

Measurements and Calculations of Beta Dose Rates on Contaminated Ground at the Fukushima Daiichi Nuclear Power Plant Site

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Abstract. The accident at the Fukushima Daiichi Nuclear Power Plant resulted in a substantial release of radionuclides into the environment. Consequently, the emergency operation workers were exposed to both external gamma and beta radiations arising from the fallout; however, their beta doses were scarcely recorded at the time due to an insufficient number of dosimeters.

To solve this problem, the authors performed Monte-Carlo simulations which estimated the gamma and beta dose equivalent rates in outdoor work environments during the initial weeks of the accident and allowed the personal beta exposure to be reconstructed from the individual gamma doses recorded. The computer model consisted of air-ground interface, and the person receiving the dose was assumed to be standing on uniformly contaminated ground. The source term used was based on the isotopic compositions of ^{129m}Te , ^{132}Te - ^{132}I , ^{131}I , ^{134}Cs , ^{136}Cs , ^{137}Cs - ^{137m}Ba , ^{140}Ba - ^{140}La , etc., all identified in soil by HPGe spectroscopy analysis. The calculated results were benchmarked by comparison with both the dose rates measured for contaminated soil samples collected from the site and those measured by stationary and portable instruments on-site in mid-March. The computed beta-to-gamma dose ratio ranged from ~10 at 50 cm to ~3 at 130 cm above the ground. The maximum possible beta exposure to the unprotected skin of the worker, engaged in outdoor operations, was estimated to be ~1 Sv, with the expectation of further dose reduction reflecting the effects of ground surface roughness and protective clothes.

Keywords: Fukushima Daiichi Nuclear Power Plant, emergency operation workers, beta rays, skin dose, Monte-Carlo calculation

1. Introduction

The accident at the Fukushima Daiichi Nuclear Power Plant in March 2011 resulted in a substantial release of radionuclides into the atmosphere and caused extensive contamination of the environment. The released fission products were principally noble gases and volatile elements (iodine, tellurium and cesium), the latter of which were deposited on the ground surface and created a harsh radiation field of gamma and beta rays. Hundreds of emergency workers were devoted to an attempt to keep the situation under control and were consequently exposed to high external dose rates. In the course of the initial plant stabilization operation, a few tens of workers received external gamma doses exceeding 100 mSv [1]. By contrast, there has been no report on the external beta exposures to the workers, except for a case in which two workers waded in highly contaminated water and thus suffered contact beta exposures to the

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skin of their legs [1,2]. This absence of reports is simply due to the fact that the doses of external beta rays were scarcely monitored due to the limited number of beta dosimeters available and operable at the time. As reported elsewhere, however, a high beta dose several times greater than the gamma dose was not unusual in radiation fields resulting from reactor accidents, and in some cases the beta dose was a primary safety concern for the workers [3-5]. This paper describes a dosimetry method which intended to reconstruct the gamma and beta mixed radiation fields in the outdoor environment at the site, which were not well defined at the time after a series of initial radiological releases, by using computational dosimetry techniques. The paper also intends to provide data applicable to assess the workers' skin dose of beta rays.

2. Measurements

Measurements were made of the beta and gamma dose rates on the surfaces of soil samples collected on 22-23 March at several locations within 1 km from the reactor Units 1 and 2. These were initially brought by the Tokyo Electric Power Co., Ltd. (TEPCO), the plant operator, to the Japan Atomic Energy Agency (JAEA) for the purpose of germanium spectroscopy and radiochemical analysis. These three samples taken in the vicinity of the reactor buildings – respectively 500 meters north, west and south – were chosen for the dose rate measurement and then each packed into a plastic container of 13 cm in diameter × 5 cm in depth and weighed. The major radionuclides identified and their activities at the time of sampling are listed in Table 1, a part of which was already published on TEPCO's website [6]. The observed radionuclides were ^{129m}Te , ^{132}Te - ^{132}I , ^{131}I , ^{134}Cs , and ^{137}Cs - ^{137m}Ba , with minor contributions of ^{106}Ru , ^{136}Cs and ^{140}Ba . The activity ratios of ^{134}Cs to ^{137}Cs were 1.0, 1.0 and 0.4 for the north, west and south samples, respectively. In contrast, the $^{131}\text{I}/^{137}\text{Cs}$ ratios were 3.1, 17, and 40 for the north, west and south samples, respectively, and correspond to 8, 40, and 100 when decay-corrected to March 12, i.e., the day of the initial primary containment vessel (PCV) venting at Unit 1. Substantial differences in the $^{131}\text{I}/^{137}\text{Cs}$ ratios were observed north to south across the site.

