backgrounds. Therefore, continuous development of new device and analysis, which should be worked more rapidly and sensitively, must be confirmed. Following subjects were studied from these view points.

The study comprise of two parts. First part (Chapter 2) is related to radionuclide measurements around several dam sites close to Kashiwazaki-Kariwa area, especially ^{137}Cs in the water, to obtain information of its distribution and behavior. Main aim was to clarify the source of ^{137}Cs in tap water from this area, because the existence of the isotope had been scarcely observed in other sites in Japan. Next part (Chapter 3) contains the development of a high-speed counting system to measure micro-seconds pulse pairs selectively derived from correlated events of ^{214}Bi (beta) to ^{214}Po (alpha). The present method was tested as a sensitive detector for artificial radionuclides in the airborne dust collected on a filter paper, on the basis of accurate counting of the contribution from ^{222}Rn and its progenies.

Keywords: ^{137}Cs in the water; dam lakes and sediments; Mt. Yoneyama; andicite soil; Time Interval Analysis (TIA); correlated events of μsecond scale; high-speed time counting; on-line dust monitoring

1. General Introduction

Niigata prefecture is located central part of Honshu, Japan and faces the sea of Japan. One of noticeable thing there is that it hosts the world's largest nuclear power plant, Kashiwazaki-Kariwa nuclear power station of Tokyo Electric Power Company. It began operation in September 1985, and has amounted to the largest capacity for electrical supply of 8,212 MW with 7 units since 1997. Niigata prefectural government has been carrying out the environmental radiation and radioactivity monitoring around the station site since 1983, and reporting the results of artificial contribution exposed to the public.

During the monitoring, some typical artificial radionuclides found are ^{90}Sr , ^{137}Cs , Pu isotopes, etc., considered entirely to be origin of fallout debris from

past atomic bomb tests in the atmosphere. Since recent concentrations are on the decline or nearly constant at very low levels, detectable data must reflect the features related to geological, topographical and meteorological information of each sampling point. However, it is necessary to investigate their accurate distributions and behaviors as backgrounds, not only for preparing for undesirable happening of nuclear disaster, but for accounting for the natural radioactive phenomena to the residents and providing a sense of security. This is just the obligation of a local government. For this purpose, continuous development of new device and analysis, which should be worked more rapidly and sensitively, must be confirmed

2. ^{137}Cs distribution around Kashiwazaki-Kariwa area

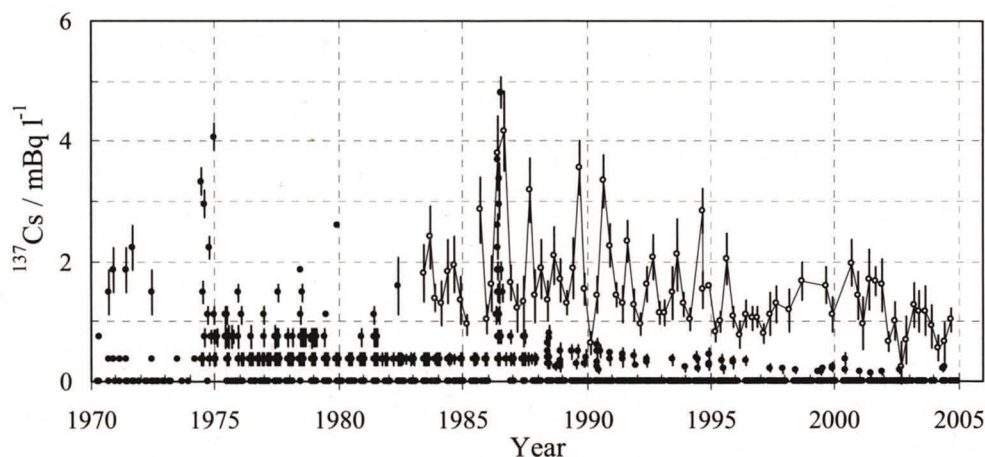


Fig. 1 Changes of ^{137}Cs concentrations in tap waters in Japan.

closed circles: results of annual researches at 47 prefectures since 1970.

open circles: results of radioactivity monitoring (once every three months) at Kashiwazaki city since 1983.

2.1 Introduction

Existence of trace amount ^{137}Cs , which has been often detected in some fresh water samples from Kashiwazaki city, will attract public concern in the field of the environmental radiation monitoring. Figure 1 shows changes of ^{137}Cs concentrations in tap waters in Japan since 1970. This radionuclide had been detected almost everywhere in Japan within tap waters during 1960's to 1970's, indicating wide pollution of water sources and rivers by fallout debris derived from atomic bomb tests in the atmosphere. However, it has been rarely found in tap waters these days in the order of mBq/l except Kashiwazaki city.

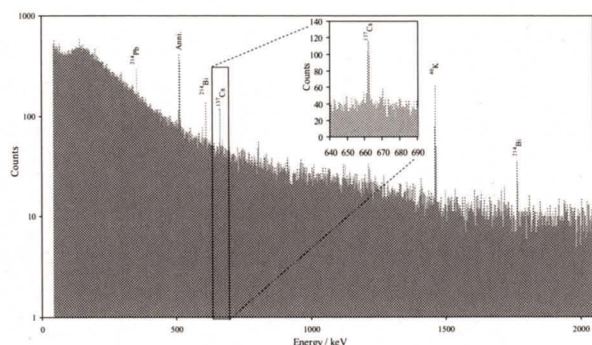


Fig. 2 Typical gamma ray spectrum of a tap water sample in Kashiwazaki city.

^{137}Cs concentration was estimated as 1.10 ± 0.107 mBq/l

Even after 1987, samples from Kashiwazaki alone have showed ^{137}Cs results of about 1.0 to 1.5 mBq/l (sample amount: 20 l). Emphasis should be placed on

the fact that no other artificial radionuclides have been detected (except ^{134}Cs just after the Chernobyl accident) in the samples as shown in Figure 2; it cannot be from the nuclear power station.

Although the evaluated internal dose for drinking ^{137}Cs of 1 mBq/l is very low, of course, as calculated below,

$$0.001 \text{ (Bq/l)} \times 2.2 \text{ (l/d)} \times 365 \text{ (d/a)} \times 1.4 \times 10^{-5} \text{ (mSv/Bq)} \\ = 1.1 \times 10^{-5} \text{ mSv/a,}$$

it is not clear why ^{137}Cs has still been detected from the district only, and it needs to be investigated.

In this study, ^{137}Cs activity in several fresh waters, dam sediments and soils were examined by gamma ray spectrometry as well as beta counting. Some other fallout radionuclides such as ^{90}Sr and Pu isotopes were also analyzed. Their concentrations were discussed with the results of some stable elements. Much attention was paid on the distribution of fallout radionuclides around Mt. Yoneyama (an altitude of 993m) which stands only 5 km away from the southwest coast of Kashiwazaki city.

(In this article, result of ^{137}Cs is mainly discussed)

2.2 Experimental

About 20 to 60 liters of fresh waters, including rivers, dam lakes and springs, were collected around Kashiwazaki-Kariwa area and 17 dam sites in Niigata during period of Nov. 2003 to Sep. 2006 (mainly on

fine days during summer seasons), as illustrated in Figures 3 and 4.

Samplings at dams were performed generally near the center of each lake on a boat, together with vertical profile measurements of water temperature. Monthly tap water in Kashiwazaki city (at the

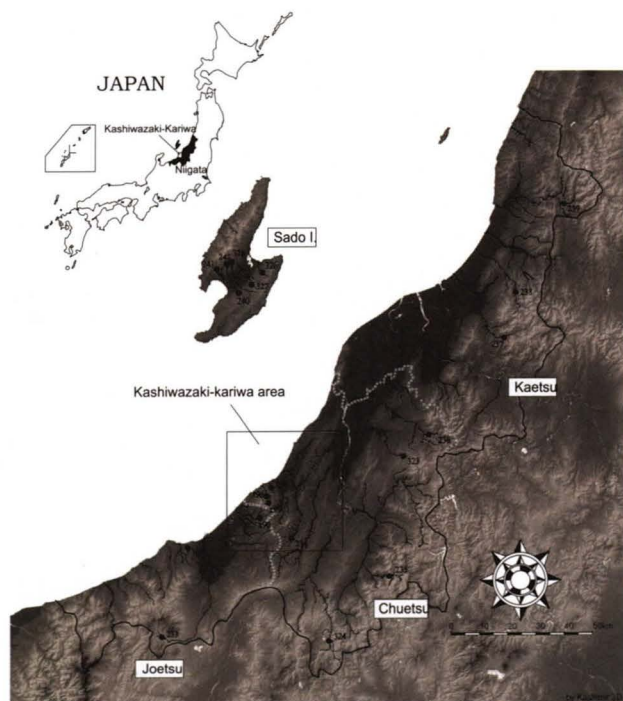


Figure 3 Sampling points of dam lake waters and sediments in Niigata.

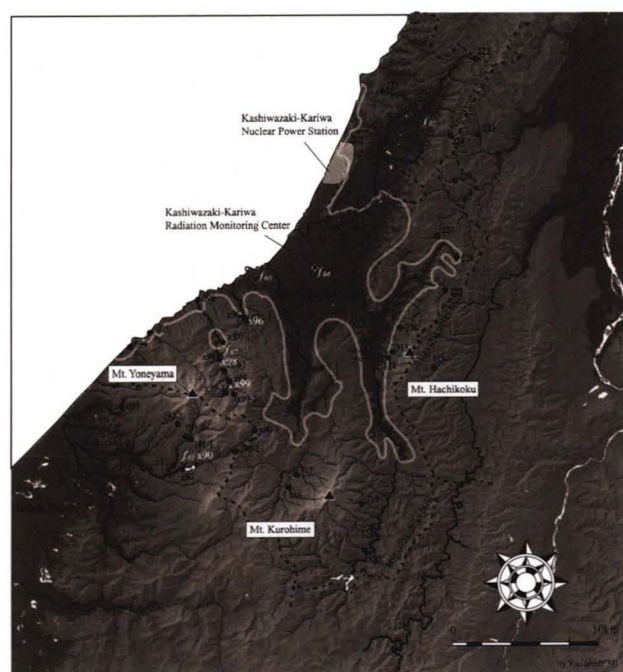


Figure 4 Sampling points in Kashiwazaki-Kariwa.

