Radiochemical Approach to the JCO Criticality Accident in Tokai-mura, 1999 : An Overview of the Radiochemistry Group

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Radiochemical Approach to the JCO Criticality Accident in Tokai-mura, 1999 —An Overview of the Radiochemistry Group—

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JCO criticality accident/Neutron activation/In-situ measurement/Gold/Soil

A few days after the JCO criticality accident in Tokai-mura, a collaborating scientific investigation group was organized to evaluate the environmental impact of the accident. The group consisted of two groups: an environmental research group (radiochemistry group) and a biological research group. This paper overviews the scientific activity of the former group based on 6 sampling campaigns conducted at the JCO campus, Tokai-mura and Naka-machi. Some of the topical results and our remaining tasks concerning the JCO accident are discussed.

INTRODUCTION

At 10:35 on September 30,1999, a criticality accident occurred in the precipitation tank of the uranium conversion building of JCO in Tokai-mura, Japan, located about 120 km north-east of Tokyo. This kind of accident was not expected even by nuclear engineers as well as nuclear and radio-chemists. The total number of fission events during 20 hours was evaluated to be 2.5×10^{18} by analyses of the fission products in the uranium solution in the precipitation tank[1)]. In spite of the best possible medical care, two workers who were severely exposed to fission neutrons and gamma-rays, died 3 and 7 months after the accident.

Within one week after the accident, apart from the actions by a governmental Investigation Committee, two investigation teams were organized to evaluate the environmental impacts of the accident and to assess the radiation effects on the local residents. A brief history and results obtained by the first group are overviewed in this report together with further tasks which remain to be performed in the near future.

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ORGANIZATION OF RESEARCH GROUP

On October 2, an urgent E-mail massage was issued from researchers of the Meteorological Research Institute (MRI) in Tsukuba to the participants of The 1st Summer School on Environmental Radioactivity and Radiation[3] held on July 28~30, 1999 at the Low Level Radioactivity Laboratory (LLRL), Kanazawa University. In this message, they emphasized the importance of an investigation to assess the environmental impact of the JCO criticality accident. This idea originated from experiences in a similar accident that occurred at the former Power and Nuclear Fuel Development Company (PNC) in Tokai-mura in March, 1997. They detected ^{134}Cs and ^{137}Cs derived from an accident in an airborne dust sample collected at Tsukuba, located at about 60 km southwest of PNC[4].

On October 4, the mailing list for the investigation group of the JCO accident was opened by LLRL. On the other hands, the president of the Radiation Research Society of Japan (Prof. M. Sasaki), started negotiation with the Ministry of Education, Science, Sports and Culture of Japan (Monbusho), Nuclear Safety Commission of Japan (NSCJ), Japanese Science and Technology Agency (STA) and Tokai-mura town office to initiate scientific research by organizing a university investigation team. More than 20 proposals were sent to him by October 6. The investigation team was organized in two groups: the first one which was to evaluate the environmental impact of the JCO accident, consisted mainly of radiochemists (environmental research group); the second one which was to assess the radiation effects on residents, consisted of radiation biologists (biological research group).

The environmental research group headed by the author (K. Komura) was composed of 9 principal members from 7 universities and 40 collaborators from 11 universities, 5 national institutes and 2 local governmental institutes. On the other hand, the biological research group headed by Prof. Sasaki consisted of 3 principal members of 3 universities and 3 collaborators from 1 university and 2 national institutes. The proposal was accepted by Monbusho on October 22, 1999. More than 600 E-mail massages were exchanged during 12 months after the JCO accident through mailing lists organized by LLRL and MRI.

SAMPLING CAMPAIGN

A brief history of the environmental research group is summarized in Table 1. Six sampling campaigns at the JCO campus were conducted before February of 2000. Most of our requests were accepted by the JCO, though the sampling time was limited to only 3~4 hours in all cases.

The main object of the survey was the collection of appropriate samples to assess the environmental impact of the accident. The samples collected at the JCO campus were soils, plants, reagents at the laboratory, ceramics, fluorescent lamps etc. Beside sampling, various kinds of investigations were conducted, such as *in-situ* gamma spectrometry using a portable Ge detector, a beta-ray survey along the building walls and the boundary fence, and neutron measurement using ^{3}He counter.