Table 1 Major radionuclides found in soil samples and their concentrations

Sample ^a	Density (g/cm ³)	Concentration ^b (Bq/cm ³)					Sampling date
		^{129m}Te	^{132}Te	^{131}I	^{134}Cs	^{137}Cs	
North	1.6	1.4E+3	3.0E+3	5.0E+3	1.5E+3	1.6E+3	Mar-22, 2011
West	1.3	3.3E+2	7.9E+2	7.5E+3	4.4E+2	4.4E+2	Mar-21, 2011
South	0.8	7.6E+0	1.7E+1	6.3E+2	7.0E+0	1.6E+1	Mar-22, 2011

^a Soil samples from north, west and south of the reactor buildings were collected from the ground near a solid waste storage facility (500 m north of the stack of Unit 1), an athletic field (500 m west-northwest), and a storage yard (500 m south-southwest), respectively.

^b Top soil to a depth of 5 cm was assumed to be collected; therefore, the deposition density (Bq/cm²) used for the calculation (described in 3.2) was estimated by multiplying the concentration by 5 cm.

The beta and gamma dose rates at the surfaces of the soil samples were measured with an air

ionization chamber (AIC), Oyo-Giken Model AE-133B. This AIC has a shallow collecting volume of 10 mm in depth with a thin and large window (diam. ~100 mm), being essentially the same design as the national standard laboratory's transfer instrument for the beta dose-rate standard [7]. The entrance window of the AIC was set 5 cm above the top of the soil. The chamber output was repeatedly measured by a digital voltmeter placed in parallel with the meter circuit and then average readings were taken. The beta and gamma dose components were discriminated with measurements with and without an 8-mm acrylic filtration placed on the window. The measurements were repeated several times from 1 April through 10 May for a rough estimate of the short-lived isotopes present. The prior calibration of the AIC with the Amersham-Buchler standard beta calibration sources presented nearly energy- and angular-independent responses for ^{90}Sr - ^{90}Y and ^{204}Tl , with a slight (~20%) under-response for ^{147}Pm .

3. Calculations

Given the fact that no on-site data of beta doses or dose rates were obtained during the initial weeks of the accident, the beta exposure for the workers must be based on computations. Two Monte-Carlo computation models were therefore created to benchmark the above-mentioned measurements, and to extend them to reconstruct the on-site radiation fields, allowing the workers' exposures to be estimated. To do this, the MCNP-4C code [8] was selected because of its ability to simulate any 3D geometry.

3.1 Model-1: Benchmarking of the measurement

The first computer model was aimed to validate the Monte-Carlo calculation results by comparison with the measurement results; therefore, it included a plastic container filled with uniformly contaminated soil, a concrete floor and surrounding air. The elemental compositions of soil and air were taken from the literature while the measured density of the soil was used. Each isotropic source of ^{106}Rh , $^{129\text{m}}\text{Te}$, ^{131}I , ^{132}Te - ^{132}I , ^{134}Cs , ^{136}Cs , ^{137}Cs - $^{137\text{m}}\text{Ba}$, ^{140}Ba - ^{140}La , etc., all identified by HPGe spectrometry, was designated in the SDEF card as being uniformly distributed throughout the soil region.

Continuous beta energy spectra for ^{131}I , ^{134}Cs , ^{137}Cs and ^{140}Ba were taken from the table in Appendix D of ICRU Report 56 [9]. The spectra for the other nuclides were calculated using the program SOURCE, a minor modified version of the subroutine SOURCE of the BETABREM code [10]. Discrete internal conversion electrons of 624 and 656 keV for ^{137}Cs were also considered. Photon energies and relative abundances, although only those > 1% were chosen, were as given in the Table of Isotopes.