- administrative districts of Kashiwazaki city and Kariwa village
- water supply districts for Kashiwazaki city (1998)
- Sampling points for fresh waters in this study

Kashiwazaki- Kariwa Radiation Monitoring Center) was also sampled at the end of every month from June 2002. Water samples were filtered by passing through a 0.45 μm membrane filter (exception of tap waters) and evaporated gently to dryness after adding HNO_3 to be 0.1 % v/v.

Lake sediments were collected at the same day to the water sampling using an Ekman-Birge type bottom sampler. Most of sampling was conducted near the center of those dam lakes, where the water depths were the deepest (about 7 to 60 m). The mud and sand samples were dried at 110 $^{\circ}\text{C}$ for 48 h, sieved through a 2.0 mm screen.

^{137}Cs measurement of evaporated residue was performed by gamma ray spectrometry with high pure Ge (HPGe) detectors (relative efficiencies of about 45 %). The residue part on the filtered papers (for water samples) was also subjected to the determination of particulate radioisotopes. The detection limit for ^{137}Cs in the water was about 0.22 mBq/l with a measurement time of 240,000 sec.

About 50 to 100 g dried fine soils were also measured with the HPGe. The measurement time was 80,000 sec. The detection limit for ^{137}Cs was estimated to be about 0.11 Bq/dry-kg.

After the HPGe measurement, some evaporated residues of fresh waters and dried fine soils were engaged to the radiochemical procedures for the determination of ^{90}Sr . The residue was decomposed with aqua regia after addition of strontium carrier solution. Soils were heated at 450 $^{\circ}\text{C}$ for decomposition of organic matters, and then strontium carrier and hydrochloric acid solution were added to the sample and was heated to dissolve. Strontium fraction was precipitated at first as carbonate, then as oxalate, and followed by further purification using a cation-exchange resin. ^{90}Sr were counted using a low background gas-flow type GM counter (Aloka, LBC-471Q) with 28,800 to 36,000 sec for ^{90}Sr . Detection limit of water and soil were about 0.10 mBq/l and 0.025 Bq/dry-kg, respectively.

Some stable elements were also analyzed by an ICP-MS and an ICP-OES.

2.3 Results and discussions

2.3.1 ^{137}Cs in tap and dam waters

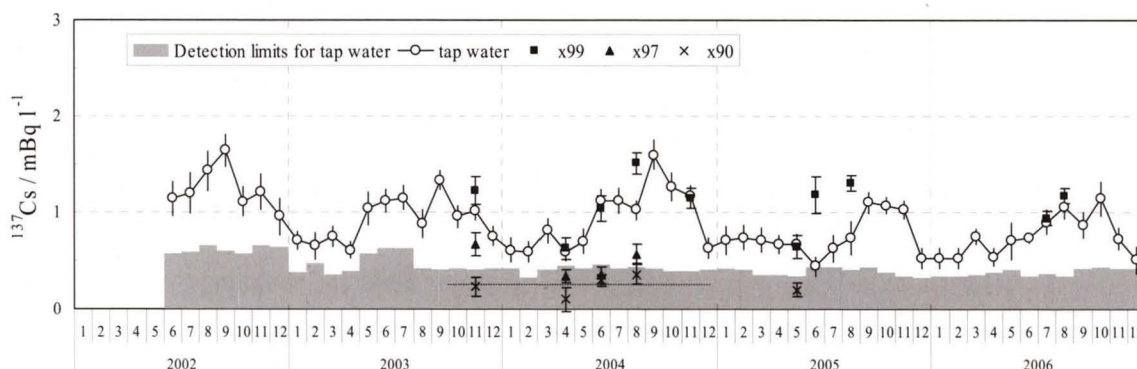


Fig. 5 Trend of monthly ^{137}Cs in tap water in Kashiwazaki city.

Sample amount: 40 l for tap water and 60 l for fresh waters.

Measurement: γ ray spectrometry of evaporated water (160,000 to 240,000 sec)

Distribution around the dam lakes

Figure 5 shows ^{137}Cs contents in monthly tap water supplied from the dams illustrated in Fig. 4 as No. x99 and x98 (as a mix) along with the data of x99's own, x97 and x90. The tap water was collected at Kashiwazaki-Kariwa Radiation Monitoring Center, as x00 in Fig. 4.

Since ^{137}Cs in the filtered residues were scarcely detectable (ND), most of discussion in this study is connected to the isotope in filtered solution. Other artificial radionuclides were not found in HPGc measurements.

Activity fluctuations for the dam lake x99, in the range of 1.6 ± 0.15 to 0.44 ± 0.10 mBq/l, are in good agreement with that of tap water. The way of water purification for dams of x99 and x98 was rapid sand filtration using polyaluminum chloride, sodium hypochlorite and sodium carbonate, which removed particulate matters in the raw water. Coincidence of ^{137}Cs contents between the dam lake and the tap water suggests this isotope was in a chemical form which was not influenced by the water purification procedure. Thus, ^{137}Cs in the water were thought to be nearly-free ion.

Figure 6 shows ^{137}Cs distribution in fresh waters around the dams of x99, x98 and x96. Relatively higher results over 1 mBq/l were observed around dams of x99 and x98, especially in the influent streams to the x99. This means water already contained ^{137}Cs before the streams reach down to the dam lake, suggesting its dissolution from the soil

surface on the upper reaches. Those streams flow from the east side of Mt. Yoneyama, where fallout radionuclide would be abundant as discussed below with the results in Figure 9.

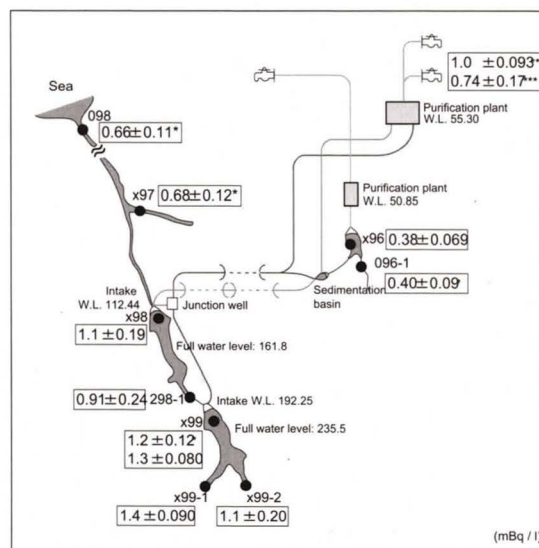


Fig. 6 ^{137}Cs distribution around water facilities near Mt. Yoneyama.

lake and river water : 20 to 60 l

* ... sampled on 27 November 2003

(no asterisk) ... sampled on 1 September 2005

tap water : 40 l

(collected at Kashiwazaki-Kariwa radiation monitoring center)

** ... sampled on 28 November 2003

*** ... sampled on 30 August

Exceptional features

It also needs to discuss about a few exceptions of ^{137}Cs relation between the dam and the tap water. As shown in Figure 5, two types of different were found; the first was the in case of June 2005, and the second

was Augusts 2005 and 2006, in which both ^{137}Cs contents were relatively higher in the dam lake of x99 but not in the tap water. Below discussions deal with the relation of tap and dam water in x99, which stands the uppermost zone of the river basin having larger catchment area (5.92 km^2) and effective water storage ($3,550 \times 10^3\text{ m}^3$) than that of x98 (3.26 km^2 and $1,300 \times 10^3\text{ m}^3$, respectively).

(In this article, only the first case is discussed)

Influences of heavy rainfall

As in the case of June 2005, it will be explained by the difference of sampling date at the dam (30 June) and the faucet (29 June), and should be discussed with the effects of a heavy rain which hit wide area of northern part of Japan during 27 to 28 June, 2005, from viewpoints of the isotopic relations of ^{137}Cs and ^{133}Cs in the dam water.

Figure 7 shows the trends of ^{137}Cs , ^{133}Cs and their ratio ($^{137}\text{Cs}/^{133}\text{Cs}$ as mBq/mg) in the dam of x99. The ratio was mostly constant except the sample of June 2005, indicating similar behaviors of both nuclides in the normal conditions. However, the ratio was increased anomalously on 30 June, which corresponded to about 4.6 times as large as the normal.

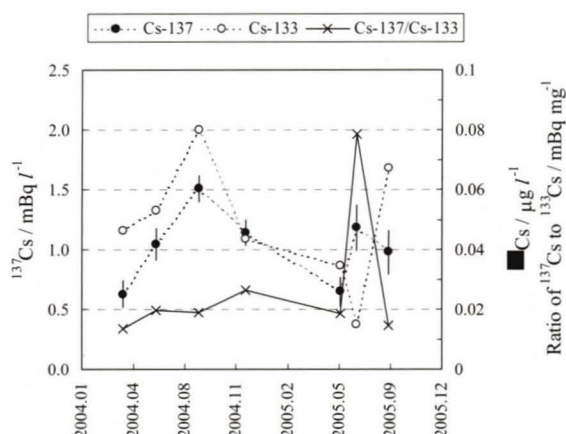


Fig. 7 Seasonal changes of ^{137}Cs , ^{133}Cs and their ratios in dam lake x99.