Table 1. Diary of the environmental research group.

Date	Actions of research group
7-Oct-99	1st survey in JCO campus
8-Oct-99	Sampling in Tokai-Mura and Naka-Machi
15-Oct-99	Presentation of preliminary results in annual meeting of Japan Society of Nuclear and Radiochemiical Sciences.
15-Oct-99	Press conference at Tsukuba concerning Monbusho Research Group on the JOC accident
22-Oct-99	Courtesy visit to Monbusho, STA and Atomic Safety Committee
23-Oct-99	2nd survey in JCO campus
24-Oct-99	Sampling in Tokai-mura and Naka-machi
9-Nov-99	3rd survey in JCO campus (sampling and In-situ gamma ray measurement
27-Nov-99	4th survey in JCO campus
28-Nov-99	Sampling in Tokai-mura and Naka-machi
11-12. Dec. 99	1st meeting on JCO research group at Tatsunokuchi
14-Dec-14	Due date of paper for JER
18-Dec-99	Recovoring of gold samples set in JCO
23-Dec-99	Send revised version of paper to Baxter
24-Dec-99	Mr. Ouchi has died
26-Dec-99	Funeral of Mr. Ouchi
22-Jan-00	5th survey in JCO campus
12-Feb-00	6th survey in JCO campus

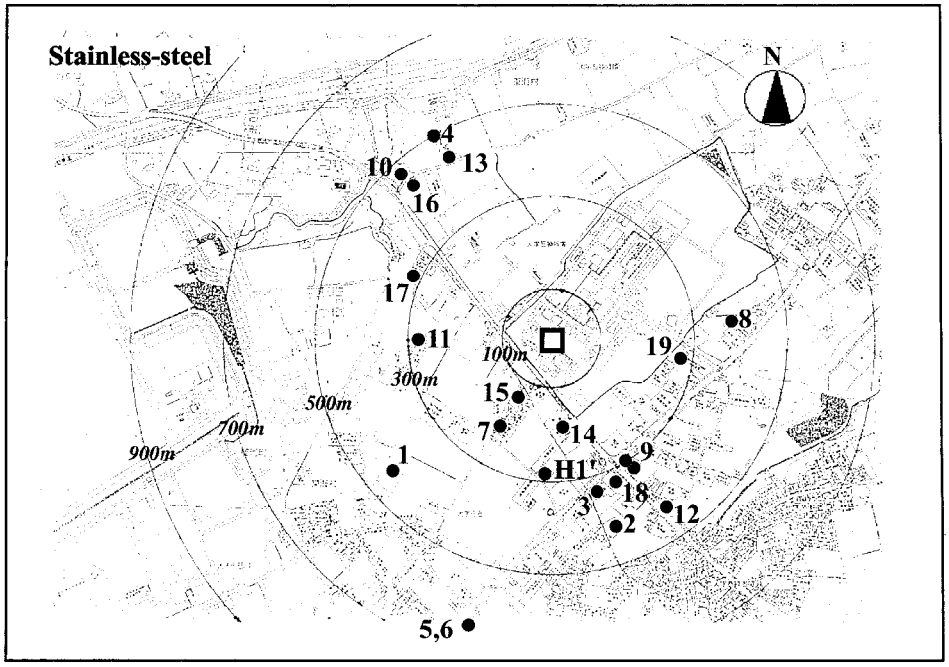

Fig. 1. Sampling points of stainless-steel samples in Tokai-mura and Naka-machi.

