ABSTRACTS

Workshop on The Criticality Accident at Tokai-mura

19 May 2000

International Conference Center Hiroshima

Japan

Japan Health Physics Society

Contents

Workshop on The Criticality Accident at Tokai-mura

Programme

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2-3) Estimation of neutron fluences for three heavily exposed patients based on activation of biological materials from the Tokai-mura criticality accident.

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K. Kobayashi, M. Umemoto, T. Matsunaga (Japan Atomic Energy Research Institute)

18:00 – 18:05 **Closing Remarks** T. Iida (Nagoya Univ.)

Participation Fees: Participants to IRPA10 or Speakers (Free), Members of JHPS (¥2,000), Non-members of JHPS (¥3,000), Students (¥1,000)

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1-1)

The Number of Total Fission Events in the JCO Criticality Accident Determined by Passive Observation of Some Fission Products Gamma-rays

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c Department of Chemistry, Tohoku University, Aramaki, Aoba-ku, Sendai 980-8587, Japan.
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e Radioisotope Research Center, Aichi Medical University, Nagakute-machi, Aichi 480-1195, Japan.

Abstract

Passive γ-ray spectrometry was carried out from outside of the Uranium Conversion Building by using a portable Ge detector (relative efficiency of 11%) and portable 4096 ch PC-PHA. The γ-rays of ^{95}Zr, ^{103}Ru and ^{140}La-^{140}Ba were observed with good counting statistics. The relative intensity of the γ-rays of ^{140}La observed at 329 keV, 487 keV, 816 keV, 1596 keV and 2522 keV allowed the inner calibration of counting efficiency and the attenuation of the γ-rays of ^{95}Zr and ^{103}Ru with high accuracy. The large attenuation of the 1596 keV γ-ray with the wall of the building was estimated experimentally by using the same specimens of the wall material and the attenuation with the uranium solution was calculated by assuming the solution was retained in the precipitation vessel in which the criticality accident took place. The total fission events (TFE) thus determined was 3.14×10^{18} with 3σ of 0.39×10^{18}. The TFE value is in good agreement with the analytical result of the uranium solution that was performed by JAERI. This agreement implies that the criticality accident took place in a small limited volume and that the majority of the fission products were retained in the precipitation vessel.

Key Word: JCO Criticality Accident, Total Fission Events, Passive Measurement, Gamma-ray Spectrometry

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1-2)

Dosimetry study on 350m zone-residents around JCO accidental site taking account the directional dependence on radiation

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A criticality accident at uranium conversion facilities in Tokai-mura Japan on September 30 1999 caused neutron exposure residents. The distance between the nearest residence and uranium source was only about 100 m. Evacuation of people in 350m zone was started after 5 hours. There was a remarkable directional distribution on neutron beam which depends on complex internal and external structures of building surroundings critical uranium. The neutron flux changed 1.0~0.2 relatively in various direction. External effective doses on residents in 350 m-zone near the accidental factory were estimated taking account the directional dependence on radiation. The maximum doses for outdoor and indoor of 350m zone are estimated to be 30 and 12 mSv respectively. Average doses for outdoor and indoor of 350m zone are estimated to be 5.6 and 2.2 mSv respectively. If the all of residents were indoor during the accident, 47 % residents in 350 m zone might get external dose less than 1 mSv.

1-3)

Activation of gold by JCO accident on Sep. 30, 1999

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According to the report of Investigation Committee for Criticality Accident at Uranium Processing Plant, 2.5 x 10^{18} of fission events might have occurred during 20 hours after the break out of the accident at JCO in Tokai-Mura. Great number of neutrons had leaked out to the environment and many materials were expected to be activated by the neutrons mainly through (n, gamma) reaction.

In order to access the environmental impact of the accident, some 400 samples were collected from the surrounding area of the JCO site mostly in the 1^{st} survey (Oct. 7-8) of the university research group of Monbusho. Collected were soils, plant leaves, table salts, sugars, chemical reagents, various kinds of coins, spoons made of stainless steel, gold items, fluorescent lamps, ceramics etc. Among these samples, gold items such as gold rings, necklaces, coins etc. are considered to be most sensitive to evaluate the neutron fluence at various distance from the JCO site, because neutron capture cross section of ^{197}Au(n,)^{198}Au reaction is extremely high (99 barns) and half-life of ^{198}Au (2.695 d) is not too short to measure by gamma-ray spectrometry.

Fourteen gold samples collected from up to 2.7km from the JCO site by 1^{st} survey and two samples by 2^{nd} survey (Oct. 23-26) was used to measure ^{198}Au. In order to avoid the production of ^{198}Au by high altitude flight, these samples were hand-carried by train connection (9 hours) from Tokai-Mura to underground laboratory, where extremely low-background Ge detectors are installed in a tunnel of former Ogoya copper mine (270 meters water equivalent of depth). Three Ge detectors (93.5 % coaxial type Ge, 65% well type Ge and 30% planar type Ge) specially designed for low-background measurement and one ordinary Ge detector (40%) were used to measure neutron-induced ^{198}Au. The gold sample collected from a distant place was measured first to obtain maximum outcome within limited time before decay of short-lived ^{198}Au. Measuring time were ranged from 51 to 250 ksec depending on the intensity of ^{198}Au activity and limitation of counting time.