In the period of 26 to 28 June, there recognized much amount of precipitation of 519 mm at the dam, which brought the inflow increasing of 16.2 m/s on 28 June. Trend of stable ^{133}Cs concentration gave minimum value of $0.015\text{ }\mu\text{g/l}$ on 30 June as shown in

Figure 7, which would be attributable to diluting effect by the increased water. Interestingly, the behavior of ^{137}Cs appeared contrary to the trend of stable isotope; the artificial isotope, ^{137}Cs increased after the heavy rainfall, raising their ratio of $^{137}\text{Cs}/^{133}\text{Cs}$. It reveals certainly that the distributions of two kinds of isotopes in the soil are different. Since fallout-derived artificial radionuclides are thought to be abundant in shallow soil, when a torrential rain carries away surface soil to downstream, much ^{137}Cs would be released in water whereas the dilution of stable ^{133}Cs would be induced. This carrying away effect also caused increasing of particulate ^{137}Cs of 1.1 mBq/l , which was not detectable in all the other sampling terms.

On the other hand, ^{137}Cs in tap water sample showed no increase on 29 June, 2005; it took about 5 to 7 days before the dam water reached to the faucet, so that the water would not be affected by the heavy rainfall.

The relation of ^{90}Sr and stable Sr took the same tendency to Cesium as shown in Figure 8, though increment of radionuclide/stable ratio after the heavy rainfall was relatively smaller (amounted to about 1.9 times as large as fine days) than that of Cs (about 4.6 times).

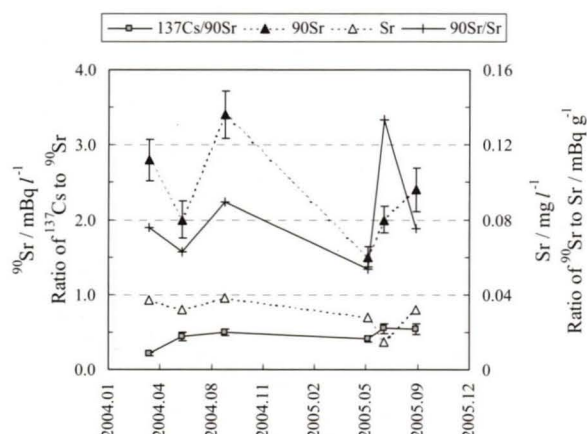


Fig. 8 Seasonal changes of ^{90}Sr , Sr, ratios of ^{90}Sr to Sr and ^{137}Cs to ^{90}Sr in dam lake x99.

2.3.2 ^{137}Cs distribution around Kashiwazaki-Kariwa area

^{137}Cs in fresh waters

Figure 9 illustrates distribution of ^{137}Cs in rivers on

the map, together with some geological information in Kashiwazaki-Kariwa area. The concentration is expressed in several shades of blue, regarding an average value of the catchment area on the upper reaches from sampling point.

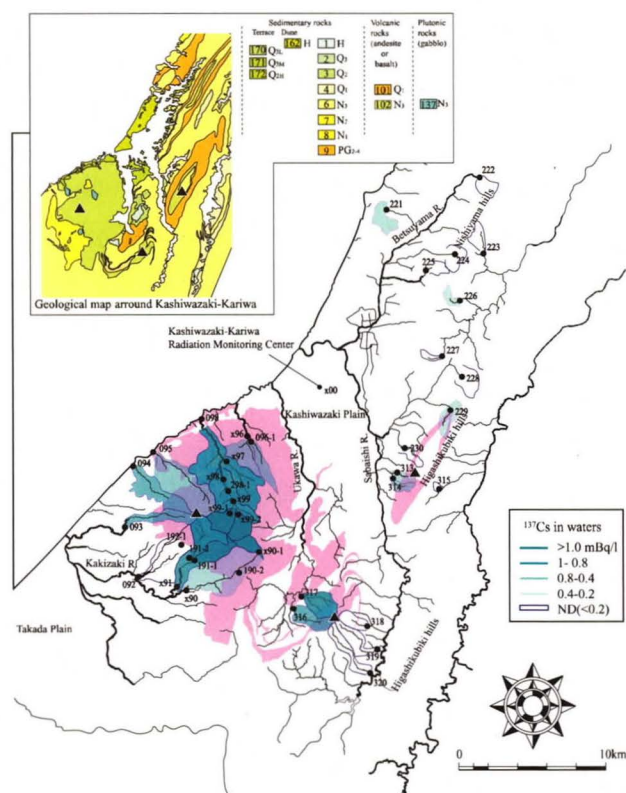


Fig. 9 Distribution of ^{137}Cs in several fresh waters in Kashiwazaki-Kariwa area.

Andesitic rock distribution
● Sampling points for fresh waters in this study
Above geological map is based on the data from "Seamless Digital Geological Map of Japan (1:200,000)", an web site of "<http://riodb02.ibase.aist.go.jp/db084/index.html>"

Figure 9 indicates clearly that the ^{137}Cs in those rivers close to Mt. Yoneyama gave higher concentration near 1 mBq/l, particularly in the east side of the mountain. Relatively higher values were also admitted in west sides of Mt. Kurohime (No. 316 and 317) and Mt. Hachikoku (No. 313 and 314) in the range of 0.36 ± 0.071 to 0.81 ± 0.075 mBq/l, though ^{137}Cs in their east sides (No. 230, 315, 318, 319 and 320) were almost ND. Those rivers flow from some hills surrounding the Kashiwazaki Plain (No. 221 to 229) displayed hardly detectable peak of 662 keV. Two spring water in the Yoneyama massif also gave the results of ND.

It is interesting that distribution pattern of ^{137}Cs is

seem to be overlapped to that of andesitic rocks on the geological map, as shown in Fig. 9, indicating that the radionuclide was dissolved in relatively higher concentration in those rivers run through the soil of andesite-origin. This would be the reason why ^{137}Cs was obtained distinctly in the west sides of Mt. Kurohime and Mt. Hachikoku.

^{137}Cs Distribution around Mt. Yoneyama

In the case of Mt. Yoneyama, the massif itself forms almost entirely huge mass of andesite volcanic rocks. Relatively higher ^{137}Cs concentrations in rivers from east side of the mountain will be due to the abundance of fallout nuclides in the soil there, which would arise from the geographical and metrological characteristics around the mountain.

Around Mt. Yoneyama, there exist more valleys cut by steep rivers in the east side, bringing on an asymmetric shape from east to west.

It is a frequent feature of mountains in Niigata, which is known as the heaviest snowfall district in the world, having precipitous gorges and cliffs in their east or southeast side (opposite to the sea side) exposed by predominant west wind, so called "monsoon (Siberian wind)". It is also well known that the snow falls in more amount in the east side, and it remains until even early summer, while in the west, the snow melts away easily to the sea in early spring.

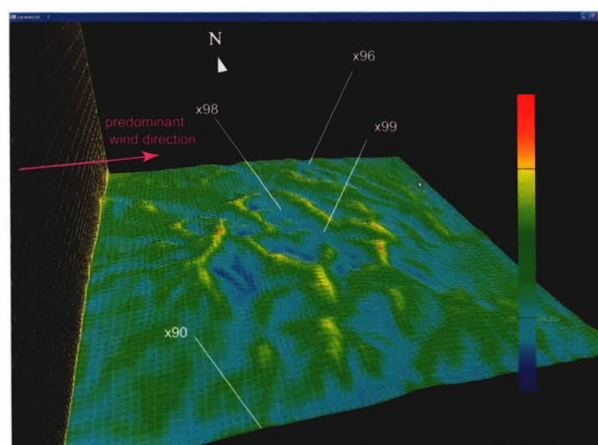


Fig. 10 Distribution of average wind velocity around Mt. Yoneyama.

Above results were culculated by LAWEPS (Low Energy and Industrial Technology Development Organization, NEDO) with multi-step wind simulation with nesting method.

Figure 10 shows distribution of annual wind velocity (as an average) around Mt. Yoneyama, calculated by multi-step wind simulation with nesting method (local area wind energy prediction system, LAWEPS). The simulation shows that the wind blows relative strongly (8 to 9 m/s) across the ridges of the mountain, whereas it drops suddenly at the east-side valleys in which watersheds of x99 and x98 are also located.

Generally, snowfall amount becomes larger where the wind velocity decreases quickly, so that east side of mountain would become snowier in winter season. In the same way, fallout debris including ^{137}Cs may have been gathered and remained in the east side area for relatively longer period, too. Then ^{137}Cs will be transferred to soil and leached out continuously into the water near the ground. This mechanism will be a reasonable interpretation occurred around Mt. Yoneyama, which is not so higher altitude (993 m) but closer to the sea.

Sampling of fallout radionuclides during spring season

To investigate above estimation about topographical effect of Mt. Yoneyama upon the concentration of fallout radionuclides, it needed actual sampling around the mountain.

In the periods of March to May and also in November, Aeolian dust has often flied to Japan from the depth of China. The dust with fine yellow sand was recognized distinctly in the sample of rain and dry fallouts during the season. A noteworthy phenomenon is that those fine particles contained small amount of ^{137}Cs , which has been also detected by gamma ray measurements in the ordinal monitoring.

Sampling of rain and dry fallouts were conducted for 4 times with 1 to 2 week intervals, during March to May 2004 at five points as shown in Figure 4. There found remarkable ^7Be in all samples, indicating the fallout amount at point- f_{02} (close to the dam of x98) was largest in every interval. Although ^{137}Cs was not detected within most of sampling terms due to the poor size of the collecting vessel, those composite samples of 4 time collections showed clear difference between these sampling sites as in Figure 11.

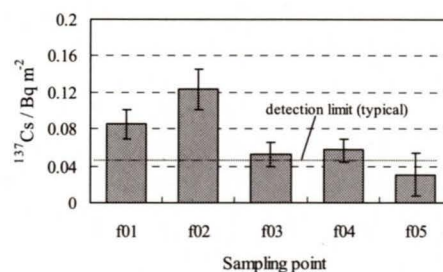


Fig. 11 Results of rain and dry fallouts collected in Kashiwazaki-Kariwa area during spring of 2004.

