Integrated Technique for Assessing Environmental Dose of Radioactive Waste Storage Installation

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INTRODUCTION

The capability to accurately predict exposure rates at large distances from a gamma radiation source is becoming increasingly important. This is because that the related regulation for the control of radiation levels in and around nuclear facilities becomes more stringent. Since the continuous increase of the radwaste storage capacity requirement on site, the requirement of a more realistic evaluation is very necessary. The environmental dose could be of course calculated by using rigorous transport and/or Monte Carlo methods despite time consuming. However, in the routine design work some simplified and dedicated computer codes, e.g. the point kernel code – QAD series(1) and the integral of line-beam response function code – SKYSHINE-III(2) as well as the recently developed SKYDOSE and McSKY(3), were normally used for saving computing time. To apply these dedicated codes on the problem of environmental dose evaluation, some simplifications need to be done firstly. It could be considered in two stages. The first stage is to determine the spectrum and strength of the source from the radwaste. It was at past time determined by using the SPECTUM(4) code, which was used to calculate the beta and gamma spectrum from a given set of isotopes and executed on IBM mainframe. The newly developed SPECTRUM-506(5), which is the enhanced version of SPECTUM with some features and ported to PC platform, is used instead of SPECTUM in this paper.

The second stage is dose evaluation. The total dose at detecting point could be considered in two components: direct and skyshine dose. In the aspect of direct dose estimation, it was usually at past time evaluated by the point kernel code - QADCG/INER-2(6), which is a revised version of QAD-CG(7). The buildup factors applied in QADCG/INER-2 were calculated by Capo’s(8) formula and for 5 materials only. As fitting function developing by time, the G-P (Geometric Progression) fitting functions were determined at Japan Atomic Energy Research Laboratory and Tokyo Institute of Technology in 1986(9). The G-P fitting function reproduces the basic buildup factor data over the whole range of energy, atomic number, and shield thickness within a few percent and is the fitting function of choice for point kernel calculations. Due to the advantages of the buildup factor calculated by G-P fitting function and in order to improve the accuracy of the dose estimation, it is reasonable to include the use of the G-P fitting function for the gamma-ray buildup factor of the QADCG/INER-2. Hence QADCG/INER-3 (10) was developed to replace QADCG/INER-2. Furthermore, for the dose of shielding design problem evaluated by the QAD series codes with a certainly conservative consideration have been well known, but for the dose of far-field detection point haven’t. Therefore the accuracy of far-field detection point direct dose evaluated by the point kernel code was investigated in this study.

In the aspect of skyshine dose estimation, it was usually at past time evaluated by the SKYSHINE-III code. This dedicated skyshine code deals with point sources and applies the integral line-beam method for the calculation of skyshine dose. In the conventional estimation of the skyshine dose, the volume source encountered normally in the practical engineering problem is first converted into equivalent point source by the point far away from the source using the QAD-CG point kernel code. The equivalent point source obtained by this way is assumed to be isotropic and in most cases treated as mono-energetic. According to the previous studies(11,12), the accuracy of the conventional estimation of skyshine dose rates is very poor and hard to be evaluated and it could be improved by representing the equivalent point source in terms of the photon emission rate out of the surface of the volume source. This equivalent point source, in consequence of transport of photons in the source region, shall bear some energy and angular distribution even though the volume source is isotropic and mono-energetic. Therefore the revised SKYDOSE and McSKY with the capability of dealing with anisotropic and multiple energetic point source were decided to replace of SKYSHINE-III. It has been shown that the accuracy of the dose evaluation for direct and skyshine could be improved by using these newly developed and revised codes, respectively. Finally, this paper will propose an integrated technique for assessing environmental dose of radwaste storage installation by using these dedicated codes to consider direct and skyshine contribution.
MATERIALS AND METHODS

The reference problems in the American National Standard ANSI-6.6.1(13) were investigated for the verification of the revised codes (e.g. QADCG/INER-3, SKYDOSE and McSKY) and the integrated dose assessing methodology we proposed. The arrangement of the reference problems are described as follows:

(a) **ANS-6.6.1 reference problem I.1**
This problem is for a point isotropic source of \(^{16}\text{N}\) gamma rays (6.2 MeV) with a source strength of 1 photon/s. The point source is positioned at a height of 60 ft (18.3 m) above the air-ground interfaces. The detectors are positioned at horizontal ranges between 200 ft (61 m) and 5000 ft (1,500 m) from the normal-to-the-ground surface through the point source and at a height of 3 ft (0.91 m) above the grade. Figure 1(a) shows the graphical representation of the problem configuration.