Despite of such a short measuring time, ^{198}Au was detected in all of the samples even for 2.7 km point one and also in mercury of a clinical thermometer collected from 600 m point.

Number of ^{198}Au atoms/g-gold was calculated from peak area of the 412 keV gamma ray from ^{198}Au. Contribution of natural ^{198}Au activity induced by environmental neutrons was subtracted from measured value to obtain net ^{198}Au activity derived from the accident. Tentatively 50 ^{198}Au atoms per gram of gold was adopted for natural production of ^{198}Au. This value is the average of experimental values measured for 20 to 50 g of gold sample exposed to environmental neutrons. Then number of ^{198}Au atoms per gram of gold sample decay corrected to 6:14 of Oct. 1 was calculated by assuming the neutrons emitted during burst and plateau periods being 11.4% and 88.6%, respectively.

Result of measurement is shown in Fig. 1 as a function of distance from the JCO site. The curve in Fig. 1 shows calculated value based on neutron transport code DOT3.5. The height of the curve is adjusted to best fit to the measured values. As known from the figure, general tendency of ^{198}Au activity (closed circle) can be well reproduced by theoretical calculation up to about 1400m point, though some values are much lower than the curve. The open circle of 1056m-necklace sample was corrected to the value shown by an arrow from exposure history of the owner. The higher value of 2130m-necklace was explained by natural production by second measurement of the same sample made 45 days after the accident (natural production of ^{198}Au in necklace is 90 atoms instead of 50 atoms). The lower value may be explained by shielding of neutrons depending on the exposed conditions. Detail analysis will be made by the data obtained by recent experiment using mock-up sample exposed to neutrons from ^{252}Cf.

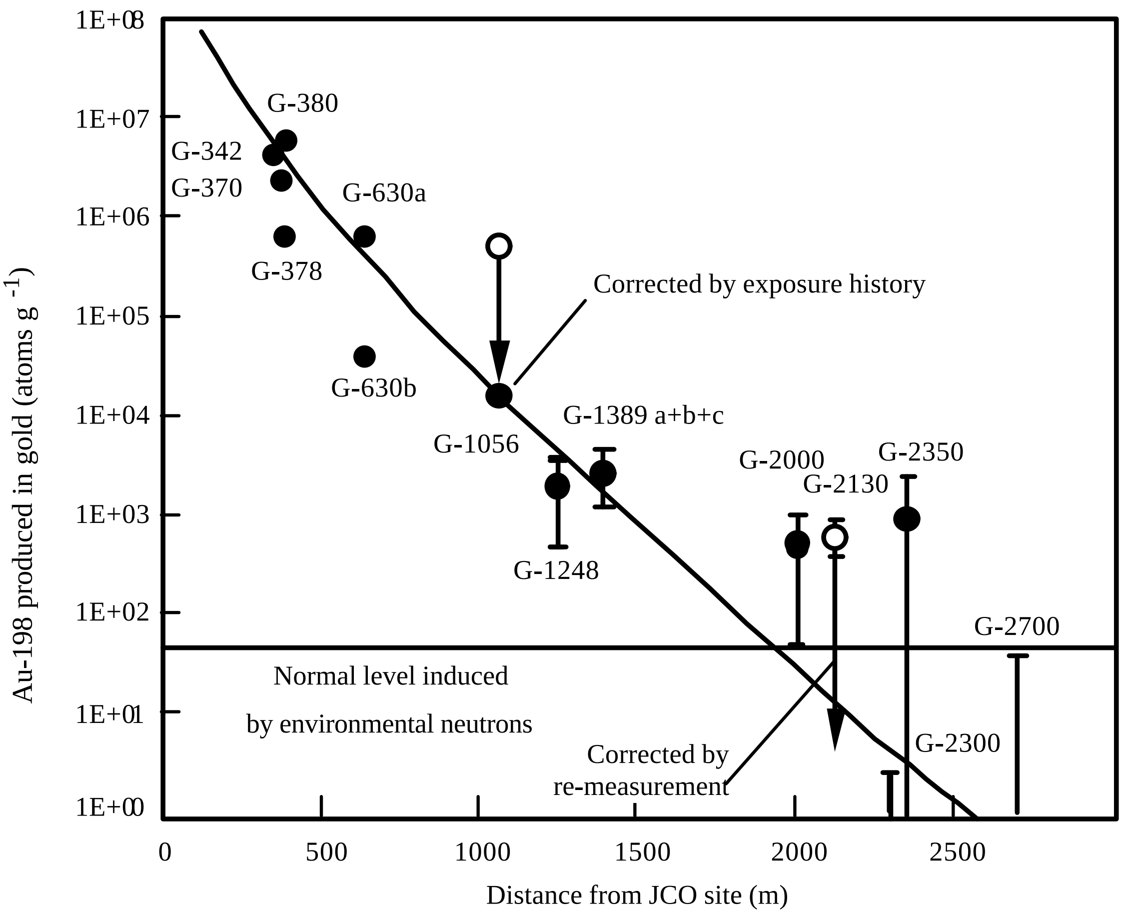

Fig. 1 Au-198 induced by JCO accident. Open circle shows data before correction.
The curve shows calculated value based on neutron transport code DOT 3.5.