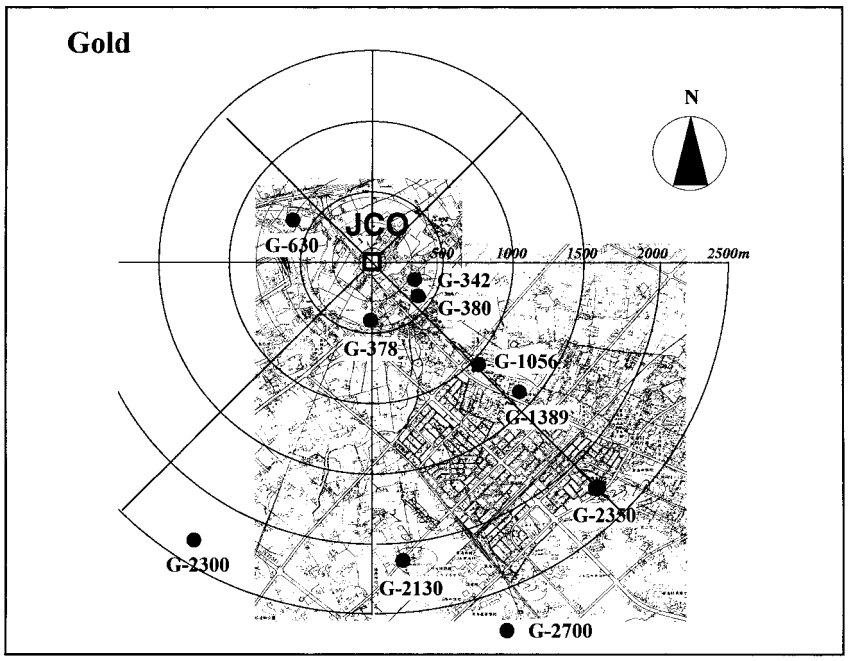

Fig. 2. Sampling points of gold items in Tokai-mura and Naka-machi.

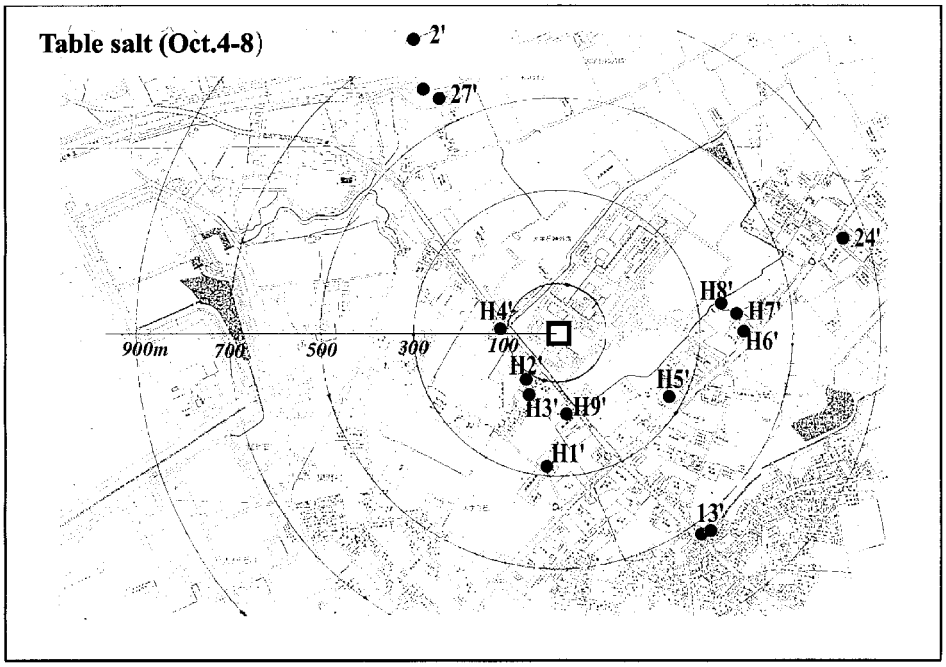

Fig. 3. Sampling points of table salts in Tokai-mura and Naka-machi.

During the 1st and 2nd JCO survey campaigns, a number of environmental samples were collected in Tokai-mura and Naka-machi within 7 km from the JCO site. The total effort in these sampling campaigns exceeded 60 man-days. The samples collected were sugar, table salt, a dry battery, stainless-steel products, coins, gold and silver items, fluorescent lamps, steel and iron products, ceramics, fertilizers, matches etc. Over 400 samples were collected with the courtesy of the JCO, town offices of Tokai-mura and Naka-machi and residents in both towns. Details of the sampling points for stainless-steel, gold items and table salts are illustrated in Figs. 1, 2 and 3. The main research activities conducted by each groups are summarized in Table 2.