Sample from point- f_{02} , which is close to dam x98 and to Mt. Yoneyama, gave the highest value of 0.12 Bq/m². This was 2.1 times as large as that collected at Kashiwazaki Plain.

2. 3. 3 ^{137}Cs distribution in Niigata

In Niigata, there are 96 dams in operation and 12 dams in construction and operation in March, 2005. They were constructed generally in mountainous riverhead regions. Thus, it is expected that ^{137}Cs also exists in those watersheds and even in waters as well as those of dam lakes near Mt. Yoneyama. Twenty dam sites including 4 dams close to Mt. Yoneyama were selected to cover main districts in Niigata, such as Kaetsu, Chuetsu, Joetsu and Sado Island as illustrated in Figure 3.

Figure 12 shows the ^{40}K , ^{133}Cs and ^{137}Cs concentrations in the dam sediments and those waters. Their ratios (concentration in the water to that in the sediment) are also presented in Figure 13.

Results of ^{40}K in the sediments indicate regional characterization clearly in Figure 12 (a), providing the following order of concentration; Kaetsu (850 to 980 Bq/dry-kg) > Joetsu (560 and 580 Bq/dry-kg) > Sado I. (370 to 480 Bq/dry-kg) > Chuetsu (260 and 640 Bq/dry-kg, varying with each site) > around Mt. Yoneyama (150 to 280 Bq/dry-kg). These trends probably explained by the difference of local geologic circumstances, in particular, Kaetsu district is well known as abundant in granitic rocks in which potassium feldspars are contained as one of main rock-forming minerals.

However, the ^{40}K concentration in the waters shows just opposite tendency; samples from those dams having K-rich sediments give relatively small amount

of activity in the waters. It is clearly showed in Figure 13 (a), in which the highest ratio of $^{40}\text{K}_{\text{wat.}} / ^{40}\text{K}_{\text{sed.}}$ was revealed from x99 near Mt. Yoneyama, whose ^{40}K in the sediment was next to last as in Fig. 12 (a). Those higher ratios were obtained from 4 dams around Mt. Yoneyama, and other 4 sites having andesitic zones on their upper reaches (No. 240, 323, 326 and 327) as well as 3 dacitic zones (No. 241, 242 and 326), and also No. 234, whose watersheds are made of alternation of sandstone and mudstone (which was the highest of 65 mBq/l).

Another alkali element, stable ^{133}Cs also thought to be showed the regional trends like ^{40}K , as shown in Figure 12 (b) and 13 (b), though 10 samples were not measured. Those dam sites in which andesitic soil is mainly distributed shows relatively higher ratio, $^{133}\text{Cs}_{\text{wat.}} / ^{133}\text{Cs}_{\text{sed.}}$, as in the case of ^{40}K . The highest ratio of 0.031 g/l was presented by the sample of No.

lava and pyroclastic rocks.

For the artificial induced ^{137}Cs contents (Fig. 12 (c) and 13 (c)), observed concentrations in the sediments were ranged 3.9 to 62 Bq/dry-kg. Those values could represent the current average-amounts of fallout ^{137}Cs within those watersheds. The results of 52 and 41 Bq/dry-kg from x99 and x98, which are close to Mt. Yoneyama, were obviously higher values among them. Some others also gave concentrations over 20 Bq/dry-kg. On the other hand, ^{137}Cs in the water did not depend on the sediment-concentration. Typical example was indicated by the sample of 239, in which ^{137}Cs was the highest of 62 ± 0.49 Bq/dry-kg in the sediments while ^{137}Cs in the water was very low at 0.26 ± 0.0023 mBq/l as in consistent with the results of natural alkali cations.

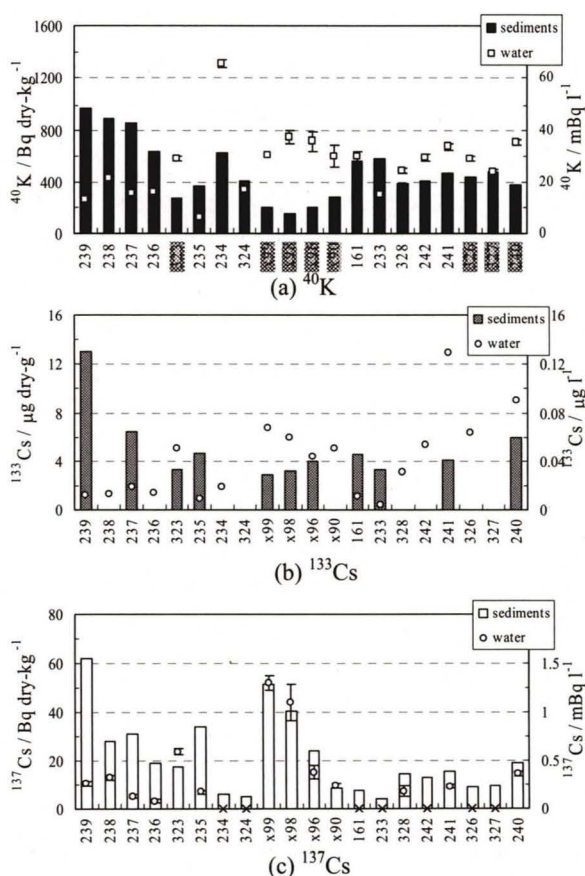


Fig. 12 ^{40}K , ^{133}Cs , ^{137}Cs concentrations in dam waters and sediments.

Hatched numbers mean those catchment areas are andesitic origin.

Lacked data in (b) are not measured.

Crosses in (c) mean ND.

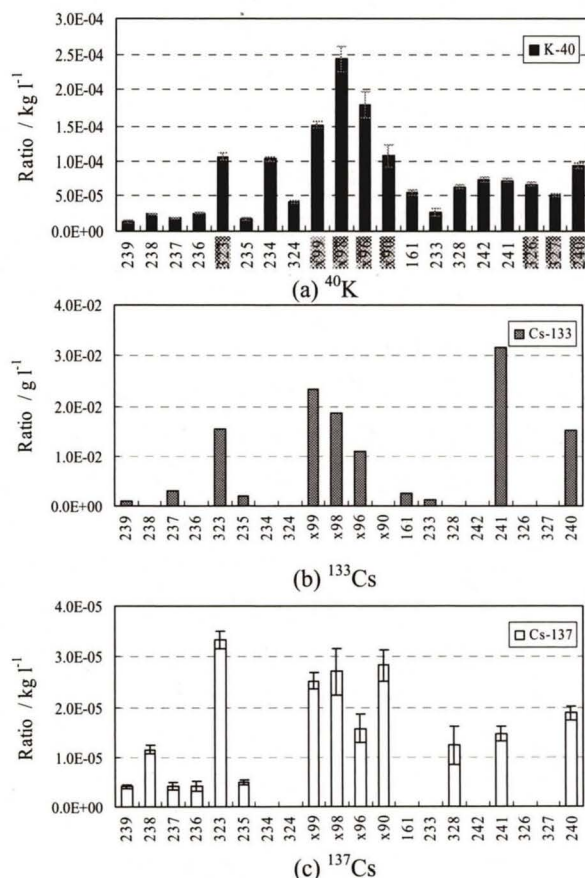


Fig. 13 The ratio of ^{40}K , s-Cs, ^{137}Cs concentrations in the dam waters to that of the sediments.

Hatched numbers mean those catchment areas are andesitic origin.

Lacked data in (b) are not measured.

Crosses in (c) mean ND.

241, whose catchment area is mainly made of dacite

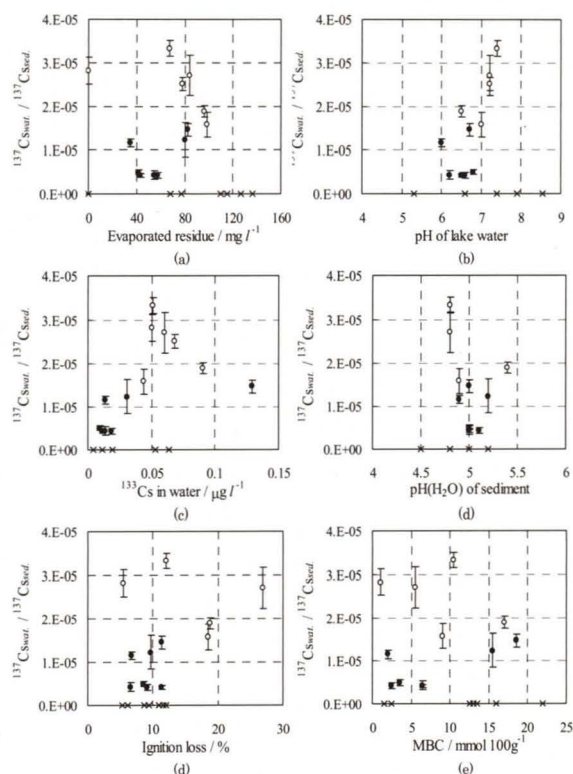


Fig. 14 Relations of $^{137}\text{Cs}_{\text{wat.}} / ^{137}\text{Cs}_{\text{sed.}}$ between several parameters of dam waters and sediments.

Open circles: watersheds of andesitic origin

Closed circles: watersheds of non-andesitic origin

Crosses: ND (water)

Relations of $^{137}\text{Cs}_{\text{wat.}} / ^{137}\text{Cs}_{\text{sed.}}$ between several parameters of dam waters and sediments are presented in Figure 14, in which no strong relation was recognized. Only geological features seemed to be main factor for the largeness of $^{137}\text{Cs}_{\text{wat.}} / ^{137}\text{Cs}_{\text{sed.}}$; the ratio was higher in the site where andesitic rocks dominate.