A disc of soft tissue with the same outer dimension as the chamber was placed with its front face 5 cm above the soil. Energy depositions at depths of 0.07 and 10 mm were calculated by an energy-modified F8 photon-electron tally and then converted to the tissue absorbed doses at equivalent depths. The dose result in units of Gy/h per Bq/cm³ for each radionuclide was summed over all the nuclides with their composition and concentrations in soil.

3.2 Model-2: Uniformly deposited on the ground

The first model was spatially extended to calculate the height variation in beta and gamma dose rates over flat open ground on which the measured radionuclide mixtures were uniformly deposited. In the second model, a person was assumed to be standing on contaminated soil at the air-ground interface, as illustrated in Figure 1(a). The elemental compositions of soil and air were again taken from the literature [11] and their densities were taken to be 1.6 g/cm^3 and 1.2 mg/cm^3 , respectively.

Isotropic sources uniformly distributed to finite depths of 0.1 mm and 1 mm in the soil were assumed. These depths were chosen to account for relatively small attenuation due to ground surface roughness, since most of the exposure occurred during the first few weeks and the deposited radionuclides were expected to be fresh, not having penetrated deeply into the soil. A shallow source of 0.1 mm would apply to ideally flat areas. The radionuclides as the source term were identical to those used in model-1. In addition, short-lived ^{133}I ($T_{1/2}=21 \text{ h}$), although not identified by gamma spectroscopy, was also taken into consideration with the estimated initial inventory of $^{133}\text{I}/^{131}\text{I}=2$ on the day the reactors halted.

The dose equivalents to various sites on a standing person would vary with the height above the ground. Therefore, they were calculated in a manner which allowed the variation in doses with height to be obtained in a single computer run. Figure 1(b) depicts an air right cylinder region, which defines the virtual body surfaces and tallies used for the spectral fluence and current of gamma and beta radiations. For beta particles, the spectral current tally (F1 tally) with both cosine multiplier and segment cards was used to score particles entering into the side of the cylinder from the outside. The incident beta particle was then segregated into angular bins of $0\text{-}30^\circ$, $30\text{-}60^\circ$, $60\text{-}75^\circ$ and $75\text{-}90^\circ$ according to the angle of incidence, where the reference vector (0°) corresponded to the normal incidence from the horizon. Besides, a set of segment cards made up of planes parallel with the ground divided the above-mentioned tally with an increment step of 10 cm in height. These energy- and angular-spectral fluences were folded with energy- and angular-dependent dose conversion coefficients [12], $H_p(0.07)/\Phi$ to obtain the beta personal (directional) dose equivalent at a depth of 0.07 mm. As for gamma-rays, while similar procedures were used to obtain the personal dose equivalent at a depth of 10 mm, the angle-integrated fluence at the surface was also folded with angular-independent (or angle-averaged) dose conversion coefficients [12], $H^*(10)/\Phi$ and $E(\text{ROT})/\Phi$, to obtain the ambient dose equivalent and effective dose in the rotational exposure geometry. In addition, to account for the fact that the personal (directional) dose equivalent from beta-rays depends on the direction considered or the worker's orientation and that most of the beta particles come from below, the beta particles were also scored with disc-shape surface tallies created as intersections between the cylinder and a set of horizontal planes with different heights. The reference vector (0°) here corresponded to the normal of the ground. The calculation results in units of Sv/h per Bq/cm^2 for each radionuclide were scaled to the total radioactivities of three soil samples from the north, west and south and each was compiled as a function of time after the disaster.

