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Dose Evaluation Based on ^{24}Na Activity in the Human Body at the JCO Criticality Accident in Tokai-mura

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Criticality accident/Neutron dosimetry/^{24}Na/JCO/External exposure

^{24}Na in the human body, activated by neutrons emitted at the JCO criticality accident, was observed for 62 subjects, where 148 subjects were measured by the whole body counter of JNC Tokai Works.

The 148 subjects, including JCO employees and the contractors, residents neighboring the site and emergency service officers, were measured by the whole-body counter. The neutron-energy spectrum around the facility was calculated using neutron transport codes (ANISN and MCNP), and the relation between an amount of activated sodium in human body and neutron dose was evaluated from the calculated neutron energy spectrum and theoretical neutron capture probability by the human body.

The maximum ^{24}Na activity in the body was 7.7 kBq (83 Bq(^{24}Na)/g(^{23}Na)) and the relevant effective dose equivalent was 47 mSv.

INTRODUCTION

The Japan Nuclear Cycle Development Institute (JNC) was actively involved in a variety of activities as emergency response actions to the criticality accident at the uranium processing facility of JCO Co. Ltd., which was occurred at 10:35 A.M. Japanese local time on 30 September, 1999. In these activities, JNC started the direct measurement of radioactivity in the body with a whole-body counter (WBC) to a part of the JCO workers and inhabitants around the JCO-site, who were supposed to be exposed to the neutrons emitted during the criticality. A total of 148 persons were measured up until 13 October with the WBC, and sodium-24 (^{24}Na) was observed from 62 persons.

^{24}Na is produced in a body due to activation by neutron irradiation. Measurements of ^{24}Na in the body had played an important role in the dosimetry in some past criticality accidents. At a criticality accident at Oak Ridge in 1958, the ^{24}Na content in the body estimated by an assay of blood samples provided the only available information on individual doses, because there were

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no personal dosimeters[1]. In this accident, an animal (burro) was exposed to neutrons using a mock-up reactor for determining the relationship between the sodium activity in the blood and radiation dose, in which the same neutron spectrum as the accident was applied.

The effectiveness in the measurement of ^{24}Na in blood for individual dose estimations at a criticality accident is described in some references[2,3]. WBC for a ^{24}Na measurement is also describing as being more sensitive method than the assay of blood samples[4].

In the same way as in past criticality accidents, the radiation doses of a few workers were estimated directly from individual dosimeters. Therefore, JNC supported the Headquarter for Accident Countermeasures of the Science and Technology Agency (STA) and the Accident Investigation Committee of the Nuclear Safety Commission of Japan (NSC) in the individual dose assessment based measurements of the ^{24}Na activity in the bodies of the subjects.

This paper describes the measurement of ^{24}Na with WBC and a dose assessment based on the determined ^{24}Na activity.

MEASUREMENT OF ^{24}Na IN THE BODY

1) WBC system

The whole-body counter used in this study is located at the Radiological Health Service Facility of the JNC Tokai Works. This WBC is usually used for special monitoring for internal contamination of radiation workers who are engaged in the Tokai reprocessing plant and other nuclear research facilities.

The detectors of the WBC include two germanium (Ge) detectors (Type: GC 5021) manu-

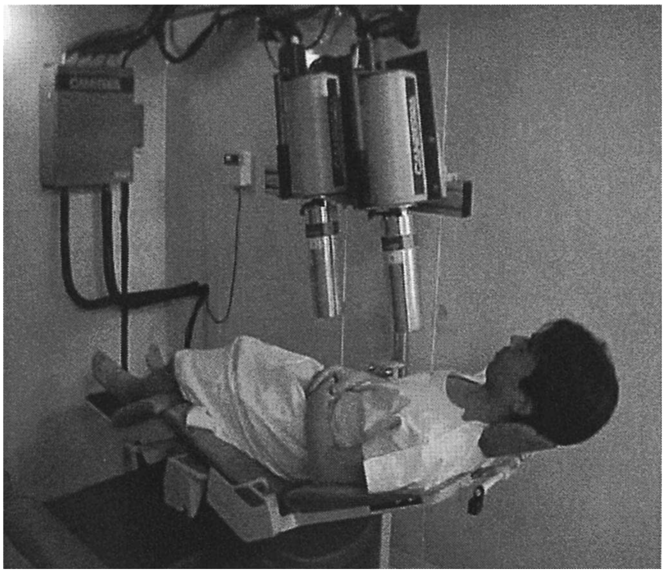

Fig. 1. JNC whole-body counting system with Ge detectors, installed in the radiation shielding container.

factured by Canberra Co. The dimensions of each detector are 3400mm^2 (window area) × 54.5mm (length). This system is installed in a container for radiation shielding, which consists of 20 cm thick iron. The inner dimension of the container are 2m × 2m × 2.5m(height × width × length).