(b) **ANS-6.6.1 reference problem I.2**
A \(^{16}\text{N}\) point source with strength of 1 photon/s is positioned at a height of 60 ft (18.3 m) within an open roof building having a 4-ft thick concrete wall on all four sides. Height of the concrete wall is taken to be 62 ft (18.9 m) and the inside dimension of the structure are 100 ft (30.5 m) by 150 ft (45.7 m). Detectors are positioned at horizontal ranges between 200 ft (61 m) and 5000 ft (1,500 m) from the normal-to-the-ground surface through the point source and at a height of 3 ft (0.91 m) above the grade. Figure 1(b) shows the graphical representation of the problem configuration.

(c) **ANS-6.6.1 reference problem II.1**
This problem is for a cylindrical tank, 12-ft (3.7 m) in diameter and 35 ft (10.7 m) high, containing water with a uniformly distributed source of 0.8 MeV photons. The volumetric source strength is assumed to be 30 MeV/cc-sec with the tank wall neglected. Detectors are positioned at a height of 3-ft (0.91 m) above the grade and at horizontal ranges up to 1000-ft (305 m) form the axis of the tank. Figure 1(c) shows the graphical representation of the problem configuration.

(d) **ANS-6.6.1 reference problem II.2**
This problem has the same geometry and source distribution as that described above for problem II.1 except that a 35-ft (10.7 m) high annular concrete shield surrounds the tank with a 2-ft (0.61 m) thickness. Figure 1(d) shows the graphical representation of the problem configuration.

Monte Carlo code MCNP was for rigorous calculation. In the calculations, \(10^8\) photon particles were traced and the cut-off energy was 1 keV. The relative error of the calculated dose rates were less than 2 %. The photon flux densities estimated by using point detectors were then converted to the dose equivalent rate by using the flux-to-dose equivalent rate conversion factors(14).

RESULTS AND DISCUSSIONS

I. The features of SPECTRUM-506
SPECTRUM-506 is newly developed for the replacement of SPECTUM, which is used to calculate the beta and gamma spectrum from a given set of isotopes. Daughter isotopes are included by utilizing the Bateman series solution for the coupled differential equations that govern isotope decay. Buildup of daughter isotopes can
also be done by assuming a negative decay time, but this not applicable for the estimation of parent isotopes, however. SPECTRUM-506 is coded in FORTRAN language and running on PC platform. The major differences between SPECTRUM-506 and SPECTUM are as follows:

1. FIDO Input System is introduced into the program.
2. Up to 506 isotopes is included in the library.
3. The output of SPECTRUM-506 could be imported into the QADCG/INER-3 code as the source term directly.

II. Update the point kernel code - QADCG/INER-2

QADCG/INER-3 code is the latest version of INER’s QADCG/INER code series. According to the recommendation of the American National Standard Institute report ANSI/ANS-6.4.3-1991(15), the G-P (Geometric Progression) fitting function coefficients is the fitting function of choice for point kernel calculations. Hence the G-P fitting function was built in the QADCG/INER-3 for more accurate buildup factor calculations. The flux-to-dose conversion factor for photon in the ICRP-51 report was applied in this newest version.QADCG/INER-3 code still keeps the combinatorial geometry and the FIDO input method as in the previous version.

III. The verification of the QADCG/INER-3 code

The major differences between QADCG/INER-3 and QADCG/INER-2 are an addition of the newest gamma mass attenuation coefficient and geometric progression buildup factor originated from ANSI-6.4.3-1991 plus the absorbed conversion factor originated in ICRP-51 report. Hence the first step of verification is to compare the difference between the buildup factor calculated by Capo’s and G-P fitting function. In addition, some parameters with the effects on the dose evaluation in this code were also investigated in this report. Finally, the verification of QADCG/INER-3 was carried by comparison of the calculations of reference problem I.1 and II.1 of ANSI-6.6.1-1987 by Monte Carlo code MCNP(16).