Figure 15 shows the comparison of typical two sediment samples from No. 239 (granite origin) and x98 (andesite origin), concerning with powder properties. Interestingly, there were not great differences on the specific surface area (SSA), particle size distribution and MBC among them, although there certainly exists the difference of cesium equilibrium between liquid and solid, in which both ^{133}Cs and ^{137}Cs were more abundant in the water of x98, as shown in Figure 12.

It was also confirmed experimentally that the soluble fraction of ^{137}Cs in x98 was larger than that of No.239 as indicated in Figure 16. Approximately 11 % of sediment- ^{137}Cs in the former was extracted with 1 M ammonium acetate, while in the latter, it

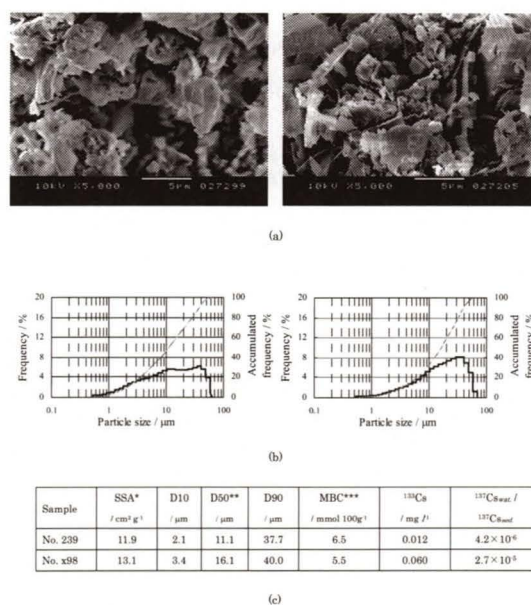


Fig. 15 Comparison of sediment samples from No. 239 and No. x98

(a) SEM images

(b) Frequency distribution of particle size (volume distribution)

(c) powder parameters and Cesium data

* ... Specific Surface Area

** ... median radius calculated from the size distribution

*** ...Methylene Blue Capacity

corresponded to be 6 %.

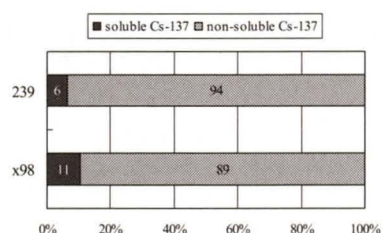


Fig. 16 Comparison of soluble fraction for ^{137}Cs in

sediment samples from No. 239 and No. x98

Reagent: 1M NH_4OAc

Shaking time: 1 h at 25 °C

There also indicated the results of strontium in Figure 17 and 18. As contrary to the alkali metal cations, Sr was in higher concentration in the sediments around Mt. Yoneyama (0.12 to 0.065 mg/dry-g), as well as those of No. 323, 241 and 240 (0.05 to 0.099 mg/dry-g), though the differences were not bigger. Sediment samples from Kaetsu and Joetsu, in which the ^{40}K was rich, showed lower values of Sr (0.022 to 0.036 mg/dry-g).

In the lake water, as in the case of monovalent ion,

relatively higher concentration was obtained from the sites in which sediment Sr was lower. The ^{90}Sr also showed similar behavior to the stable Sr, just like the relation of ^{137}Cs and ^{133}Cs .

Above behaviors of stable elements will be

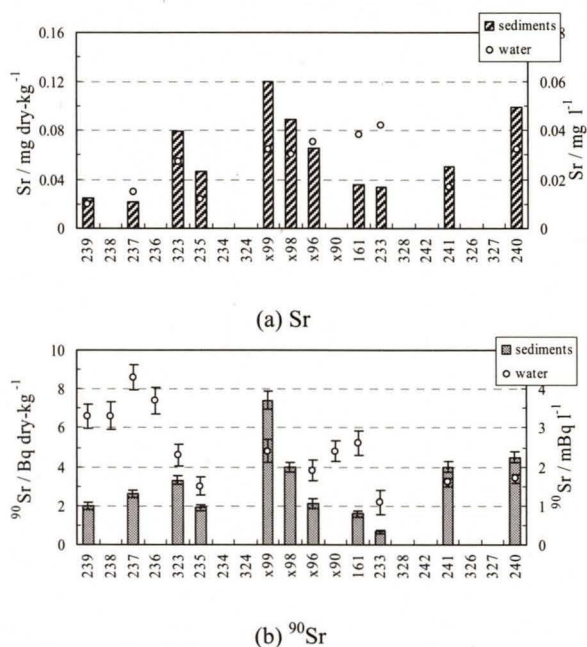


Fig. 17 Sr, and ^{90}Sr concentrations in dam waters and sediments.

Hatched numbers mean those catchment areas are andesitic origin.

Lacked data in (b) are not measured.

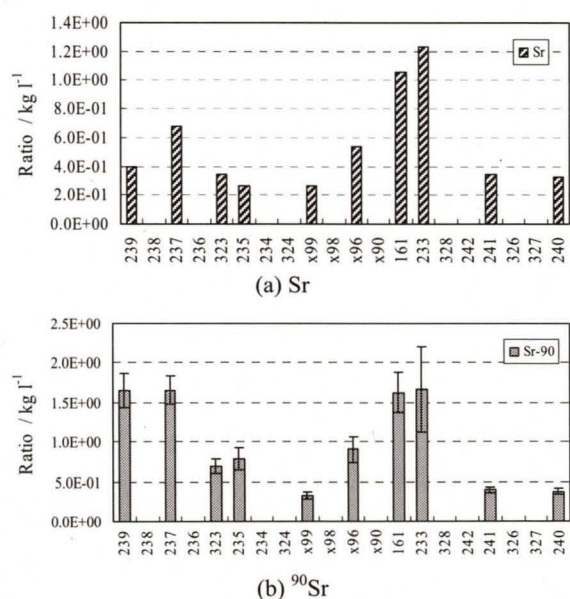


Fig. 18 The ratio of Sr and ^{90}Sr concentrations in the dam waters to that of the sediments.

Hatched numbers mean those catchment areas are andesitic origin.

Lacked data in (b) are not measured.

which exists in a rock or a mineral as a main component, is considered to be hardly released into water. For example, plagioclases contain a lot of alkali earth metals, such as Ca and Sr, in which these cations are well-fitted in the Si-O structures, whereas alkali metals such as K is hardly accepted in the mineral, and easily dissolved to water. Cesium will act as K. In fact, andesite rocks around Mt. Yoneyama are well known to contain such plagioclases, which would be a reason for higher concentration of alkali metal cations in the lake water. As for artificial ^{137}Cs , it showed similar behavior to ^{133}Cs in the normal condition. It is possible that artificial radionuclides are in similar state to their stable isotopes at the surface of soil particles.

2. 4 Conclusions

To trace the origin of dissolved ^{137}Cs in fresh water samples from Kashiwazaki-Kariwa area, radioactive measurements of several fresh waters and sediments were conducted in Niigata, Japan, particularly near Mt. Yoneyama which is located on the southwest coast of this area. A dam lake close to the east side of the mountain showed ^{137}Cs of 1.5 to 0.62 mBq/l in the water, which was higher than those in other dams in Niigata. This is thought to be explained by two reasons; firstly, relatively large amount of fallout radionuclides (about 2 times as large as in plain area) were gathered there by the predominant west wind and resultant asymmetrical shape of the mountain (steeper in the east side). Secondary, there occurs ^{137}Cs transfer from soil to the streams, which will be enhanced by the characteristic geology of the soil originated from volcanic andesite. There abundant plagioclases in the andesitic soils, in which divalent cations such as Sr is preferable as a main component (resulting lower concentration of it in the water), while the Cs will be easily released into the surface water. Artificial radionuclides are thought to be in similar state to the stable isotopes at the surface of soil particles, then they will act together concerning interaction with surrounding water.

connected to a basic property concerned with the interaction of minerals and waters. The element,

3. A rapid TIA (Time Interval Analysis) system for the on-line monitoring of artificial radionuclides in the airborne dusts using a phosfitch type alpha/beta detector

3.1 Introduction

Time interval analysis (TIA) method, based on the analysis for time intervals of some radioactive decaying, has been developed for selective determination of correlated decay processes within millisecond time scales from other random decay events. However, the TIA application to U-series was difficult because their pulses from the correlated events due to ^{214}Bi into ^{214}Po ($T_{1/2} = 164 \mu\text{s}$) are too fast to capture with the ordinary electronic system. On the other hand, recent advancement of PC hard- and software has enabled to measure pulse detection time accurately with microsecond levels, so that calculation of their intervals and result visualization on a PC display could be realized in real-time.

In this study, a new system for TIA method using high speed PC combined with some custom circuits was initially constructed and applied to the continuous monitoring of radioactive airborne dust around a nuclear power station.

3.2 Experimental

3.2.1 TIA (Time Interval Analysis)

Normally, there exist some radionuclides of different half lives in the measurement samples, one of which is expressed as follows,

nuclide $n \rightarrow$ nuclide $m \dots$,

the probability of occurrence for all the radiation during a time of t to $t + dt$ is expressed as follows,

$$\begin{aligned} \sum_n P_n(t) dt &= \sum_n W_n [\exp(-\lambda_n t) - \exp\{-\lambda_n(t + dt)\}] \\ &= \sum_n W_n \{1 - \exp(-\lambda_n dt)\} \exp(-\lambda_n t), \\ &\dots (1) \end{aligned}$$

where W_n is the proportion of the objective decaying to the total events, and λ_n the decay constant of a nuclide n .

In the case of dt is at least 10 times smaller than T_n ($= \ln(2) / \lambda_n$), Eq. (1) is approximated as,

$$\approx \sum_n W_n \lambda_n \exp(-\lambda_n t) dt. \quad \dots (2)$$

When the half-life period of a radionuclide k , expressed as T_k , is much longer than $T_{fix} = 1,000 \times dt$ (T_{fix} is an appropriate upper limit of time intervals),

nuclide $k \rightarrow$ nuclide $l \dots$,
 $T_k \gg T_{fix}$,

decay of k is considered to be constant during T_{fix} , so that the occurring rate of decaying pulse, as $P_k(t)$, appears to be random. This is theoretically expressed from Eq. (2), at the case of λ_i is quite small,

$$\begin{aligned} P_k(t) dt &\approx W_k \lambda_k dt \\ &= r_k dt (\text{constant}). \quad \dots (3) \end{aligned}$$

where r_k is proportion of k (as a random events) to the total events.