1-4)

Emergency Environmental Monitoring for JCO Criticality Accident

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The criticality accident has occurred in JCO Co. Ltd. (Uranium conversion facility) on 30th September 1999. JCO is located in the northwest of Tokai-mura and it leaves about 6km east of Japan Nuclear Cycle Development Institute (JNC) Tokai works.

In this accident, rare gas and iodine isotopes were discharged to the atmosphere, and neutron and gamma radiation were directly emitted to the environment. The emergency environmental monitoring was carried out immediately after the accident in order to grasp the effect to the environment. JNC participated in the emergency monitoring with Ibaraki Prefectural government and Science, Technology Agency, JAERI and others. The airborne radioactive materials, leafy vegetables, milks, drinking water, rain water, sea water, marine products, soils and radiation exposure dose rates were collected and measured. Real time radionuclides diffusion were estimated by SMAP (Simulation and MAPping system of emergency environmental effect).

As a result of the emergency environmental monitoring, some of the iodine sample collected by charcoal filter was detected ^{133}I ((3.6〜3.9)×10^{-7}Bq/cm^3) and ^{135}I ((1.6〜3.4)×10^{-6}Bq/cm^3). In some dust samples, concentrations of ^{138}Cs, which is progeny of ^{138}Xe were 0.016 0.39Bq/cm^3. In a part of soil sample, ^{24}Na that was produced by neutron irradiation was detected, their concentrations were 1.3〜56Bq/kg. In a vegetable and grass samples, ^{131}I (〜4.7Bq/kg), ^{133}I (〜7.1Bq/kg) and ^{135}I (〜10Bq/kg)were detected. The detected activities were less than the level that was affected to the public health or serious damage in the environment.

1-5)

Dose Evaluation on the Basis of Na-24 Activity in the Human Body for the Criticality Accident at JCO Tokai Nuclear Fuel Processing Plant

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Sodium-24(^{24}Na) generated in human body due to neutron activation was measured by whole body counter at JNC Tokai Works. Total 148 persons (JCO employees and contractors, public member, Tokai-mura emergency service workers, etc.) were measured and a significant activity of ^{24}Na was detected in the 62 persons*.

Neutron energy spectrum around the uranium conversion facility was calculated using ANISN and MCNP4B codes and spectrum averaged capture probability of neutron for human body was about 0.25 at any distance from the center of the precipitation tank. Effective dose equivalent for the 62 persons were estimated based on the calculated conversion factors from ^{24}Na specific activity to neutron dose. Maximum ^{24}Na activity was 7.7 kBq (83 Bq(^{24}Na)/g(^{23}Na)) in total body and the relevant effective dose equivalent was 47 mSv.

* except three JCO employees who were severely overexposed

Table 1 Number of persons exposed to external gamma and neutron and estimated doses

Persons / Group	Number	Range of estimated effective dose equivalent (gamma + neutron) [mSv]
Public	7	6.7 ~ 16
Tokai-mura emergency service workers	3	4.6 ~ 9.4
JCO employees	36	0.6 ~ 47
JCO employees engaged in the operation to drain water from the cooling jacket	16	5.9 ~ 45

Figure 1 JNC-Whole body counting system with Ge detectors, installed in the special shielding room

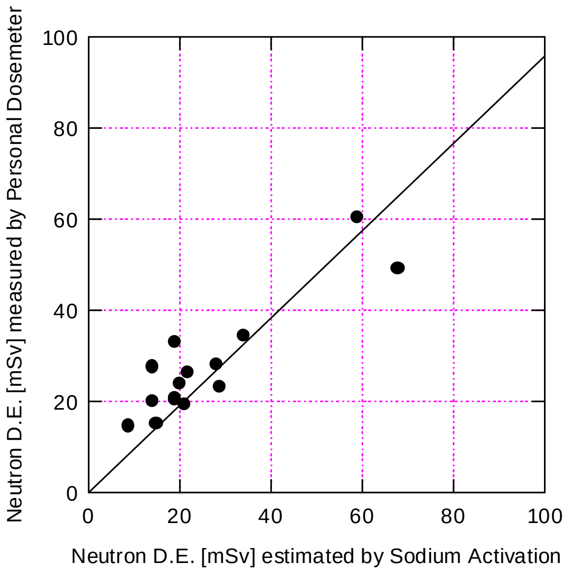

Figure 2 Comparison of neutron doses (*Hp(10)*) estimated by Na-24 activity and electric personal neutron dosemeter (worn by workers engaged in the operation for termination of criticality)

1-6)

A School Teacher and Policemen Measured the Gamma Dose Rates Around the JCO Tokai Plant with GM Survey Meters for Their Own Safety During the Criticality on September 30, 1999

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INTRODUCTION

Ibaraki National College of Technology (INCT) is located in Hitachinaka-shi, Ibaraki-ken, which is surrounded by Tokai-mura, Oarai-machi and Naka-machi. Since there are a lot of nuclear facilities in those areas, residents in Hitachinaka-shi could suffer from damage if a severe nuclear accident happens. According to the antidisastar plans (nuclear accident section) established by Ibaraki Prefectural Government and Hitachinaka-shi, our school is designated as a "concrete - building - indoor - evacuation" site with capacity of 1,330 persons. Nevertheless, except for an appointment notice we received from the local government, we so far have nothing but a wireless radio (a loudspeaker) connected to the administration, which is installed outside the building. We are not equipped with any radiation survey meters. While the administration has been working on a revision of these emergency plans, including a new two-way communication system, our school (INCT) is excluded from this plan because our school is a national one. This means that detailed information would never be provided to us directly, even if a severe accident were to happen. We must establish our own safety and radiation protection measures, regardless that our building is an appointed evacuation site for severe nuclear accidents.