Table 2. Task sharing of the environmental radioactivity group.

Purpose of Measurement	Technique or Sample	Research group	Method, Nuclides, Detector
Dose	Dose survey on road	Ibaraki National Coll.	GM-counter
	Dose survey of building wall or boundary fence	Hiroshima Univ.	NaI (Tl) survey meter
	Ceramics	Niigata Univ., Hiroshima U.	TL meas
	Sugar	NIRS	ESR meas
Fission number	In-situ Ge measurement	Tohoku U., Kanazawa U., Tsukuba U., Aich Med. Coll.	FP by portable Ge
Theoretical calculation	Neutron survey and activation data	Kyoto U., Hiroshima U. ERI	Model calculation
Neutron fluence by activation data	Soil	Kanazawa U., Hiroshima U., NIRS	all radioactive nuclides by Ge
	Concrete	Kanazawa U., Tohoku U.	all radioactive nuclides by Ge
	Table salt	Kanazawa U., Tohoku U.	^{32}P by Si b counter
		Tsukuba U.	^{36}Cl by AMS
	Reagent	Kanazawa U., Tohoku U., Tsukuba U., Aich Med. Coll.	all radioactive nuclides by Ge
	Stainless steel	Hiroshima U., Kanazawa U., Aich Med. Coll., HADES (Belgium)	^{51}Cr, ^{58}Co, ^{60}Co, ^{54}Mn by Ge
	Gold items	Kanazawa U., Tohoku U.	^{198}Au by Ge
	Coins	Kanazawa U., (NIRS)	^{65}Zn, ^{110m}Ag, ^{58}Co by Ge
	Fluorescent lamp	Kanazawa U.	^{152}Eu, ^{122}Sb etc. by Ge
	Human hair	Kanazawa U., (NIRS)	^{32}P by Si β counter
	Plant leaves	Ibaraki U., NIRS	^{32}P and FP etc.
Leak of radioactivity	Airborne dust	Meteorol. Res. Inst., NIRS, Natl. Inst. Env. Sci.	Fissio Product by proportional counter or Ge
	Soil	NIRS, Natl. Inst. Env. Sci., Kanazawa U.	^{235}U/^{238}U by ICP-MS
	Plant leaves	Ibaraki U.	^{3}H by LSC

PURPOSE OF MEASUREMENTS MADE BY THE ENVIRONMENTAL RESEARCH GROUP

Immediately after the outbreak of the accident, JCO, JAERI (Japan) and JNC (Japan) conducted intensive monitoring actions to evaluate the neutron and gamma-ray doses. The Ibaraki prefectural Institute of Public Health and Environmental Radioactivity conducted monitoring of vegetables and surface soils. The number of fission events that occurred during 20 hr before the cease of the criticality was evaluated by the JAERI team using a uranium solution collected from the precipitation tank in the Uranium Conversion Building. They also measured the concentration of neutron-induced radionuclides in the stainless-steel net of the cooling tower located 1.8 m from the precipitation tank to evaluate slow and fast-neutron fluence. These measurements were conducted under the supervision of governmental authorities. Their data were adopted in the official report of committee[1,2].

On the other hand, our team, which was composed of members of a number of universities as well as national and prefectural institutes, tried to assess the environmental impact of the accident from NGO-like policy. A variety of techniques were applied, such as *in-situ* Ge spectrometry[6], gamma- and beta-ray measurements of neutron-induced nuclides and fission products in various samples collected in the JCO campus and in Tokai-mura and Naka-machi[7–15], TL measurement[16] etc. The measured data were used for theoretical simulation based on neutron transfer model[12,17,18]. It is also noted that the uranium isotopic ratio in surface soils and plant leaves were measured to assess the possibility of leakage of ^{235}U-enriched uranium[19,20].