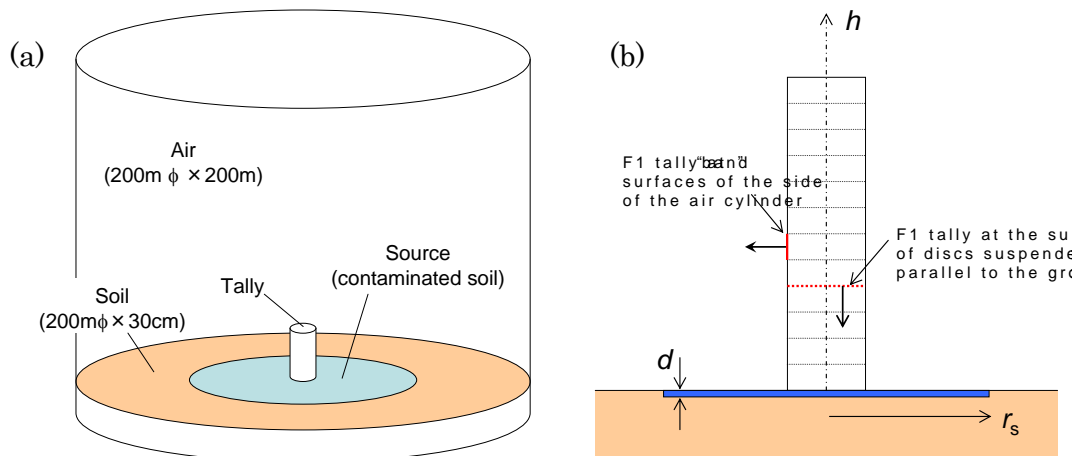


Figure 1. MCNP calculation geometry, showing (a) the whole view of the air-ground interface and (b) the tally used in the calculations. The arrows shown in (b) represent the reference orientations of F1 tallies, and particles coming from the opposite side are disregarded in the calculation of both $H_p(0.07)$ and $H_p(10)$. The source radii of r_s are 20 m for beta sources and 100 m for gamma sources, with the source depths of $d=0.1\text{mm}$ and 1mm .

4. Results

Figure 2 shows the measured beta and gamma dose rates 5 cm above the surface of the north soil sample against the number of days following the accident. Also plotted are the calculated directional dose equivalent rate for beta rays and the ambient dose equivalent rate for gamma rays, both of which are expressed as lines of decay curves. The agreement between the experimental and calculated results was satisfactory, thus lending credibility to the validity of further transport calculations for beta and gamma rays in the soil-air interface geometry. Most of the dose rates came from ^{132}I during the initial week, followed by ^{131}I . The beta-to-gamma dose ratio near the surface was ~ 2 as a result of 5-cm-deep contaminated soil, which was caused by stirring during repacking of the soil samples.

The variations in the beta and gamma dose rates and the beta-to-gamma dose ratio with height are shown in Figures 3 and 4 for major dose contributors of ^{131}I , ^{132}Te - ^{132}I , ^{133}I , ^{134}Cs , and ^{137}Cs - $^{137\text{m}}\text{Ba}$. All the plots are normalized to $1\text{Bq}/\text{cm}^2$ of the parent nuclide. For distributed sources like fallout, the curves of the dose rates depend not on the inverse square relationship with distance but on air attenuation. The graph shows that the beta dose rate for ^{131}I with a maximum beta energy of 0.606 MeV decreases by a factor of ~ 100 when the height is increased from 0 to 100 cm. In contrast, the beta dose rate for ^{132}I with a maximum beta energy of 2.14 MeV decreases by at the most a factor of 4 in the same height range, thus dominating the beta exposure to the worker's body off the ground. Because the gamma dose rate is less dependent on height although the beta radiation is reduced with increasing height, the beta-to-gamma dose ratio becomes relatively smaller as the height increases. Moreover, a comparison of the beta dose rates between the source depths of 0.1 mm and 1 mm demonstrated that the beta dose rate was very sensitive to the actual depth distribution. Dose rate reduction between the two depths