METHOD

A Fuji Electric GM survey (NHJ110), made in 1994 with indications of μSv/h, and an Aloka GM survey meter (TGS-111), made in 1981 with indications of mR/h units, were used for this study. The latter was calibrated by the Institute of Radiation Measurement, Japan, in 1998.

MEASUREMENTS on ROUTE 6 (15:55 to 18:00 local time on Sep. 30. 1999)

Firstly, the first author carried a Fuji Electric GM survey meter in a car, measuring gamma dose rates as he drove on Route 6 down to Tokai-mura.

MEASUREMENTS AROUND the JCO FACILITIES (20:00 to 20:30 local time on Sep. 30. 1999)

Carborne measurements were conducted in a patrol car, from the Nikenjaya intersection to the western corner of the JCO campus on Prefectural Road 62. Measurements were made at around 20:00 local time, September 30, 1999. A barricade placed at the western corner of JCO was set for an origin, and fixed measurements were made with 100m increments, by comparing two GM survey meters' indications. In order to follow the spread of the radiation, additional measurements were made at an intersection near the entrance of the Tokai-mura municipal office and at a bridge over the JR Joban line along the Prefectural Road 62.

RESULTS

Indications of both GM meters were well correlated with each other. A maximum dose rate along Route 6 of 20.8 μSv/h was observed at 280 m away from the source. Along Prefectural Rad 62, a maximum dose rate of

510 μSv/h was recorded at 70 m away from the source, the border of JCO. It should be noted that, within this point, a GM survey meter carried by the police exceed its upper limit by 3 points around JCO.

DISCUSSION

After watching the report on the JCO criticality accident on TV, the first author started the gamma survey in the INCT campus and confirmed that the gamma dose rate was not showing any increase above the normal level (0.1 μSv/h). Also, a guidance was given to approximately 170 students living in a school dormitory, to make their choice of going home, or staying in the dorm; made in the interests of their safety. After finishing all tasks at school, the first author started the actual investigation in the evening of September 30, 1999. Since there was no information regarding radiation and radioactivity from the JCO criticality accident at that time, the first author started taking carborne measurements with a GM survey meter. On the way through Hitachinaka-shi and Naka-machi, no increase of the gamma dose rate was observed, while in the area around Nikenjaya intersection in Tokai-mura it increased up to 20 μSv/h . However, the gamma dose rate along Route 6 was not so high compared to that observed along Prefectural Road 62, simply because of the distance.

The first author visited a police base, which was set up at a parking lot of an electronic shop near the Nikenjaya intersection, and asked a police chief for information. Since the police were being engaged in guard duty without receiving any information regarding the radiation and radioactivity from the accident, the police appreciated the author's visit and asked him for the information instead. The first author then cooperated with their traffic control unit and made radiation dose measurements along the Prefectural Road 62. Later on, the authors went to the headquarters of the Ibaraki Prefectural Police to explain the results of the gamma dose rate measurements.

Our data[1] seem to be comparable to those obtained by specialists and professionals. Data obtained by professionals such as JCO, the Japan Nuclear Cycle Development Institute and the Japan Atomic Energy Research Institute were all returned to the Science and Technology Agency (STA) and were not disclosed to the public until the joint meeting of the ministries that was held on the night of the accident. Although residences and public roads were exposed to the radiation from the criticality accident, there were no radiation warning signs nor indications posted. Even the policemen guarding the vicinity of the JCO campus were not notified of the radiation information. In other words, such important data obtained by the professionals were not properly utilized for the actual protection of the public. The official data of the radiation dose rate were eventually released in the end of October. The authors would like to emphasize that in such an emergency case, those data should have been disclosed as soon as possible, regardless of their exact preciseness.

It is difficult for the first author to understand the reason why the specialists are not discussing radioactive fallout from China's atmospheric nuclear tests in the 1970's as a comparison in a discussion of the radiation exposure due to the JCO criticality accident. It was only the second time, since Chinese tests, that the first author witnessed the indication of a GM survey to become saturated.

ACKNOWLEDGEMENT

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REFERENCE

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2-1)

Environmental Radiation Monitoring on JCO Nuclear Criticality Accident

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[2]Ibaraki Prefectural Consumer Information Center