TOPICAL RESULTS OBTAINED BY THE ENVIRONMENTAL RESEARCH GROUP

(a) Evaluation of the total fission events by passive in-situ gamma spectrometry using a portable Ge detector[7]

In-situ Ge spectrometry is known to be the most powerful method for the rapid identification of radionuclides released by nuclear accidents. It is unbelievable that no spectrometry attempt was conducted immediately after the accident, though many portable Ge systems are equipped at JAERI, JNC and/or Ibaraki Prefecture for emergency cases of nuclear accidents.

On October 26 (2nd survey of JCO) an *in-situ* measurement by a portable Ge detector was first performed at the JCO campus by university group members. The Ge detector was set at the top of the northwest corner of a shielding wall made of sandbags heaped up around the Uranium Conversion Building. In this measurement they detected intense γ-ray peaks from ^{140}La in radioactive equilibrium with precursor ^{140}Ba (half-life 12.75 d). They invented a very sophisticated method to estimate the total fission events (TFE) using the fission yield of ^{140}Ba, the emission probability of γ-rays from ^{140}La and the detection efficiency of the Ge detector. This technique was applied more comprehensively in a 3rd survey (November 9). Thus-estimated TFE was $(3.1 \pm 0.4) \times 10^{18}$ with 3σ of uncertainty by using γ-rays from ^{95}Zr, ^{103}Ru and ^{140}La-^{140}Ba. This value

is in fairly good agreement with the value obtained by a direct measurement of a uranium solution[1,2].

(b) Measurement of neutron-induced nuclides in gold items, stainless-steel products, and table salts

It seemed most important to evaluate the range-dose relationship. In order to evaluate this, various neutron-induced nuclides were measured at many laboratories of research groups for the samples collected by the 1st and 2nd sampling campaigns.

Among the nuclides measured, ^{198}Au induced by ^{197}Au (n,γ) ^{198}Au was found to be the most useful to assess the range-dose relation, because of the high neutron-capture cross section of ^{197}Au (99 barns for thermal neutrons) and the appropriate half-life of ^{198}Au (2.7 d). Measurements of gold samples were performed by using extremely low-background Ge detectors set in the Ogoya Underground Laboratory[21]. As a result, ^{198}Au induced by the JCO criticality accident was detected up to 1400m from the JCO campus. Also the production rate of ^{198}Au could be well reproduced by a theoretical calculation based on the DOT-3.5 code[12].

The range-dose relation was also measured for the slow neutron product, ^{51}Cr, in stainless-steel samples collected from a range of 130–450m[9,18] and a fast neutron product, ^{32}P, induced by ^{35}Cl (n, α) ^{32}P and/or ^{32}S(n, p) ^{32}P reactions in table salt and sulfur containing reagent collected from a range of 70 to 360 m from the site[11]. The result of ^{32}P showed a strong directional dependence on the fast neutron fluence in a close vicinity (130–170 m) extended in the south-west direction. This observation was supported by beta-ray survey along the concrete fence boundary (Matsuzawa, T., private communication, 1999). These facts indicated that fast neutrons were shielded by the nearest buildings and some other materials between the precipitation tank and the sampling points (see Fig. 1 of Ref. 10). The range-dose (activity) relations obtained by ^{198}Au, ^{51}Cr and ^{32}P measurements are compared in Fig. 4 together with the result of the neutron counter