On the other hand, if there exist successive decay events whose half life is less than T_{fix} , such as,

nuclide $h \rightarrow$ nuclide $i \rightarrow$ nuclide $j \dots$,
 $dt < T_i < T_{fix}$,

decaying probability of i during a time of t to $t + dt$ is expressed as in the form of Eq. (2),

$$P_i(t) dt = W_i \lambda_i \exp(-\lambda_i t) dt. \quad \dots (4)$$

$P_i(t) dt$ is distinguished from $r_k dt$ in the Eq. (3), called as "correlated events".

Therefore, probability of occurrence for the system which contains one kind of correlated events and other random events is expressed as (from Eq. (3) and (4)),

$$\begin{aligned} \sum_n P_n(t) dt &= W_i \lambda_i \exp(-\lambda_i t) dt + \sum_n r_k dt \\ P(t) dt &= \{W_i \lambda_i \exp(-\lambda_i t) + r\} dt, \end{aligned}$$

where r is proportion of random events to the total events.

For example, if there exist R_n progenies, such as $^{214}\text{Bi} (\beta) \rightarrow ^{214}\text{Po} (\alpha) \rightarrow ^{210}\text{Po}$ in the U-series, since the half life of derived correlated events of $\beta \rightarrow \alpha$ is very short (164 μs), other radiation pulses are generally regarded as random events with dt of 1 μs . Then, only the time interval component of these correlated events selectively emerges on the TIA spectrum as an exponential distribution.

The number of the correlated events is expressed as (when the detection efficiency is 100 %),

$$\begin{aligned} NP(t) dt &= N \{ W_i \lambda_i \exp(-\lambda_i t) + r \} dt \\ &= K_1 \exp(-\lambda_i t) + K_0, \quad \dots(5) \end{aligned}$$

where N is total count number and K_0 and K_1 are experimentally determined coefficients.

The total counts of correlation event component, corresponding to $K_1/\lambda_i = N W_i$ in the Eq. (5) in which K_1 is also experimentally determined by the fitting of the TIA spectrum to the equation, give the radioactivity or counting rate of the decay series.

3. 2. 2 A rapid measurement system

A circuit diagram of original measurement system for TIA is shown in Figure 19.

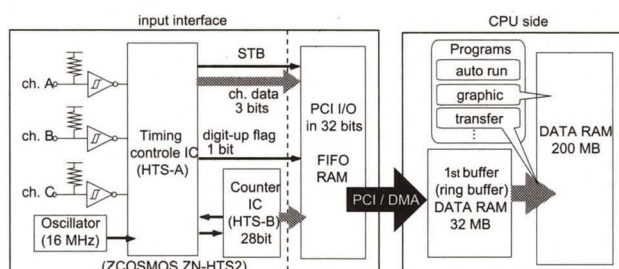


Fig. 19 Circuit diagram of new TIA measurement system.

It had two main parts; an input interface part and DATA RAM part in CPU side. The former consists of a high-speed time counting board (Zcosmos, ZN-HTS2) with three input channels, timing-control IC (Zcosmos, HTS-A) and counting IC (Zcosmos, HTS-B), and PCI/IO unit with FIFO RAM. It could analyze individually three kinds of TTL pulses (from A-, B-, and C-channels). In addition, time intervals

between pulses from different two channels and their sum (A to B, B to A and A+B) were also obtainable simultaneously. Two custom IC provided excellent performance as a time counter with 1 μsecond level. The data were stored at a FIFO RAM and transferred immediately into a ring buffer of 32 MB by PCI-DMA of 33 MHz. In the CPU side, both data transfer program into a main RAM of 200 MB (corresponding to the 5×10^7 pulses) and graphic processing for TIA spectrum could run apparently at the same time.

3. 2. 3 Sampling of airborne dust and detection of radioactivities

A phosfitch type α/β detector (dual phosphor consisting of ZnS (Ag) layer + thin plastic scintillator, 50 mm in a diameter) was employed for the continuous monitoring of radioactive dust in air around the Kashiwazaki-Kariwa nuclear power station, Niigata, Japan.

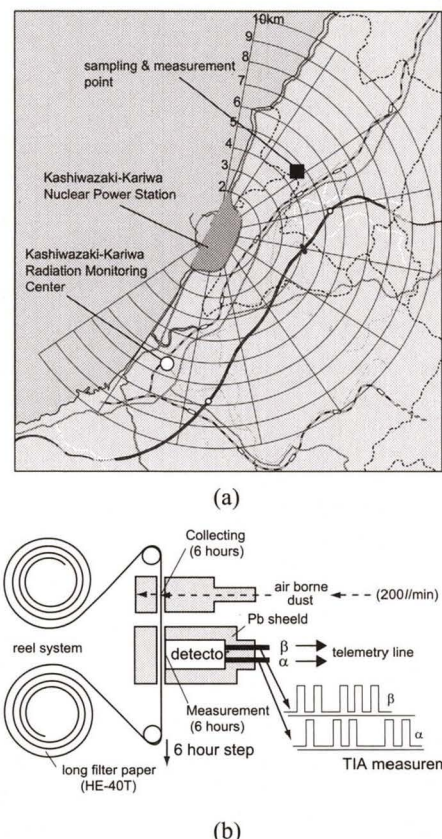


Fig. 20 Sampling point and measuring apparatus for the airborne dust.

- (a) Sampling and measurement point
- (b) Schematic arrangement of continuous sampling and measurement

Figure 20 shows the sampling point and measuring

apparatus. This system was installed in a monitoring post which was located 6.7 km away from the nuclear power station. Airborne dust was collected on a long filter paper (glass-cellulose fiber, Advantec, HE-40T) by an air evacuating sampler with about 200 l/min. After 6 hour sampling, the collected position was moved to a front of phosfitch type detector by a reel system, where α and β rays were measured separately for 6 hours. Those α and β detected pulses were sent to a main monitoring station as TTL type signals (pulse width: 1 microsecond) by a telemetry system. Discrimination of α/β was conducted by use of their different pulse heights, 200 mV for α rays and 50 mV for β rays. The counting efficiency for α rays was about 27 % and 29 % for β , using ^{241}Am and U_3O_8 source, respectively.

Generally, observed radioactivities are thought to be dominated by ^{222}Rn progenies. One problem of this measurement is that the obtained data give only total activities in the collected sample, and lack in information about the kind of nuclides. It is hard to judge whether those counts were derived from artificial or natural origin, in particular just after sampling. Therefore, it is necessary to establish a new method for the real-time monitoring of specific radionuclide in the collected airborne dust.

TIA is induced for this purpose, in which the ratio of ^{222}Rn progenies to the total β activity is expected to be measured, and served as an indicator for the occurrence of artificial radioactivities.

3.3 Results and discussion

3.3.1 Pulse generator tests

Figure 21 demonstrates a result of TIA measurement by the present system using a commercially available pulse generator (Aloka, MXG-111) with 0.5 MHz. This graph confirms that the system could count pulse incident time of 1 microsecond level and calculate their intervals precisely.

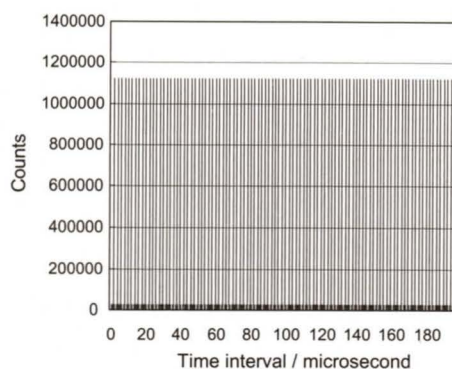


Fig. 21 A TIA spectrum of regular pulse input using a pulse generator.

Pulse frequency: 0.5 MHz

(corresponding to time intervals of 2 μs)

Pulse width: 0.5 μs Measurement time: 1 sec

Analyzed time interval: 1 to 1000 μs

3.3.2 Measurements of air borne dust

Figure 22 illustrates a typical TIA spectrum for the correlated events of ^{214}Bi (β) to ^{210}Po (α ; $T_{1/2}=164$ μs) from the dust sampled on the filter paper for 6 hours. Measurement time was 1 hour. There existed no contribution from artificial radioactivity. It is obvious that the system satisfactorily catch the decay events with very short lives of 1 μs -scale.

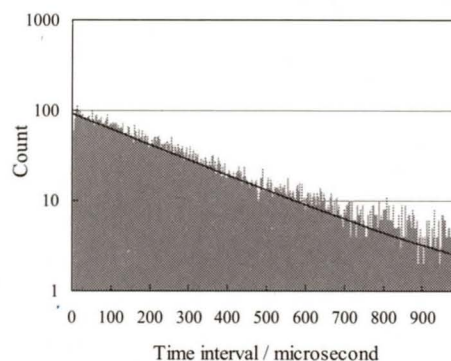


Fig. 22 Typical TIA spectrum due to β - α correlated events from an air borne dust sampled on the filter paper.

A curved line indicates a fitting of $NP(t)dt = K_1 \exp(-\lambda_1 t) + K_0$, from which $T_{1/2}$ is estimated as 170 μs .