The geometry of the WBC is chair type and the view fields of the two detectors are chest/thorax and abdomen respectively. Fig.1 shows the WBC system. This system is calibrated with an anthropomorphic phantom (170cm height) which is filled with a composite radionuclide solution (^{40}K,^{60}Co,^{137}Cs and ^{133}Ba) in an acrylic resin container. The detection limit of ^{24}Na in the body was estimated to be approximately 50 Bq for a ten-minute measurement.

2) Measurements of ^{24}Na in the body

The 1.369 MeV gamma-ray from ^{24}Na with an emission rate of 100% was measured with the WBC. A total of 148 persons were measured, and a significant activity of ^{24}Na was detected in 62 persons who are categorized into 4 groups as follows:

Group-A: 7 persons who worked at a construction materials stock yard neighboring the JCO sites; they stayed until 4:00 p.m. of 30 September at their office or outdoor, which were located about 100 m northwest from the uranium processing facility.

Group-B: 3 emergency service officers who transferred 3 severely exposed workers to Mito National Hospital by an ambulance.

Group-C1: 30 JCO employees and contractors who stayed within a 75 m (about the nearest boundary of the site to northwest direction) radius from the precipitation tank when the criticality occurred.

Group-C2: 6 JCO employees and contractors who stayed at over 75 m radius from the precipitation tank when the criticality occurred.

Group-D: 16 JCO employees who were engaged in the terminating the criticality.

Figure. 2 shows a map indicating the locations of the persons belonging to the above 4 groups during the criticality. The decrease in the ^{24}Na activity in the body was corrected with the effective half-life of 14.08 hours for ^{24}Na in the human body (physical half-life, 14.96 hours; metabolic half-life, 10 days according to ICRP metabolic model[5)]). It is supposed that the total induced activity of ^{24}Na in the body of the persons of group A and C was produced at the time when the criticality accident occurred at 10:35 A.M. of the day stayed in and around the JCO site. For those persons of group D, it is supposed that the total induced activity of ^{24}Na in the body was produced at the time when they started the work for terminating the criticality. The human body contains 1.4 g of stable ^{23}Na per 1 kg of body weight on the average[6)]. Subjects were weighed on a scale before the measurement with the WBC. Based on the weight data, the mass of stable ^{23}Na contained in the body was estimated and the specific activity (the activity of ^{24}Na per gram of stable ^{23}Na; here and after described as Bq(^{24}Na)/g(^{23}Na)) was obtained.

The measured activities of ^{24}Na in a body were in the range 0.16 kBq ~ 7.7 kBq, and thus the specific activities were in the range 1.5 Bq(^{24}Na)/g(^{23}Na) ~ 83 Bq(^{24}Na)/g(^{23}Na).