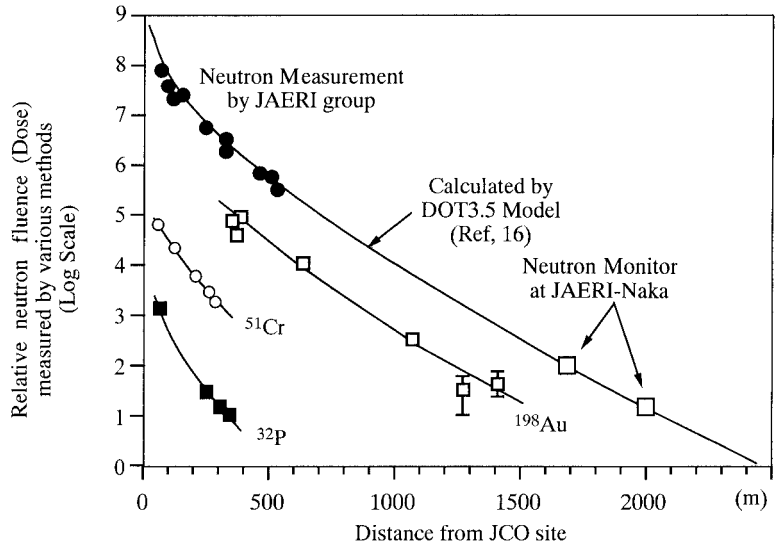

Fig. 4. Distance-dose (activity) relations of ^{198}Au (340–1400m), ^{51}Cr (70–270m), ^{32}P (70–360m) and neutron counter (90–2000m).

and a calculation by the DOT 3.5 Model.

Measurement of neutron-induced nuclides were made by many groups: soils[14,15], chemical reagents[13], gas cylinder[22], stainless steel net[23] and table salt[24,25].

(c) Visualization of the distribution of radioactivity in plant leaves using an imaging plate[26]

An imaging plate was applied to visualize the distribution of radionuclides in plant leaves. By this technique it was found that the concentration of radioactivity is higher in the veins

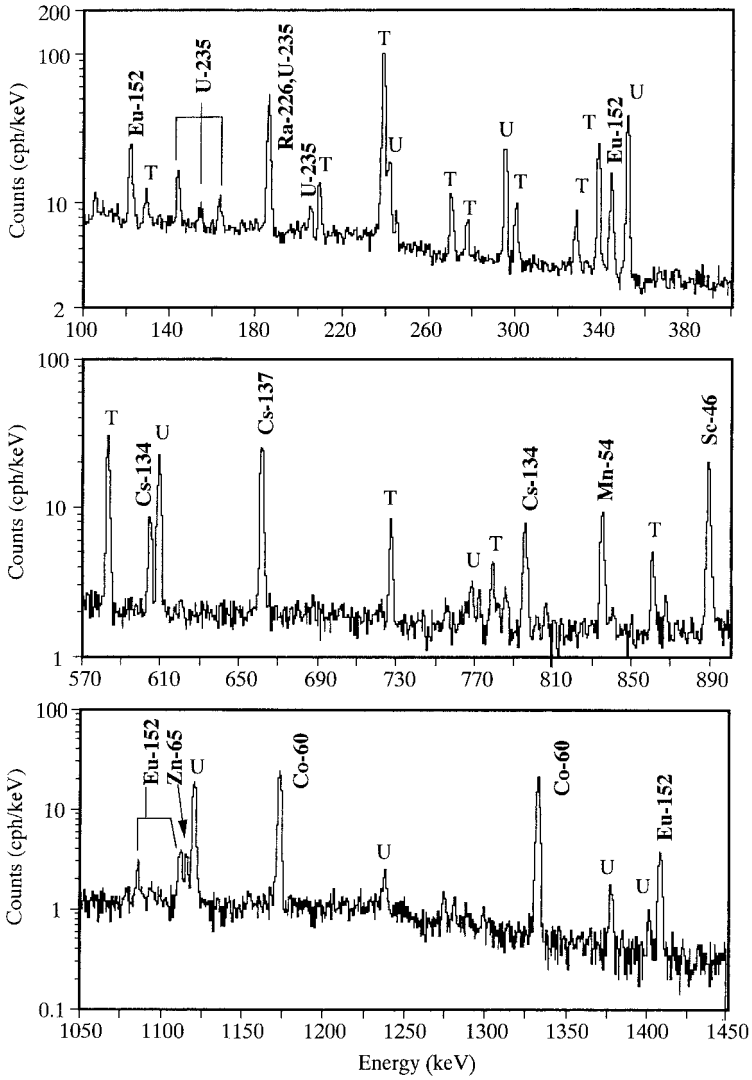

Fig. 5. Gamma-ray spctrrum 14 months after the criticality accident for soil collected 6m from the accident. T: thorium series nuclides, U: uranium series nuclides.

of leaves. By gamma-ray measurements, ^{131}I, ^{133}I, ^{137}Cs and ^{140}Ba-^{140}La were detected in these samples. All of these nuclides detected are volatile, or descendant of short-lived gaseous products ^{137}Xe and ^{140}Xe, probably leaked through an HEPA filter of the Uranium Conversion Building. Besides fission products, ^{32}P was detected by a radiochemical analysis, followed by low-background beta-ray counting at the Ogoya Underground Laboratory. The existence of ^{32}P indicates *in-situ* production by either ^{31}P(n,γ) ^{32}P or ^{32}S(n,p) ^{32}P reactions from phosphorus and/or sulfur in the leaf.