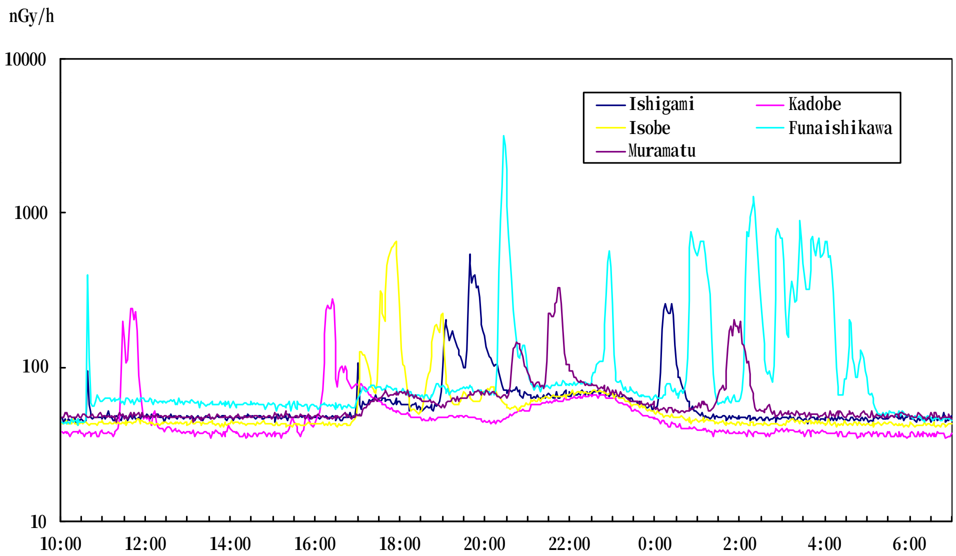
1. Environmental gamma-ray monitoring

1) Measurement by Centralized Environmental Radiation Monitoring System

2"φ×2"NaI detector with DBM module and ionization chamber instantaneously detected prompt γ-ray at 10:38 at Funaishikawa station(1.4km south from JCO), where air absorbed dose rate(NaI) was 0.40 μGy/h for 2 min. γ dose rate increased with the plume of radioactivity as follows. Dose rate recorded 0.20 μGy/h at 11:28 at Kadobe station(6.3km west), and kept the level for 20 min, then returned to normal level. When the wind direction changed at about 16:00, dose rate rose again from 16:12 to 16:42. At Isobe station(4.4km north) 3〜10 times normal level was detected from 17:00 to after 18:00. Dose rate also rose from about 19:00 at 2km north-east and 2〜4.5km east, and from about 20:00 at the place of 5〜11km south-east and 3.6〜7.7km south.

After the nuclear criticality came to an end at 6:00 next morning, γ dose rate went back to normal level at all stations. The highest record was 3.2 μGy/h at 20:26 at Funaishikawa(1.4km south).

The dose increase by this accident was 2.1 μGy maximum, subtracting 1μGy as the increase part by natural radioactivity of rain.



Trend of γ-dose rate at monitoring stations

2) Gamma-ray survey

γ-ray survey by monitoring car was performed 4 times, i.e. from the point of 3km north-west to the JCO(12:49〜13:21), the area within 4km(14:44〜15:50), on the north-west and south-west border line of JCO (where dose rate was 18〜440 μGy/h(15:50〜16:01)), and on the border line of JAERI Naka at south-west of JCO(16:17〜16:33). The survey provided a good grasp of γ-ray situation at the circumference of JCO.

3) Thermo-luminescence dosimeter

The TLD placed in the field from before the accident was read to measure the integral dose. The number of measurement points was 101 total in Tokai area placed by 7 institutions(JAERI(2), JNC, JAPCO, NDC, Tokyo

Univ., EPRCI).

The integral dose was 270 μGy at 300m from JCO conversion testing plant, 30 μGy at 600m and 20 μGy at 600〜800m, but was the same level as usual at other 93 points.

2.Measurement of environmental samples by Ge semi-conductive detector

The number of samples and the result of measurement are presented in the table.

Samples were measured for more than 2000sec by three HPGe detectors for quick result in early monitoring stage and measured for more than 20000sec in second stage after Oct. 12th. Radioactive inert gas and gaseous radio-iodine were released to environmental circumference by the accident, but particle materials were almost removed by HEPA filter. Sodium in soil were activated by the neutron emitted from JCO conversion testing plant, and Na-24 was detected in soil.

3. Requests for contamination survey

In addition to emergency monitoring program, requests for survey of agricultural products etc. rushed to the center from agricultural associations, commerce and industry associations, schools and individual persons. Measurements were done with GM or NaI survey-meter for ease of mind. The results showed all normal levels. The number of survey by the center and JAERI, JNC, JAPCO were 1400 for agricultural products etc., 95 for school grounds and 700 for well waters by Ge detctor.

4. Conclusion

1) γ-ray dose detected by TLD and centralized monitoring system were 270 μGy at a nearest point. This value is the same in Tokai area for cumulative one year.

2) A part of environmental samples indicated the radioactivity originated in the accident, but the activity was very low level and decreased in short time.

Therefore, the radioactivity released to environment had no impact on health of inhabitants.