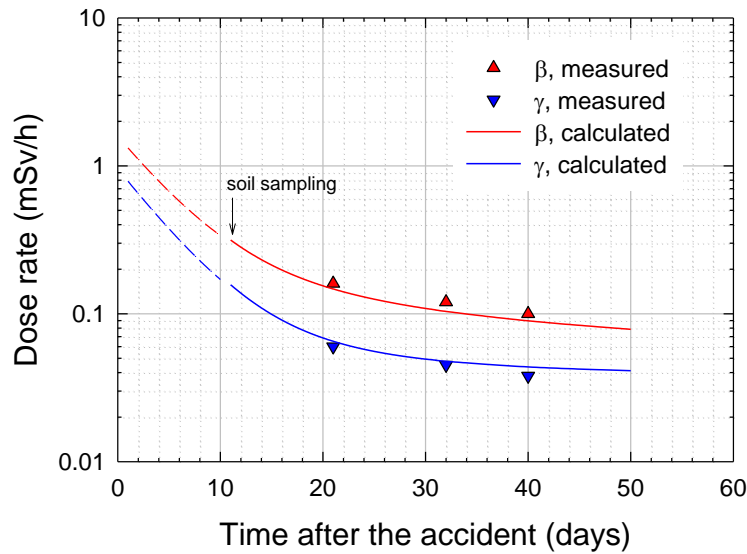


Figure 2. Measured and calculated gamma and beta dose rates 5 cm above the surface of the north soil sample plotted against the number of days after the accident. Dashed curves were extrapolated by simple decay corrections.

was by a factor of ~ 5 for ^{131}I and ~ 2 for ^{132}Te - ^{132}I . It is also noteworthy that beta doses showed a dependence on the orientation of the tally by a factor of ~ 2 .

Figure 5 presents the early time course of the ambient dose equivalent rates of gamma rays measured at 1 m above the ground on-site and off-site. Plotted are readings of: temporary stationary monitors at the front and west gates of the site, a temporary stationary monitor in front of the administration building adjacent to the quake-proof building (in which TEPCO's response headquarters was established) [13], portable survey instruments used for the purpose of dose rate mapping around the reactor buildings [14], and an off-site monitoring station at Okuma-town, located ~ 5 km west of the plant [15]. In addition to the measurements, gamma ambient dose equivalent rates calculated for three soil samples are also shown for purposes of comparison. These rates were simply decay-corrected back to the day the disaster occurred, since the actual time-dependence of the deposited radionuclides was unclear. The overall trend and peaks of the measured dose rates from 12 through 16 March corresponded to a series of PCV-venting operations and an accidental explosion around the suppression chamber of Unit 2. The radiation fields appeared to have been built up by the ground deposition of fallout until the afternoon of 16 March; since then the measured dose rates followed the decay curve with a half-time of a composite of ^{131}I and ^{132}I . It is considered that the gamma dose rates, although their absolute values were slightly different, had been able to be sufficiently simulated to reconstruct the outdoor radiation environments around the damaged reactor buildings.

The topical beta dose rates, calculated in this paper at the surface of and 1 m above the ground, are also exhibited in Fig. 5. The curves calculated only for the north soil sample with the assumption of uniform 0.1-mm-deep deposition on the ground are drawn to avoid excessive overlapping of the plots. The calculation results revealed that there had existed an intense beta