(4) Discovery of ^{235}U-enriched uranium leacage[19,20)]

One of the interesting results is the finding of ^{235}U-enriched uranium in surface soils and plants collected inside and outside of the JCO campus. The maximum value of the ^{235}U/^{238}U isotopic ratio observed was 0.0218[19)], which is 3-times higher than 0.0072 of natural uranium. Contamination by ^{235}U was also confirmed by non-destructive gamma spectrometry (see Fig. 5). The leakage of enriched ^{235}U is considered to be not by the criticality accident, but by some other accidents that occurred before.

IMPORTANCE OF A NUMBER OF LOW-BACKGROUND COUNTING FACILITIES AND OF INTERNATIONAL COOPERATION

As mentioned above, more than 400 environmental samples were collected, mostly during the 1st and 2nd sampling campaigns. These samples were distributed to 5~7 groups for gamma-ray measurements, mainly by Ge detectors. Unfortunately, the number of Ge detectors available to the environmental radioactivity group was too low (probably less than 30 Ge detectors); therefore, many of the samples could not be measured before the decay-out of short-lived nuclides. Furthermore, the background count of most of the Ge detectors are rather high for measuring the extremely low-level rationuclides produced by the accident. Without an extremely low-background Ge system and Si beta-ray counter in the Ogoya Underground Laboratory[27)], the detection of ^{198}Au in gold samples collected from more than 500 m distance and ^{32}P in table salts could not be detected. At least 3 to 5 ultra-low-background counting facilities like Ogoya should be constructed at various locations in Japan to prepare for future nuclear accidents.

Scientist of EC-JRC-IRMM in Belgium (IRMM) belonging to the European Commission offered to measure stainless-steel samples exposed by the JCO accident. In IRMM, 4 extremely low-background Ge systems are installed in an underground laboratory at 500 meters water equivalent of depth[27)]. Before sending the samples, a preliminary experiment was made to assess the contribution of cosmic-ray neutrons during 12 hrs of air-flight from Japan to Belgium. The productions of ^{51}Cr and ^{60}Co during flight were found to be negligibly low, even if the samples were not wrapped in cadmium sheet to suppress the contribution of cosmic-ray neutrons. The stainless-steel samples were sent to IRMM in January of 2000. The results of ^{51}Cr and ^{60}Co measurements will be published in the near future[28)]. This kind of international collaboration should be organized for future nuclear accidents like in the case of JCO.

OUR REMAINING TASKS CONCERNING THE JCO ACCIDENT

Although most short-lived nuclides induced by the JCO accident have already decayed out, some nuclides with a half-life longer than several hundred days still remain at detectable levels in the soil, fluorescence lamps, coins, stainless-steel products etc., though the activity levels are expected to be extremely low, and can hardly be detected without using an ultra-low-background counting system. Candidates of residual nuclides expected to be detectable in the JCO samples are summarized in Table 3. As an example, the gamma-ray spectrum of a soil sample measured 14 months after the accident is shown in Fig. 5, which clearly shows the neutron-induced ^{54}Mn, ^{65}Zn, ^{60}Co, ^{134}Cs, ^{152}Eu, ^{154}Eu and ^{155}Eu and fission product ^{137}Cs. Most of the ^{137}Cs may be derived from past global fallout, but partly derived from the leakage of the short-lived precursor ^{137}Xe. Gamma-ray peaks from ^{235}U indicate a leakage of ^{235}U-enriched uranium, as mentioned above. The measurements of long-lived nuclides are still considered to be very important to assess an accident. Some radionuclides cannot be detected without radiochemical purification before a measurement. Fig. 6 shows the effectiveness of chemical separation in the case of a ^{54}Mn measurement in a soil sample.