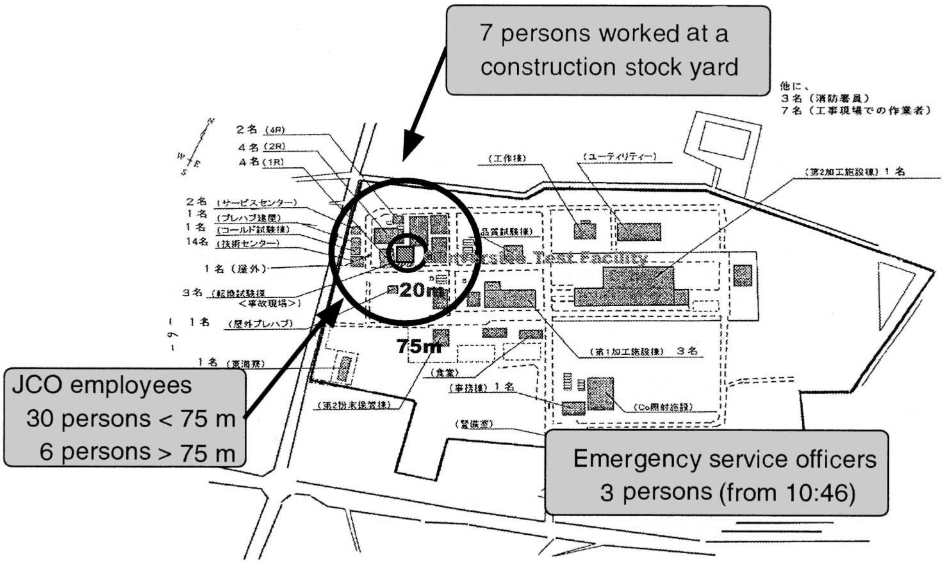

Fig. 2. Location of 46 persons at 10:35 A.M. for whom ^{24}Na in the body was detected by WBC.

PRELIMINARY ESTIMATION OF THE ABSORBED DOSE

In order to screen a subject for early medical treatment, a rapid dose estimation was necessary. Considering the IAEA report[7)], we assumed the following relation:

1Bq of ^{24}Na in total body 0.5 – 3 μ Gy (neutron + gamma rays).

Using this empirical relation, the absorbed dose of each person was estimated from the measured activity of ^{24}Na with WBC that is carried out on the day when the criticality accident occurred and reported to the Accident Investigation Committee and the related medical practitioners as a preliminary estimation. The maximum estimated absorbed dose was approximately 20mGy.

EVALUATION OF DOSE

The relation of the ^{24}Na specific activity (S) and the neutron total fluence (Φ [n/cm^2]) at the field can be expressed by following equation[8)]:

$$S = 0.61\bar{\xi}\,\Phi\ [\mu\,\mathrm{Bq}(^{24}\mathrm{Na})/\mathrm{g}(^{23}\mathrm{Na})], \quad (1)$$

where $\bar{\xi}$ is the neutron capture probability averaged over the neutron energy spectrum, and $\bar{\xi}$ can be expressed by

$$\bar{\xi} = \int \xi(E)\,\phi(E)\,dE / \int \phi(E)dE. \tag{2}$$

Here, ξ (E) is the calculated values of neutron capture probability in the anthropomorphic BOMAB phantom,[2] and ϕ (E) is the neutron energy spectrum [n/cm^2/eV].

At the same field, the neutron effective dose equivalent (H [Sv]) can be expressed by

$$H = \int h(E)\,\phi(E)\,dE \text{ [Sv]}, \tag{3}$$

where h(E) is the fluence to the dose equivalent conversion coefficient [Sv·cm^2].

The fluence to the effective dose equivalent conversion factor and the fluence to 1cm depth dose equivalent conversion factor are described in ICRP Publication 51[9]. The fluence to effective dose conversion factor is described in ICRP publication 74[10].

Equation (3) can be changed to

$$H = \bar{h}\Phi \text{ [Sv]}, \qquad (3)'$$

where,

$$\bar{h} = \int h(E)\,\phi(E)dE / \int \phi(E)dE.$$

Therefore, using equation(1) and equation(3)', H can be expressed as

$$H = (\bar{h} / 0.61\,\bar{\xi})\,S \text{ [Sv]}. \tag{4}$$

If the exposed neutron energy spectrum can be calculated or measured, the effective dose equivalent, the 1cm depth dose and the effective dose can be evaluated from the ^{24}Na specific activity by using equation(4). For a simple description, we define the dose conversion factor (K) in relation to the specific activity as

$$K = 0.61\,\bar{\xi} / \bar{h} \text{ [}\mu\text{Bq(}^{24}\text{Na)/g(}^{23}\text{Na)/Sv]}. \tag{5}$$

The neutron energy spectra within a range up to 500 m from the precipitation tank in which the criticality occurred was calculated using the ANISN code[11] (Multigroup One-Dimensional Discrete Ordinates Transport Code System with Anisotropic Scattering). A 40 liter solution of uranium nitrate in the precipitation tank was approximated by a sphere with a radius of 21 cm. The water-cooling jacket (2.2 cm thick), precipitation tank structural materials (SUS3mm, 3mmx2), the conversion building outer wall (10 cm thick lightweight foam concrete,0.5g/cm3) and the air were taken into consideration in this calculation. Other buildings and equipments were excluded from the consideration because of details, such as the structure, were not clear. The neutron energy spectra around the facilities was also calculated[12] using the MCNP-4B code[13] (Monte Carlo N-Particle Transport Code System for Multiparticle and High Energy Applications).