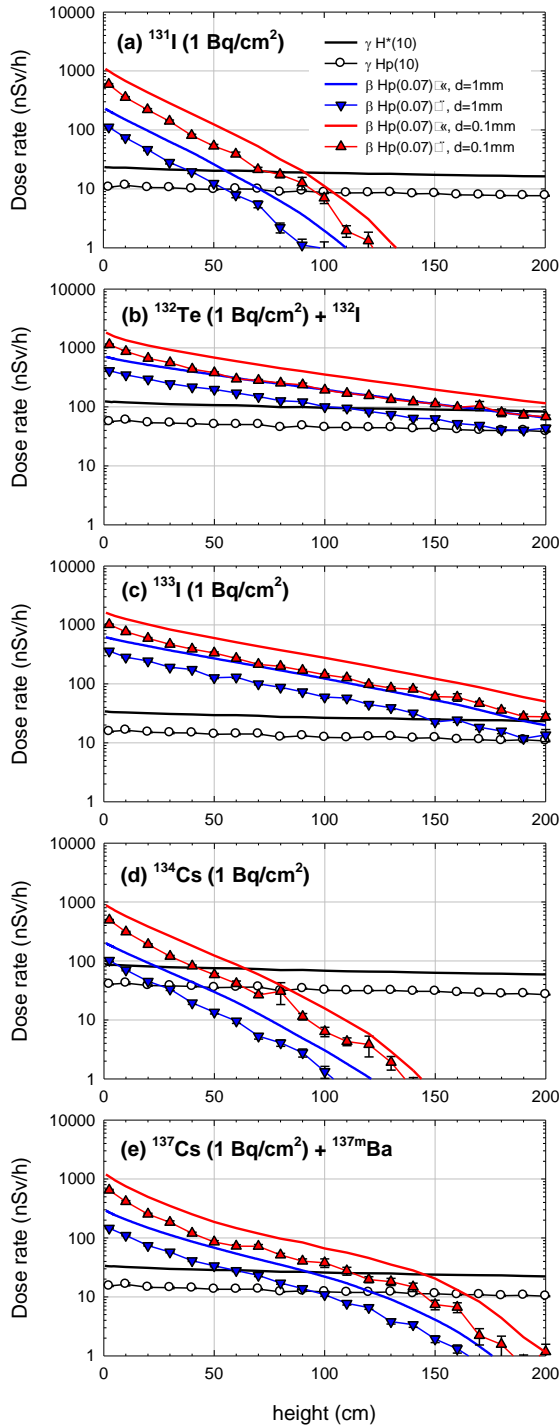


Figure 3. Variation in beta and gamma dose rates with height from the ground for major dose contributors of (a) ^{131}I , (b) ^{132}Te - ^{132}I , (c) ^{133}I , (d) ^{134}Cs , and (e) ^{137}Cs - $^{137\text{m}}\text{Ba}$. Arrows (\downarrow , \rightarrow) in the caption represent the reference orientation of the FI tally in the space to calculate angular-dependent $H_p(0.07)$ and $H_p(10)$.

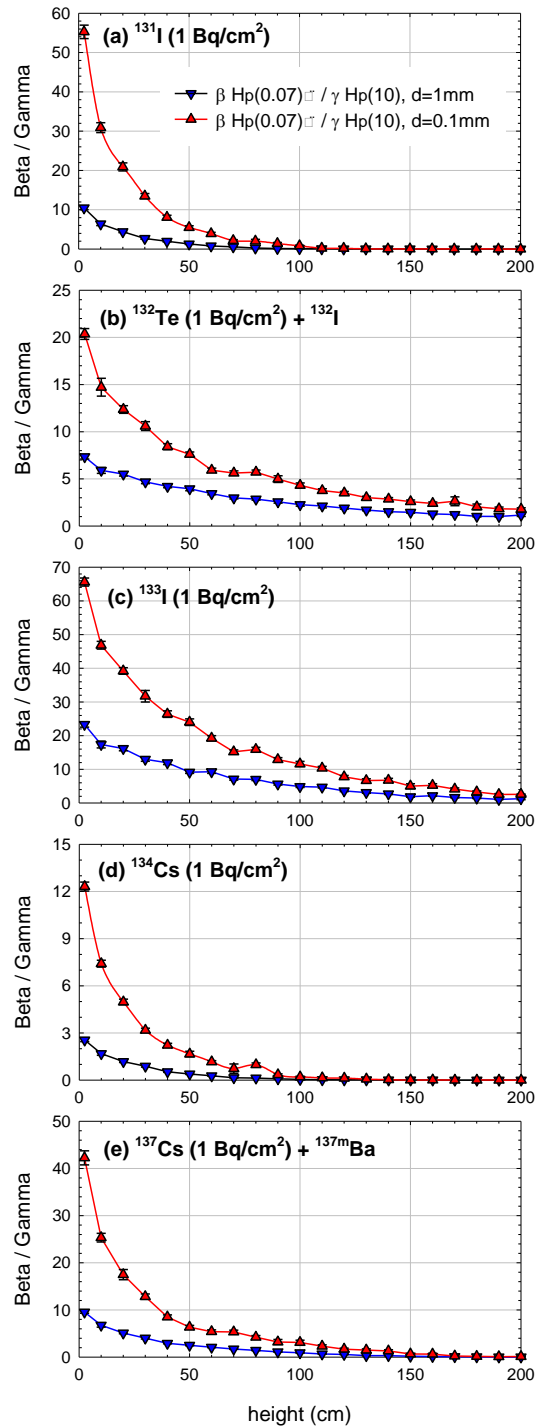


Figure 4. Variation in beta-to-gamma dose ratios with height from the ground for major dose contributors of (a) ^{131}I , (b) ^{132}Te - ^{132}I , (c) ^{133}I , (d) ^{134}Cs , and (e) ^{137}Cs - $^{137\text{m}}\text{Ba}$.

field in which it was likely for the workers to have experienced beta exposures higher than the gamma exposures. The beta dose rates at 1 m on March 16, deduced from the March 22 gamma measurements around the reactor building, were estimated to have been on the order of 100 mSv/h.