Except for uranium solution, no sample could be collected from the Uranium Conversion Building, because the building is under the control of judicial police. Details of the neutron fluence, energy spectrum and directional dependence of neutron emission, which are most essential to

Table 3. Candidates of radionuclides expected to be still detectable in environmental samples.

Product	Half-life	Major radiation γ-ray energy (abundance)	Production Reaction	Sample
^{3}H	12.33 y	β 18 keV	^{6}Li (n, α)	Li battery
^{14}C	5730 y	β 156 keV	^{14}N (n, p)	Reagent, Fertilizer
^{22}Na	2.60 y	1275 (99.94)	^{23}Na (n, 2n)	NaCl
^{36}Cl	3×10^{5} y	β 709 keV	^{35}Cl (n, γ)	NaCl
^{54}Mn	312 d	834 (99.978)	^{55}Fe (n, p)	Soil, Iron product
^{55}Fe	2.73 y	X ray	^{54}Fe (n, γ)	Iron product
^{60}Co	5.27 y	1173 (99.86), 1332 (99.98)	^{59}Co (n, γ)	Soil, SS Nickel coin
^{63}Ni	100 y	X-ray	^{63}Cu (n, p)	Copper wire
^{65}Zn	244.3 d	1115 (50.75)	^{64}Zn (n, γ)	Soil, Iron sheet
^{109}Cd	462.6 d	X-ray	^{108}Cd (n, γ)	Reagent
^{108m}Ag	418 y	434 (90), 614 (90), 723 (90)	^{107}Ag (n, γ)	Coin, Silver product
^{110m}Ag	249.9 d	648 (94.74), 885 (72.86)	^{109}Ag (n, γ)	Coin, Silver product
^{119m}Sn	293 d	X-ray	^{118}Sn (n, γ)	Tin plate, Tinware
^{134}Cs	2.06 y	605 (97.6)	^{133}Cs (n, γ)	Soil, reagent
^{137}Cs	30.0 y	662 (85.2)	Fission	Soil
^{152}Eu	13.33 y	122 (29.24), 344 (27.0)	^{151}Eu (n, γ)	Soil, Fluorescent lamp
^{154}Eu	8.593 y	123 (40.6), 1274 (35)	^{153}Eu (n, γ)	Soil, Fluorescent lamp
^{155}Eu	4.76 y	105 (21.2)	^{154}Sm (n, γ) Fission	Soil, Fluorescent lamp

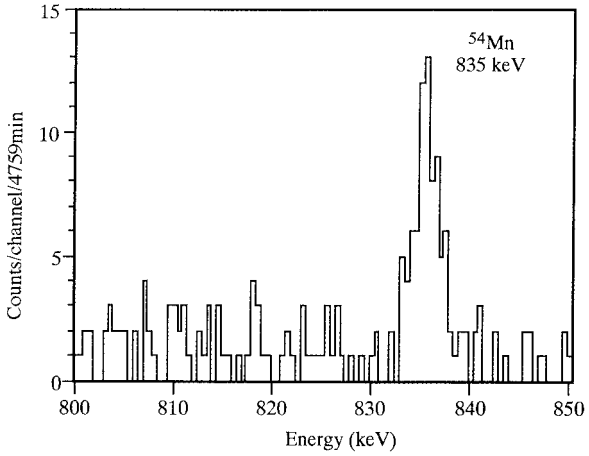

Fig. 6. Effectiveness of chemical separation for the measurement of extremely low-level ^{54}Mn in soil sample at 12m from uranium precipitation vessel. The sample was measured by an ultra-low-background well-type Ge detector in the Ogoya Underground Laboratory.

assess more details of the accident, will be obtained by analyses of samples collected directly in the Uranium Conversion Building. The samples of concern are cores of a concrete wall, ceiling and floor, fluorescent lamps, door knobs, electric wires and stainless-steel products at various distances and direction from the precipitation tank.

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