The calculated neutron energy spectra using ANISN and MCNP-4B at reference points 1.8 meters (outside of the conversion building) and 75 meters (about the nearest boundary of the site) from the center of the precipitation tank are shown in Fig. 3. The estimated mean capture probability ($\bar{\xi}$) based on a calculation using the ANISN code was almost constant and in the range of 0.25 – 0.28 at any distance from the center of the precipitation tank.

Figure. 4 shows the estimated conversion factor (K) defined by equation (5) as a function of the distance from the precipitation tank. The conversion factor (K) based on the calculation of ANISN code is lower than that based on MCNP-4B code at all distances.

Table1 gives the ^{24}Na specific activity per effective dose equivalent which was adopted for

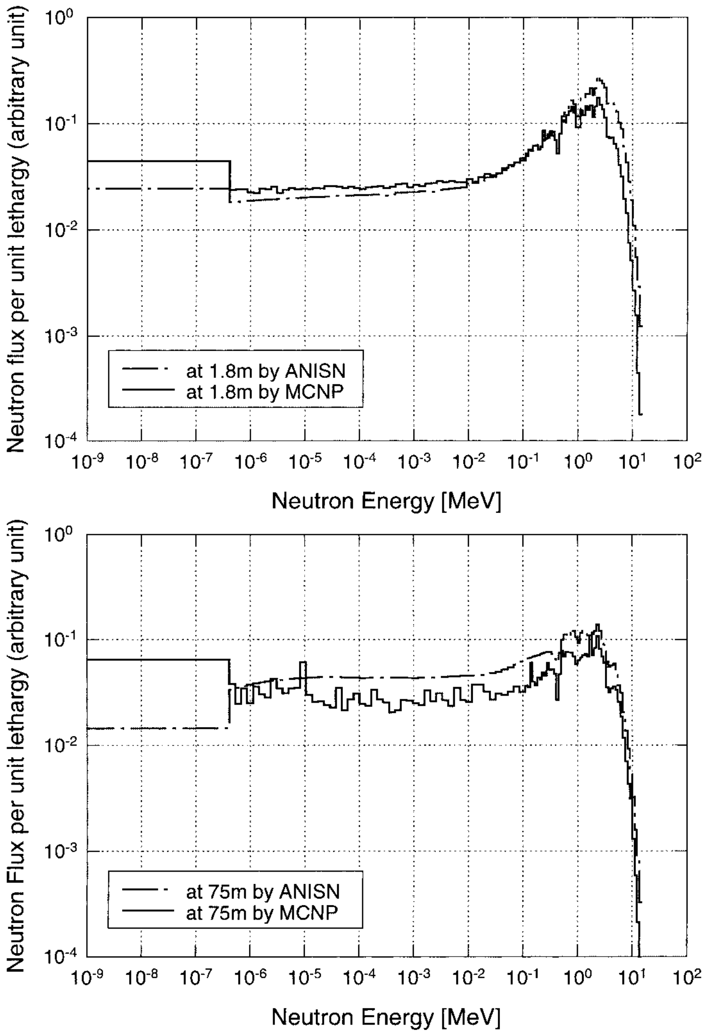

Fig. 3. Neutron energy spectrum calculated with the ANISN code and the MCNP code.