5. Beta exposures to the workers

The Japanese government relaxed the annual limit on the occupational effective dose from 100 mSv to 250 mSv under emergency conditions [1], while the limit on the skin equivalent dose of 1 Sv was not raised. Consequently, to ascertain whether the skin dose was four times (=1000/250) greater than the effective dose is important both to comply with the dose limits and to avoid deterministic health effects.

Figure 6 presents the time-variation in the ratios of the beta doses at various heights to the gamma dose at 130 cm above the ground on which radionuclides were uniformly deposited to a depth of 0.1 mm. The ratio allows the estimate of beta doses at any location on a worker's body from the gamma dose expected to be recorded with the worker's personal dosimeter [16]. It is evident from the graph that partial beta exposures to the lower body exceeding 4 times the gamma dose may be imparted.

According to the TEPCO, the maximum worker's gamma exposure as of the end of March 2011 was reported to be ~200 mSv; however, how much of the dose resulted from outdoor work was not disclosed. In contrast, the maximum gamma exposures to members of the Tokyo Fire Department and the Self Defense Force, who were engaged in dousing the damaged plant, were reported to be 27 and 81 mSv, respectively, as a result of exclusively outdoor operations [17].

Based on these facts, we assumed here that the maximum possible gamma dose which had been received in the outdoor work was 100 mSv. By use of the ratios of 3 and 10 extracted from Fig. 6, the maximum possible beta doses received were estimated to have been 300 mSv at 130 cm (chest level) and 1,000 mSv at 50 cm (knee level), respectively. These dose estimates were, however, significantly conservative values neglecting the beta dose reduction due to both the surface roughness of the actual ground/paved areas and the protective clothing worn by the workers. The former effect might be roughly accounted for by the beta dose rate reduction of up to a factor of 3 found between the source depths of 0.1 mm and 1 mm. As for the latter, a worker was generally required to wear (his) normal working clothes, an anti-contamination suit and gloves, etc., and, if required, an anorak [1]. An assumed combined mass thickness of these outfits of more than 50 mg/cm² would have reduced the beta skin dose by a factor of ~2. With these considerations in mind, the estimated maximum beta dose to protected skin was expected to be far below the skin dose limit of 1 Sv.

6. Summary

The accident at the Fukushima Daiichi Nuclear Power Plant resulted in a substantial release of radionuclides into the environment, producing harsh radiation fields to which the emergency operation workers were exposed. The authors performed a computer simulation which estimated