Table 1.Concentrations of radionuclides in environmental samples

Sample item	Number of measurement	Results of measurement: No. of detected	Nuclides	Detected range
First stage monitoring				
Air dust	81(17)	1(1)	Sr-91	2.1×10^{-8} Bq/cm^3
		8(0)	Cs-138	1.6×10^{-8}〜1.4×10^{-3} Bq/cm^3
		1(1)	Ba-140	2.2×10^{-9} Bq/cm^3
		1(1)	La-140	1.6×10^{-9} Bq/cm^3
Air iodine	50(10)	3(0)	I-131	1.3×10^{-10}〜4.1×10^{-7} Bq/cm^3
		3(0)	I-133	1.3×10^{-9}〜3.9×10^{-7} Bq/cm^3
		2(0)	I-135	1.6×10^{-6}〜3.4×10^{-6} Bq/cm^3
		3(0)	Cs-138	9.2×10^{-6}〜1.4×10^{-5} Bq/cm^3
Rain	3(2)	0		
Service water	17(13)	0		
Well water	31(31)	0		
Lake.marsh	3(0)	0		
Soil	138(29)	16(7)	Na-24	1.3×10^{-3}〜1.3×10^{-1} Bq/g
		2(0)	Mn-56	3.7×10^{-2}〜1.3×10^{-1} Bq/g
		1(0)	I-131	4.5×10^{-4} Bq/g
		1(0)	I-133	1.6×10^{-3} Bq/g
		88(20)	Cs-137	1.4×10^{-3}〜2.6×10^{-2} Bq/g
Agricultural products (including weeds)	115(29)	12(4)	I-131	9.5×10^{-4}〜3.7×10^{-2} Bq/g
		7(2)	I-133	3.9×10^{-3}〜3.8×10^{-2} Bq/g
		2(0)	I-135	1.3×10^{-2}〜1.4×10^{-2} Bq/g
Stock farm products	19(16)	0		
Marine products	13(13)	0		
Sea water	7(4)	0		
Second stage monitoring				
Air dust	57(27)	0		
Air iodine	45(27)	2(0)	I-131	7.3×10^{-11}〜1.2×10^{-10} Bq/cm^3
Soil	18	18	Cs-137	1.8×10^{-3}〜1.7×10^{-2} Bq/g
Leaf vegetable	18	0		

note:Values in parentheses were measured by EPRCI

2-2)

Determination of Fission Products and Uranium Isotopes in Soil and Plant Samples Collected around the Uranium Conversion Building in JCO Campus

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INTRODUCTION

Enriched U (18.8% of ^{235}U by mass) was used in the uranium conversion building of JCO Company Limited, Tokai-mura, Ibaraki Prefecture, Japan, on the day of the criticality accident (September 30, 1999). In the purification process, uranium oxide (U_3O_8) powder was dissolved in nitric acid in a precipitation tank. Three workers of JCO mixed the uranium oxide powder with nitric acid in a bucket made of stainless steel. They then poured the solution containing U into the precipitation tank directly from the bucket by hand. A mass limitation prescript in the license for this tank was 2.4 kg U (enrichment level: 16 - 20 %). However, about 16 kg of U (total amount of solution: about 40 l) was added into the tank without mass control equipment, causing the occurrence of the criticality event. In the process, three workers were heavily exposed to neutron and gamma-ray produced by the fission. The chain reaction continued for about 20 hours. The self-sustaining nuclear chain reaction was terminated by removing cooling water, which acts as a neutron reflector, from the water jacket of the tank. The solution was estimated to be not boiled.

Appreciable amount of gaseous fission products such as ^{131}I, ^{133}I, ^{135}I, ^{138}Cs and ^{91}Sr and activation products such as ^{24}Na and ^{56}Mn were found in air samples and in some soil and vegetation samples collected near the JCO campus immediately after the criticality accident (1, 2). However, there was almost no information on the levels of fission products inside the JCO campus. In addition, there was no data on the levels of U in the environment around the uranium conversion building. Since enriched U was used in this facility, the $^{235}U/^{238}U$ ratio might be useful in assessing the possible contamination of U.

In the framework of "Studies on environmental effects of the criticality accident", we collected soil and plant samples in JCO campus (3, 4). The purpose of this study is to determine the fission products and U isotopes (^{235}U and ^{238}U) in soil and plant samples collected near the uranium conversion building in the JCO campus.

SAMPLING AND MEASUREMENTS

Sampling of soils and plants:

Plant samples belonging to four species were collected around the uranium conversion building on October 7 and 26, 1999. Sampling points were decided considering the distance and direction from the center of the building. The plants were cut at points of about 3 cm above the ground surface in order to avoid the direct contamination from soil. In laboratory, leaves were collected and measured for radionuclides using Ge-detector. Remaining leaves were oven-dried at 80 ^{o}C until constant weights and pulverized with a blender. Surface (0 - 5 cm) soil samples were collected around the uranium conversion building on October 7, 1999, November 27, 1999 and January 22, 2000. Soil cores were also sampled at some points in order to get depth profile. After removing stones, the soils were oven-dried at 80 ^{o}C until constant weights, and ground into powder.

Gamma-ray measurements of plants:

About 50 g of raw samples of leaves were packed in plastic bottles (diameter, 60 mm). Radionuclides were determined using a Ge-detector (Ortec GMS-30185) coupled to a multichannel analyzer (Seiko EG&G 7800). Counting times for each sample were 6000 - 80000 sec. Counting errors for the measurements of radionuclides were usually better than 10%. If peaks were not detected, limits of detection (3 times of the background counts) were calculated.