Sampling time: 6 hours Measurement time: 1 hour

Analyzed time interval: 1 to 1000 μs

In the present case of α/β separable counting for TIA, the Eq. (5) is modified into the following equation;

$$\begin{aligned} N_{\beta}^{-1} e_{\beta}^{-1} P(t) dt &= N_{\beta}^{-1} e_{\beta}^{-1} \{W_i \lambda_i \exp(-\lambda_i t) + r_{\beta\alpha}\} dt \\ &= K_1^{-1} e_{\beta}^{-1} e_{\alpha}^{-1} \exp(-\lambda_i t) + K_0^{-1}, \end{aligned}$$

where N_β' is apparent total count number for β rays, e_α and e_β are detection efficiencies for α and β rays, W_i is a proportional constant of ^{214}Bi (^{210}Po) to the total β activities, λ_i is the decay constant for ^{210}Po , $r_{\beta\alpha}$ is the background or random event rate of β to α successive decaying, and K_0' and K_1' are experimentally obtainable coefficients, respectively.

In this case, because $r_{\beta\alpha}$ does not contain random event components from α to α , α to β and β to β decaying, $r_{\beta\alpha}$ is less than r in Eq. (5), suggesting lower background for the measurement. As compared with $r_{\beta\alpha}$ in the α/β separable counting and r in the $\alpha+\beta$ counting, the former was amounted to be 23 % of the latter.

Figure 23 shows continuous monitoring of α , β and β - α correlated events activities for the airborne dust, as well as W_i , the ratio of correlated events to the total β activities. TIA could count ^{214}Bi (^{210}Po) selectively even in the first 10 minutes after 6 hour sampling. They were amounted to 0.11 ± 0.01 to 2.3 ± 0.04 Bq/m³ in the present atmospheric stabilities. In addition, W_i monitoring indicated that the proportions of ^{214}Bi (^{210}Po) were almost constant at first 2 hours (about 0.60 to 0.73) through the measurement, although those total activities of α and β were fluctuated in the order of ten. W_i also depended on the time elapsed after sampling, decreasing swiftly after 3 hours.

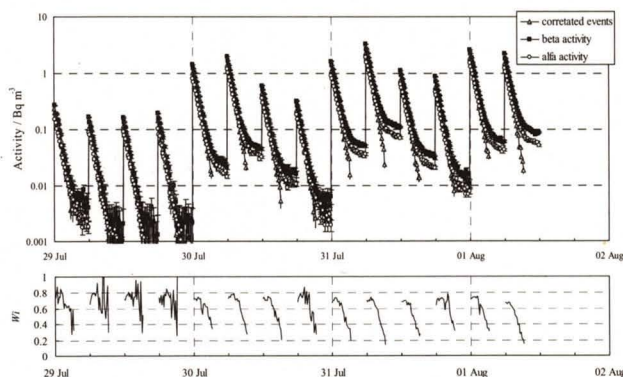


Fig. 23 Results of continuous monitoring for alpha, beta and correlated events.

Sampling time : 6 hours. Measurement time: 10 min.

If there exists artificial β radioactivity, apparent W_i' is described as

$$W_i' = \frac{N_\beta' e_\beta^{-1}}{N_\beta' e_\beta^{-1} + b_x} W_i, \quad \dots (7)$$

where b_x is the added artificial β radioactivity. As a continuous monitoring indicator, $W_i'^{-1}$ is suitable for judgment of any contribution due to artificial β nuclides. If $W_i'^{-1}$ becomes three times larger than those of ordinal variations,

$$W_i'^{-1} > W_i'^{-1} + 3 d(W_i'^{-1}),$$

where $d(W_i'^{-1})$ is counting errors of $W_i'^{-1}$, b_x is expected to be detected. Therefore, Lower detection limit for b_x by present TIA measurement was given by

Lower detection limit for b_x

$$= 3 d(W_i'^{-1}) \times (\text{obtained correlated event activity}). \quad \dots (8)$$

It was calculated for those data indicated in Figure 23, which was clearly depended on the Rn concentration of the sampling terms, varying in the range of 0.17 to 0.029 Bq/m³ at the first 10 minutes measurement. They were also decreased with time elapsed after sampling, due to decreasing of Rn progenies.

For the precise measurement of radon progenies collected on the filter paper, some experimental conditions are needed to be identified; e.g. sampling efficiencies on the filter and emerging efficiencies from the deposited surface based on self-absorption. Effects by different energies of radiation should also be taken into account. Though it is hard to examine all of these conditions, relative variations of $W_i'^{-1}$ in this study are sufficiently useful for surveying artificial radioactivity.

Figure 24 shows a result of 10 minute cycle of sampling and measurements, which corresponds to the emergency monitoring mode for the airborne dust. Sampled air volume was about 1.9 m³ / 10 minutes. TIA spectra from the correlated events of ^{214}Bi (β) $\rightarrow$ ^{214}Po (α) could be also extracted from such a low amount of sampling, in which the activities were calculated as 1.4 to 3.3 Bq/m³. $W_i'^{-1}$ was ranged at

2.5-1.5, which was not influenced by the change of α -, β - and the correlated events. Detection limit of artificial radioactivity in the present condition was estimated about 0.52 to 0.84 Bq/m³. In the practical detection of accidental addition due to artificial β nuclides, this measurement system could be powerful tool for the estimation of the contribution from artificial radioactivity.

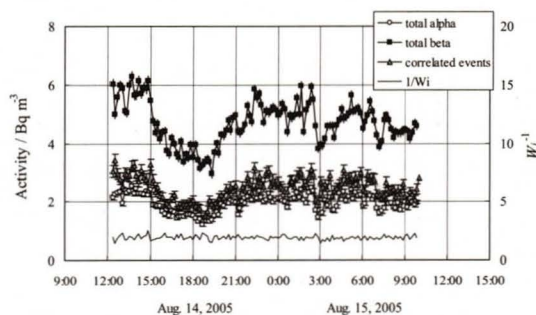


Fig. 24 Results of rapid sampling and counting technique.

Sampling time : 10 min Measurement time: 10 min

3.4 Conclusions

A new system for TIA method using high speed PC with some custom circuits was assembled and successfully applied to the measurements of pulse incident of 1 MHz. Pulse data of 32 bits from three input channels were recorded individually in 1 microsecond-scale and immediately transferred to ring buffer of 32 MB with PCI/DMA.

It could capture the correlated events of ²¹⁴Bi (β) to ²¹⁰Po (α) decaying of $T_{1/2} = 164 \mu\text{s}$, and could serve as real-time monitoring of radioactivities for airborne dust around the nuclear power station, combined with a phosfitch type α/β (ZnS(Ag)+plastic scintillator) detector. The ratio of total β activity to that of correlated events will act as an indicator for the existence of artificial β nuclides. In the case of 6 hour sampling and measurements cycle, detection limit for artificial β radioactivity was estimated about 0.17 Bq/m³ in first 10 minutes. As for the emergency mode of 10 minute sampling and measurement cycle, it corresponded to 0.84 Bq/m³.

Acknowledgements

The author is grateful to Professor T. Hashimoto, for providing useful discussions concerning throughout the study. He would also like to thank Dr.

K. Sawada, Dr. T. Horigome, Dr. T. Imaizumi and Dr. H. Kudo for critical reading of the manuscript. His thanks are due to Mr. Ibe, Mr. Wakai and staff member of Gas and Waterworks Bureau of Kashiwazaki City for the kind help of sampling waters around Mt. Yoneyama, and providing useful information and records about dam operation. He also appreciates Mr. S. Itou of Zcosmos Co., Ltd. for producing a high-speed time counting system for TIA measurement, and also Mr. M. Horiguchi of Kamioka Mining & Smelting Co., Ltd. for the measurements of particle size and specific surface area of sediment samples.

H. Sakaue wishes to thank to the member of the Niigata Prefectural Institute for Environmental Radiation Monitoring for their technical assistance and discussions throughout this study, especially to H. Fujimaki, F. Maruta, K. Kato and Dr. S. Tonouchi for their great support in the experimental work.

The author finally likes to thank most deeply his wife for understanding and support.

References

- Sakaue, H., Maruta, F., Fujimaki, H., Tonouchi, S. and Hashimoto, T. Distribution of Cs-137 around Kashiwazaki-Kariwa area. J. Radioanal. Nucl. Chem. **273**, 123-127 (2007).
- Sakaue, H., Fujimaki, H., Tonouchi, S., Itou, S. and Hashimoto, T. A new TIA (Time Interval Analysis) system for the continuous monitoring of artificial radionuclides on air borne dusts using a phosfitch type alpha/beta detector. J. Radioanal. Nucl. Chem. **274**, 449-454 (2007).

和文要旨

坂上 央存

論文は3章構成とし、第1章では本研究の意義、位置づけについて論じた。原子力発電所周辺の環境放射能モニタリングは、住民の健康と安全を守る観点から地方自治体である県の責務である。近年検出される人工放射能のレベルは非常に低く、下限値未満となることも多くなっている。しかし、万一の災害時に備えるため、また地域住民に対する説明責任を果たし、その安心感と信頼を得るためにも、バックグラウンドの濃度分布及び核種の挙動を詳細に把握しておくことが重要である。また、より迅速かつ高感度の測定・解析手法を常に検討する必要もある。これらの観点から研究を行なった。

第2章では、柏崎刈羽地域における特徴の一つである陸水中の溶存 ^{137}Cs について、その起源、分布、挙動について論じた。これは発電所に由来するものではなく、過去の大気核実験等により生成されたものであり、またごく低い濃度であるため健康上の問題はないことがこれまでの調査からわかっているものの、上水中にも時折検出されることがあり、全国的にも稀有の事象といえるものである。この原因を調査するため、当該河川及び湖上流の集水域が米山（標高993 m、海岸からわずか5 kmにそびえる）山麓にあることに注目し、山麓一帯で環境試料（陸水、土壌、堆積物、降下物）を、また柏崎刈羽を含む県内全域の主なダム湖等で陸水、堆積物等の採取・分析を行った。陸水は採取後0.45 μm のミリポアフィルターでろ過し、蒸発乾固したものを測定対象とした。