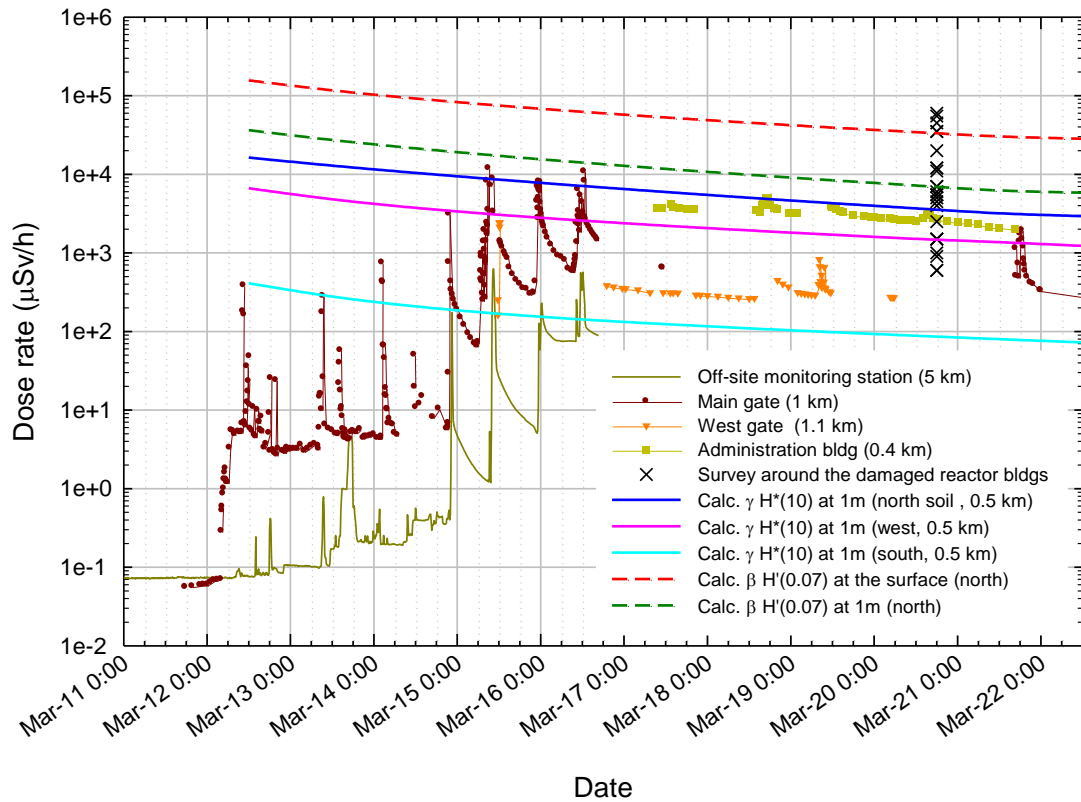


Figure 5. Early time course of dose equivalent rates on-site and off-site. Plotted are the gamma dose rate readings of an off-site monitoring station [14], on-site temporary stationary monitors [12], and survey instruments [13]. The calculated gamma and beta dose rates, based on the soil samples, are also presented in bold solid and dashed lines, respectively.

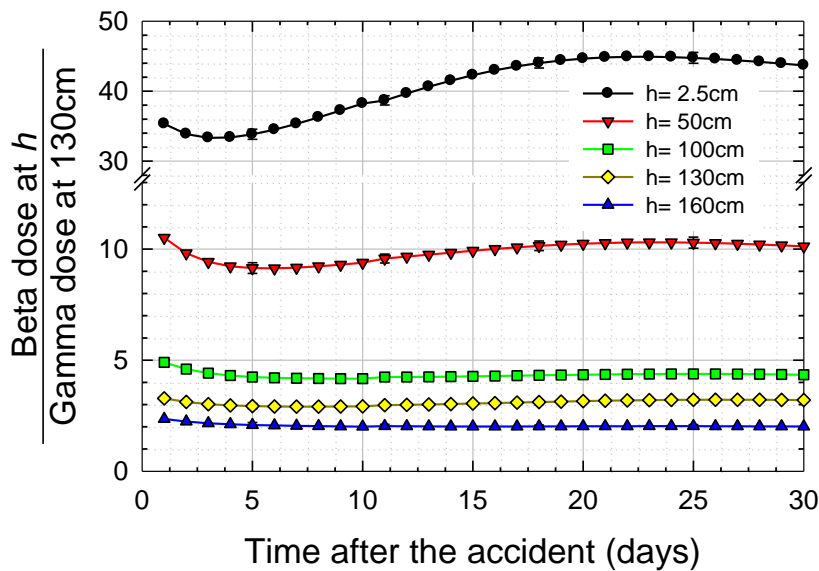


Figure 6. Time-dependence of the ratios of the beta doses at various heights (h) to the gamma dose at 130 cm above the ground. The plots are for uniform deposition of 0.1 mm in depth with the same isotopic compositions as in the north soil sample.

the gamma and beta dose equivalent rates in the outdoor work areas during the initial weeks of the accident and allowed the individual beta exposure to be reconstructed. As a result, it was concluded that the maximum possible beta dose to workers engaged exclusively in outdoor operations would not exceed the skin dose limit of 1 Sv. The dose estimates were, however, conservative and preliminary in the sense that the computer model did not reflect the effects of actual protective clothes appropriate to each group of workers. Further simulations that account for the effects of protective clothes are in progress.

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