Determination of ^{235}U, ^{238}U and ^{232}Th:

Samples (0.1 g for soils and 0.2 g for plants) were digested in Teflon^{TM} PFA pressure decomposition vessels with HNO_3, HF and $HClO_4$. A microwave digester (CEM, MDS 2000) was used for heating the samples. After digestion, the samples were evaporated to dryness on a hot plate. Then, the residues were dissolved in 1-2% HNO_3 to yield the sample solutions. Decomposition and analysis were duplicated for each sample. ICP-MS (Yokogawa PMS-2000) was used for the analysis of ^{235}U and ^{238}U. ^{232}Th was also measured for soil samples. An internal standard, Bi, was used to compensate for changes in analytical signals during the operation. Standard solutions were prepared from SPEX Multi-Element Plasma Standards (SPEX Industries Inc.) and used to get calibration curves. Standard reference materials such as JB-1a (basalt) and Tomato Leaves (1573a) were used to validate the analytical procedure.

RESULTS AND DISCUSSION

Artificial radionuclides detected in plants were ^{131}I, ^{133}I, ^{140}Ba-^{140}La and ^{137}Cs. The nuclides ^{140}Ba-^{140}La and ^{137}Cs were thought to be produced from ^{140}Xe and ^{137}Xe, respectively, in the following reactions:

^{140}Xe (13.5 s) ^{140}Cs (63.7 s) ^{140}Ba (12.8 d) ^{140}La (1.68 d)

^{137}Xe (3.82 m) ^{137}Cs (30.1 y)

The highest ^{131}I concentrations of 6300 Bq/kg-wet (decay corrected to October 1) was observed in fern collected near the uranium conversion building. The concentrations of ^{140}Ba-^{140}La and ^{137}Cs were the highest near the exhaust which was located at 18 m above the ground on the building next to the uranium conversion building. The concentrations of radionuclides decreased markedly with the distance from the building (or from the exhaust). Compared to the Japanese intervention levels of these radionuclides for foodstuffs, the concentrations observed in plants outside the JCO campus (>60 m from the uranium conversion building) were markedly lower.

Analytical results of U for soils and plants collected on October 7, 1999 were summarized. Concentrations of U in soils were comparable to the U values for common Japanese soils. However, the U/Th ratios for these soil samples were markedly higher than the control value, suggesting the possible contamination of U. The ^{235}U/^{238}U atom ratios in soil and plant samples were notably higher than the natural ratio, 0.00725. The highest values, 0.0162 for soil and 0.0193 for plant were found in samples collected near the conversion building. However, relatively high ratio was observed at the site more than 50 m from the building, indicating that enriched U may have been released not only from the conversion building but also from other unknown sources. The ^{235}U/^{238}U atom ratios were in general higher in plants than those in soils collected at the same points. The vertical profiles of U concentration and ^{235}U/^{238}U ratio in soils were also discussed.

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2-3)

Estimation of neutron fluences for three heavily exposed patients based on activation of biological materials from the Tokai-mura criticality accident.

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Three workers were carrying out a purification procedure in the uranium conversion building of JCO Company Limited, Tokai-mura (Ibaraki Prefecture), which resulted in the criticality accident of September 30, 1999. About 16 kg of U (enrichment of U-235: 18.8%) dissolved in nitric acid had been added to a precipitation tank causing the nuclear fission chain reaction to start. The three workers were heavily exposed to neutrons and gamma-rays produced by the fission event. They were transported by helicopter to the National Institute of Radiological Sciences (NIRS), Chiba, where intensive medical care was provided for them.

Immediately after their arrival at NIRS radiological surveys were performed and activation products such as Na-24 were found in samples from the patients. Contamination by fission products such as Sr-91, Y-91, Ba-140 and La-140 was found in their bodies (hair) and remaining clothing (underwear), although the levels were very low. From the first survey results we could confirm that the most important radiological effect for the patients would be due to the irradiation by neutrons and not to the contamination by fission products or uranium. In order to understand the amounts of radionuclides produced in the body, we have carried out quantitative measurements of radioactivities in biological samples such as blood, urine, vomit and hair with Ge-detector systems. The radionuclides (activation products) determined in these samples were Na-24 (half-life: 15.0h), Br-82 (35.3h) and K-42 (12.4h), which were produced by the n-gamma reaction of stable elements (Na-23, Br-81 and K-41) in the body.

In addition to the radioactivity measurements, concentrations of the stable elements in the same samples were determined by ICP-AES and ICP-MS to obtain specific activities (concentration ratio between the produced radioactivity and the target element), which are necessary in the estimation of an individual's neutron fluence. The specific activities for Na-24 in blood samples collected on 1 October from the three patients A, B and C were 75, 39 and 11 kBq/g, respectively. The values obtained for urine from A, B and C of 87, 50 and 14 kBq/g, respectively, were somewhat higher than those for blood. (The values were decay corrected to the beginning of the accident at 10:35, but the biological half-life of the nuclide was not considered in the calculation of the specific activities.)

The specific activities of Br-82 in blood collected from A and B were 56 and 28 kBq/g, respectively. Br-82 in blood from C was under the limit of detection. In contrast to Na-24 the values of Br-82 for urine from A, B and C (48, 23 and 6.3 kBq/g) tended to be lower than those for blood. The specific activities of K-42 in blood collected form A and B were 15 and 7.2 kBq/g, respectively. The values of K-42 observed in urine from A, B and C were respectively 14, 6.9 and 2.3 kBq/g, which were similar to those for blood. We also obtained values for specific activities of these nuclides in other samples such as vomit and hair collected from the patients.