調査の結果、蛇口端末の ^{137}Cs 濃度はダム湖水及びその源流水中の濃度とほぼ同じであった（ろ過残渣については豪雨後の採取試料をのぞき、全て検出下限値未満であった）。陸水中の ^{137}Cs は夏季に高く（約1.5 mBq/l）、冬から春季にかけて低い（約0.62 mBq/l）季節変動を示す一方、安定同位体である ^{133}Cs との比は豪雨直後を除きほぼ一定であった。堆積物中の ^{137}Cs 濃度は24~52 Bq/kg 乾であった。湖の流入水（源流）中に ^{137}Cs がすでに存在していることから、集水域の土壌表層を水が流れる間に、 ^{137}Cs が土壌粒子から溶出して供給されているものと考えた。集水域とは米山そのものであり、湖の位置する山麓東側（海と反対側）一帯に、フォールアウト由来の ^{137}Cs が比較的高濃度で存在するものと推定した。これは、フォールアウトを運んで来るのが中国大陸からの偏西風であること、また米山は東西に非対称な形であり、東側に急な谷などが多いことから、気象的・地形的要因によりフォールアウトが溜まり易いものと考えた。このことは、春季の黄砂を含む降下物中の ^{137}Cs 濃度の比較からも裏付けられた。

一方、県内のいくつかのダムには、米山周辺と同程度の ^{137}Cs が堆積物中に存在しているものの、それらの湖水中の濃度は0.16~0.41 mBq/l程度であり、米山周辺ほど高くはなかった。各地のダムの堆積物の粒度分布、比表面積、メチレンブルー吸着量、強熱減量あるいは土壌pH (H_2O)と ^{137}Cs の湖水/堆積物濃度比には相関は認められなかったが、集水域の地質が安山岩由来の土壌であるダムでは ^{137}Cs の湖水/堆積物濃度比が高いことがわかり、地質との関連性が強く示唆された。実際に、米山はそれ自体が安山岩の巨塊である。また同じ柏崎刈羽の八石山及び黒姫山では、安山岩が偏って分布しているが、それらの水系調査では、安山岩地帯を上流域に持つ河川水中で ^{137}Cs 濃度が比較的高かった。興味深いことに、花崗岩由来の土壌を集水域に持つダムでは（安山岩由来のダムと反対に）CsやKといった1価イオン濃度は低く、2価のSr（安定Sr及び ^{90}Sr ）濃度が高かった。このことは、それぞれの土壌に多く含まれる造岩鉱物の構造と、Cs、K、Srなどのイオンの適合性から説明できると考え、人工の ^{137}Cs や ^{90}Sr は土壌粒子表面でそれぞれの安定同位体により希釈され、安定同位体と似たような結合状態で存在していると推察した。

第3章では、原子力発電所からの漏洩人工放射能の早期検出を目的として、空气中ダスト連続モニタリングにおける新型測定・解析装置を開発し、その能力を検証した。このモニタリング手法の大きな問題点として、得られるデータが計数率のみであり、核種情報に乏しい点があげられる。そこで本研究では、人工放射能の検出の妨害核種となる、天然の ^{222}Rn 子孫核種を迅速に定量するために、 ^{214}Bi (β : $T_{1/2}=19.9$ 分) $\rightarrow$ ^{214}Po (α : $T_{1/2}=164$ μ 秒)の連続壊変事象に注目して時間間隔解析(TIA)法を適用した。 μ 秒単位の $\beta \rightarrow \alpha$ という線質の異なる高速連続パルスについて、その検出時間を正確に記録しながら間隔解析を行うことを目的として、高速かつ大容量のメモリと専用ICを組み合わせた新型PCIボードを開発した。また測定しながら結果をグラフ表示させるための解析ソフトウェアも開発した。この新システムは従来のTIA装置では不可能であった1 μ 秒単位のパルスを正確に記録することができ、かつ測定と同時に解析を行うことができた。新システムをダスト連続モニタリングに適用したところ、6時間集塵（約70 m^3 ）直後の10分測定で相関事象を捉えることができ、0.17 Bq/ m^3 の人工放射能検出能力があると見積もることができた。

2 他誌発表

新潟県における放射能調査

霜鳥達雄, 笠原貢, 昆信芳, 山崎興樹, 加藤健二, 藤巻広司, 坂上央存, 殿内重政, 鈴木博之
前年度に引き続き, 平成 17 年度に実施した文部科学省委託の環境放射能水準調査及び放射線監視交付金による原子力発電所周辺の環境放射線監視調査について, 調査結果の概要を報告した。

調査結果は次のとおりである。

1 全ベータ放射能

定時降水等について測定したが, 各試料とも例年と同レベルであった。

2 空間線量率

・ 柏崎刈羽原子力発電所周辺地域及び新潟市において測定したが各地点とも例年と同レベルであった。

3 核種分析

大気浮遊じん等について, ゲルマニウム半導体検出器により機器分析した結果, 海水等に ^{137}Cs を検出した。また, 放射化学分析により ^{90}Sr , ^3H , $^{239+240}\text{Pu}$ を検出した。これら検出された人工放射性核種は, いずれも例年と同レベルであった。

第 48 回環境放射能調査研究成果論文抄録集 (平成 17 年度), 167 (2006)。

新潟県における北朝鮮実験に係る環境放射線監視調査

春日俊信, 殿内重政, 丸田文之

平成 18 年 (2006 年) 10 月 9 日, 北朝鮮が地下核実験を実施した。放射線監視センターは, 県原子力安全対策課の指示により, 放射線 (能) 調査を行った。過去の核実験等に係る放射能影響調査では, 昭和 55 年 (1980 年) までの中国大気圏内核実験及び昭和 61 年 (1986 年) 4 月に起きた旧ソ連チェルノブイル原発事故により人工放射性核種が検出された事例がある。

今回の実験は地下核実験であったが, 県民の健康と安全・安心を守る観点から放射線監視センターの総力をあげてモニタリングを実施した。結果として, 核実験の影響は確認されなかったが, 今回の対応については県民からも高い評価を受けており, 安全・安心に貢献できたと考える。また, モニタリング内容や体制, 県民への情報提供, 他機関との連携等について, 貴重な経験を得ることができた。報告では, 当県の実施したモニタリングの詳細について解説した。

新潟理化学, 32, 33-41 (2007)。

METEX による流跡線解析

山崎興樹

近年, 偏西風によってアジア大陸から輸送されるさまざまな化学物質や粒子状物質の大気環境への影響が指摘されており, 輸送されてくる物質の起源を探索する手法の一つとして後方流跡線解析が注目されている。後方流跡線解析は, 気象データを用いて風の流れをさかのぼり, ある地点に到達した空気が通ってきた流れの経路 (トラジェクトリ) を示すもので, この方法を用いることにより, 汚染物質の濃度上昇時にどの地域からの影響が強いと推測することが可能となる。当所で実施している環境中ラドン濃度調査においても, ラドン濃度の季節変化の主要因は大陸を中心とする遠方起源成分の変動によるものと考えており, 後方流跡線解析は有効なツールの一つとなると考えている。

METEX (METeolorogical Data Explorer) は, (独) 国立環境研究所地球環境研究センター (CGER) の Dr. Jiye Zeng が大型計算機用のシステムを改良して PC 上での高速計算を可能とした気象データの解析・表示システムである。インターネットのウェブサイト上で公開されており, 誰でも無料で使うことができる。流跡線解析システムとしてこれまで広く使われてきた米国海洋大気庁 (NOAA) の HYSPLIT4 モデルに比べ, 手続き等が一切不要で説明も日本語であるため使いやすいシステムとなっている。本稿では, これから METEX を使って流跡線解析をしてみようと思う人のために, プログラ

ムのダウンロードから流跡線の作図までの基本的な使い方を解説した。

新潟理化学, 32, 42-49 (2007).

複数入力に対応した高速パルス時間間隔データ処理およびリアルタイム表示システムの構築とその性能評価

八幡崇, 坂上央存, 伊藤成樹, 橋本哲夫

A new high-speed multiple pulse time data processing and real-time display system combined with time interval analysis (TIA) was developed for counting either β - α or α - α correlated decay-events. The TIA method has been so far limited to selective extraction of successive α - α decay events within the milli-second time scale owing to the use of original electronic hardware. In the present pulse-processing system, three different high-speed $\alpha/\beta(\gamma)$ pulses were fed simultaneously to original 32 bit PCI board (ZN-HTS2) within 1 μ sec. This original PCI board is consisting of a timing-control IC (HTS-A) and 28 bit counting IC (HTS-B). The pulse data were saved to FIFO RAM, followed by the sequential transfer into Main RAM at 1 MHz rate. These data were registered rapidly to construct TIA distribution spectra for determining correlated decay events having short half-life nuclides of micro-second order. By using this new system combined with liquid scintillation counting (LSC) apparatus, both a natural micro-second order β - α correlated decay-events, due to $^{214}\text{Bi} \rightarrow ^{214}\text{Po}(164 \mu\text{sec}) \rightarrow$ (U-series), and a milli-second order α - α correlated decay-events, due to $^{219}\text{Rn} \rightarrow ^{215}\text{Po}(1.78 \text{ msec}) \rightarrow$ (Ac-series), could be identified in a mixture of natural radionuclides and TIA distribution was displayed in real-time. As a result, the present new system has proven to be succeeded for the determinations of micro-second order radioactive decay-events in environmental samples by displaying in real-time.

日本原子力学会和文論文誌, 5, No. 3, 221-228 (2006).

3 学会等発表

冬季雷に伴って発生する空間線量率上昇現象の特徴：

山崎興樹, 坂上央存, 藤巻広司, 霜鳥達雄, 殿内重政

平成 18 年度放射能分析確認調査技術検討会, 東京都千代田区, 東京国際フォーラム, 平成 19 年 3 月 14 日 (平成 18 年度放射能分析確認調査技術検討資料, 235-242, 2007).