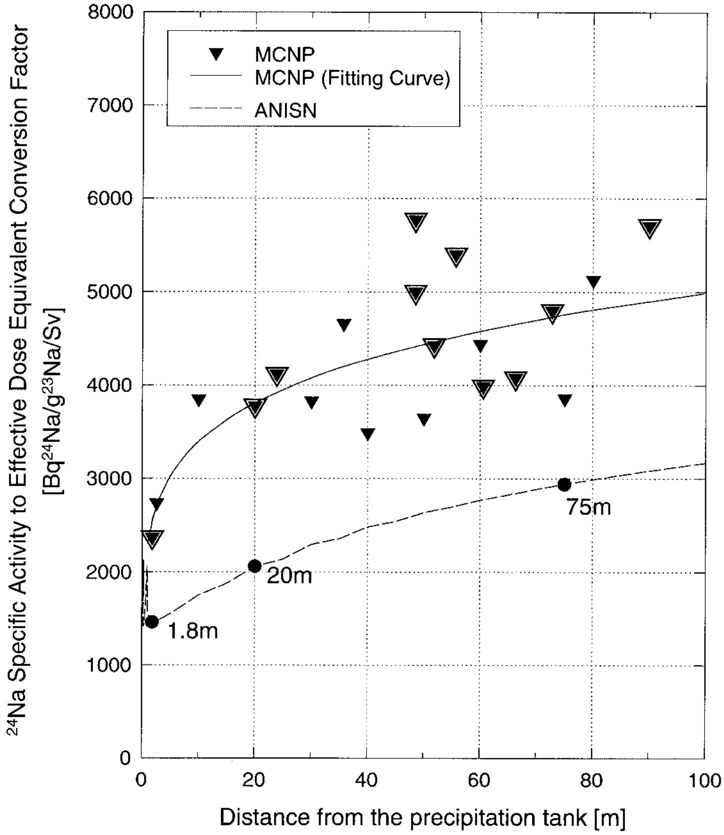

Fig. 4. Conversion factor (K) estimated using equation (5) as a function of the distance from the precipitation tank. Large ▼ marks corresponded to places where the subjects were exposed at 10: 35 a.m.

Table 1. ^{24}Na specific activity per unit effective dose equivalent at the reference points.

(AP geometry*).

Range of distance from the precipitation tank (m)		Distance from the precipitation tank to the reference point for derived conversion factor at each area (m)	^{24}Na specific activity per unit effective dose equivalent (Bq(^{24}Na)/g(^{23}Na)/Sv)
Area I	< 20	1.8	1460
Area II	20 ~ 75	20	2060
Area III	>75	75	2940

* Antero-posterior geometry: The irradiation geometry in which the radiation is incident on the front of the body in a direction orthogonal to the long axis of the body.

an individual dose estimation, where the applicable range of each conversion factor (K) was divided into three areas, such as within 20m radius from the center of the precipitation tank (area I), 20m~75m radius (area II) and over 75m radius (area III). The categorized groups in 2–2) were classified by the area based on their staying places during the criticality, as follows:

1. Group-A: Classified in area III,
2. Group-B: Classified in area II,
3. Group-C1: Classified in area II,
4. Group-C2: Classified in area III,
5. Group-D: Classified in area I.

The individual dose due to a photon was evaluated based on the neutron dose evaluated from ^{24}Na using the ratio of the neutron dose to the photon dose.

The ratio of 1-cm depth dose equivalent for a neutron to 1-cm depth dose equivalent for the photon was set at 10:1 (n:γ), based on the measured values with survey instruments[14] (the neutron rem counter and the ionization chamber) in and outside of the JCO site when the criticality continued. Table 2 gives the results of an evaluation of the individual effective dose equivalent expressed by a range with the sum of the neutron and gamma doses.

Table 2. Results of evaluating the individual effective dose equivalent

Group	Number	Range of effective dose equivalent (gamma + neutron) (mSv)
Group-A (neighboring inhabitants)	7	6.7 ~ 16
Group-B(Emergency service officers)	3	4.6 ~ 9.4
Group-C1 and Group-C2 (JCO employees and contractors)	36	0.6 ~ 47
Group-D (JCO employees who engaged in the termination of criticality)	16	5.9 ~ 45

DISCUSSION

1) Uncertainties of the evaluation

The major factors affecting the uncertainty in the evaluation of individual doses are summarized below:

(a) Uncertainty in the neutron spectrum calculation

The neutron spectrum calculated by the ANISN code does not include the moderated (scattered) neutrons by the ground surface and the shield materials, except for the conversion building walls. In actual conditions, as the distance from the precipitation tank becomes greater, the neutron spectrum becomes more moderated. Thus, as the specific activity increases, the dose-equivalent value tends to decrease. In this calculation, the effective dose equivalent for neutrons tended to be overestimated, particularly for those who were in Areas II and III. The maximum degree of this over estimation is estimated to be about 50%.