Finally we have estimated the neutron fluences for the three patients using these obtained specific activities and available nuclear data in the literature. We also have considered the cause of some variations observed in the specific activities for different biological samples. Details on the results will be reported at the meeting.

2-4)

Radiological Doses Due to Radionuclides Discharged into the Atmosphere

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The radiological doses due to radionuclides discharged into the atmosphere from JCO was estimated by using an emergency response system SPEEDI.

Calculation conditions are as follows:

(1) Objective region: 25 x 25 km squire around the JCO site. For atmospheric transport simulations, the region is divided into 100 x 100 grid cells with a horizontal resolution of 250 m.

(2) Meteorological data: Meteorological forecasts over Japan initialized at 9:00 JST, 30 Sept. from Japan Meteorological Agency (JMA) and observed meteorological data around JCO.

(3) Release Condition: Continuous unit(1 Bq/h) release of noble gases and Iodine from 10:35 JST 30 Sept. to 6:00 JST 1 Oct.

Atmospheric dispersions of radionuclides are predicted by using SPEEDI of JAERI. According to the prediction, meteorological conditions during the accident are divided into four characteristics. The first was the sea breeze from E or ESE with wind speed of 2-3 m/s during the period from 10:00 to 17:00, 30 Sept. The second was transient period from sea to land breeze around 17:00 to 18:00. The wind became almost calm and wind direction changed widely. The third period from 19:00, 30 Sept. to 3:00, 1 Oct. was land breeze from NE. The last was after 4:00, 1 Oct. whose winds were strong and from NE. During the first period of sea breeze, the radioactive plume was transported to the direction of WNW and this situation continued till 17:00. In the transient period, the week southerly wind transported the plume to the north slightly. After 20:00, the plume was transported to the seacoast area by land breeze and covered the southeast area. This situation continued till 3:00, 1 Oct., and finally the strong northeast winds transported the plume to the southeast around daybreak. Since the HEPA filter was working at the ventilation duct and, consequently, discharged radionuclides were mainly noble gases, the air dose rates observed at the far area were due noble gases. Thus, the ratio of observed values to predicted ones means the release rate of noble gases. The release rates are calculated by using the ratios of pair of the peaks from prediction and observation. The estimated release rate is 8 $x10^{12}$ Bq/h by geometrically averaging the values in the table.

The outline of external doses due to noble gases can be calculated by the multiplication of the estimated release rate, 8×10^{12} Bq/h, to predicted external doses under the assumption of 1 Bq/h release. The external doses were higher at the region of the northeast and southwest where were downwindword for long time. However, the estimation shows that the doses were considerably lower than guideline values for sheltering even at the vicinity of the site. The maximum of effective dose equivalent was 1/10 of the dose limit for 1 year to the public. Thus, it is concluded that radiological doses due to radionuclides discharged into the atmosphere have no harmful influence to the human health.

2-5)

Information Transfer Problem Related to Emergency Response Measures during the Criticality Accident at Tokai Uranium Processing Facility

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1. Purpose

On 30 September in 1999, the reactivity accident occurred at the Tokai uranium processing facility of J. C. O. Co. Ltd (JCO) at Tokai-mura, Japan. Before the accident had been brought under control, the authorities evacuated the population living within a few hundred meters and advised people within about 10km of the facility to take shelter for a period of about one day.

To enhance the effectiveness of protective measures dealing with a nuclear disaster, information transmission systems both from the local authorities to the public and among organizations concerned have been developed.

In the criticality accident, however, some of the information transmission systems did not work appropriately. This caused lack of appropriate decision-making procedures for emergency response measures.

This report is to describe the information transmission problems related to emergency response measures taken during the criticality accident.

2. Method

The chronology of information transfer among JCO, governments and the public related to emergency response measures taken was developed based on the information obtained through hearing from JCO and the governments. The chronology was analyzed to find the information transfer problems related to the emergency response measures. Moreover, a questionnaire was conducted to know when, where and how the inhabitants received the information of protective action request from Tokai-mura authorities.

3. Result

(1) At around 15:00, evacuation of residents living within 350 m of the facility was initiated by the Mayor of Tokaimura. This decision was based on three factors: a request from JCO to evacuate people from a specific area; on a report that JCO personnel had been evacuated from near the facility and on the gamma dose rate of 0.84 mSv/h measured at the site boundary. By around 17:00, 86 persons had been evacuated, reportedly with little panic or confusion. The last person was evacuated at 20:10, making a total of 161 people from 39 house-holds within 350 m range of the JCO facility.

The information related to the first and second factors were given from JCO to Tokai-mura not only by FAX but also through face to face communication.

On the other hand information from JCO to Japanese and Ibaraki prefecture government was given only by FAX and no information from JCO to local governments adjacent to Tokai-mura. The difference in the quality and quantity of information transferred from JCO to governments caused the difference in understanding of the accident situation and the necessity of imminent emergency response measures.

(2) Information transfer from the local government to the public was often made through both indoor and outdoor radio speakers prepared by the local government. Important information through some of the outdoor radio speakers was disturbed by mass media's helicopter noises. Moreover, indoor radio speakers were not set in each school, nor in each company. Results of the questionnaire which are under preparation will be summarized at the workshop.