(b) Statistical and systematic deviation in whole body counter measurements

There was a statistical deviation of about 5% (1 σ) for counting the subjects for whom the largest ^{24}Na activity was detected. The systematic error caused by the geometrical instability of the human body setting in WBC and the discrepancy between the human body and the calibration phantom was supposed to be within 10% based on a consideration that the counting efficiency for the 1.369 MeV photon emitted from ^{24}Na changed within 10% when the distance from a body surface to the detector is changed ±5 cm from standard position.

(c) Difference of individual ^{23}Na body content

The ICRP reference man contains 1.4g of ^{23}Na per 1 kg of weight. It was described in a reference[15)] that ^{23}Na in a human body is in a range of 0.9~1.6g per 1kg of weight. If the actual human body contains ^{23}Na lower than the reference value of 1.4g/kg, the ^{24}Na specific activities become an under estimation.

(d) Neutron-to-photon dose ratio

It is supposed the ratio of 1cm depth dose equivalent for neutron to 1cm depth dose equivalent for photon is 10:1. This ratio changes depend on the distance from precipitation tank, and then different doses may be obtainable but not so changeable.

2) Comparison of Evaluated Dose to the Results of the Personal Dosimeters

The JCO employees who were engaged in operations to terminate the criticality attached

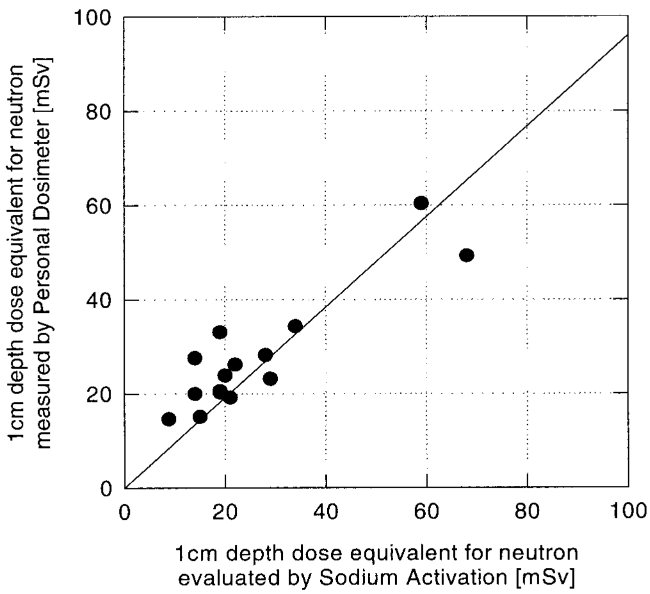

Fig. 5. Comparison of the 1cm depth doses equivalent evaluated from the ^{24}Na specific activity and that with personal dosimeters.

electric personal neutron dosimeters, the PDM-303 manufactured by Aloka Co. The relation between the 1cm depth doses equivalent evaluated from the ^{24}Na specific activity in the body and that for the personal dosimeters is shown in Figure5. The Japan Atomic Energy Research Institute (JAERI) implemented a calibration of those personal dosimeters as the post accident investigation using the Transient Experimental Critical Facilities (TRACY), of which the neutron energy spectrum in radiation field was similar to that in the JCO accident[16)]. The 1cm depth dose equivalent for neutrons measured by the personal dosimeter in figure5 adopted the corrected value based on this calibration.

CONCLUSION

The effective dose equivalent for the 62 persons were evaluated based on the ^{24}Na activity measured with the WBC. The maximum ^{24}Na activity in the body was 7.7 kBq (83 Bq(^{24}Na)/g(^{23}Na)) and the relevant effective dose equivalent was 47 mSv. The evaluated doses have a good relation with the measured dose with personal dosimeters.

These results can be used to investigate the effects on the health of the workers and residents together with findings obtained by other means. It was also proven that ^{24}Na counting with WBC is one of the effective methods for individual dose assessment at a criticality accident because this method is prompt and relatively more sensitive than the blood-analysis method.

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