III.1 The comparisons of buildup factors

The major difference of QADCG/INER-3 with previous version is an addition of using the buildup factor calculated by G-P fitting formula, and the differences between the results calculated by Capo’s and G-P fitting functions for commonly used medium, e.g. water, concrete, iron and lead, are shown in Table 1. As it shows, there are some differences between G-P and Capo’s results. For 6.2 MeV, the results of G-P are larger than Capo’s for 4 mediums within 10 mfp, except for concrete in 1 mfp. The differences of lead are relatively large than other mediums. The data of Capo’s fitting function do not include secondary sources, such as annihilation, fluorescence, and bremsstrahlung, and the contribution of this part is getting more important for higher energy source and heavier medium. Hence the results of G-P are significantly larger than Capo for 6.2 MeV in lead. For 1.25 MeV, the differences between G-P and Capo’s are small, no more than 3%, except in concrete medium. In concrete medium, the results of G-P are smaller than Capo about 11~17%. In the case of radwaste shielding design, source energy of 1.25 MeV and shielding material of concrete is in most case we encountered. As it revealed, a replace of results of G-P fitting function for concrete by Capo’s would overestimate the dose. For 0.662 MeV, the results of G-P are smaller than Capo’s in 4 mediums and it is significantly smaller in concrete.

Figure 2 shows the G-P buildup factors of 1.25 MeV in 5 mediums, e.g. water, concrete, iron, lead, and air. As it shows, the buildup factors of water and air agree very well. There was no buildup factors of air in the Capo’s data. Hence the replacement of buildup factors of air by those of water would be appropriate in the case where lack of buildup factors of air and the effect of buildup in air is significant.

![Fig. 2 G-P buildup factors of 5 mediums for 1.25 MeV photon](image-url)
### Table 1. The comparison of buildup factors calculated by G-P and Capo’s fitting function

<table>
<thead>
<tr>
<th>Photon Energy</th>
<th>Water MFP</th>
<th>Concrete MFP</th>
<th>Iron MFP</th>
<th>Lead MFP</th>
</tr>
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<tr>
<td></td>
<td>G-P</td>
<td>Capo</td>
<td>G-P</td>
<td>Capo</td>
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<tr>
<td>6.2 MeV</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>0</td>
<td>1.0000</td>
<td>1.0000</td>
<td>1.0000</td>
<td>1.0000</td>
</tr>
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<td>1.4657</td>
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<td>1.8564</td>
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<tr>
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<td>5.0119</td>
<td>5.9658</td>
<td>6.7962</td>
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</tbody>
</table>

| 1.25 MeV       |           |              |          |          |
| 0             | 1.0000    | 1.0000       | 1.0000   | 1.0000   |
| 1             | 1.9894    | 2.0525       | 1.8812   | 1.7641   |
| 2             | 3.2517    | 3.3166       | 2.9699   | 2.6912   |
| 3             | 4.7352    | 4.7803       | 4.2169   | 3.7335   |
| 4             | 6.4209    | 6.4367       | 5.6124   | 4.8816   |
| 5             | 8.2940    | 8.2793       | 7.1475   | 6.1922   |
| 6             | 10.3420   | 10.3010      | 8.8147   | 7.4710   |
| 7             | 12.5510   | 12.4960      | 10.8130  | 9.7015   |
| 8             | 14.9080   | 14.8570      | 12.1520  | 10.4190  |
| 9             | 17.4030   | 17.3770      | 14.0000  | 12.0160  |
| 10            | 20.0200   | 20.0500      | 16.6670  | 13.6900  |

| 0.662 MeV      |           |              |          |          |
| 0             | 1.0000    | 1.0000       | 1.0000   | 1.0000   |
| 1             | 2.3045    | 2.2926       | 2.0336   | 1.8817   |
| 2             | 4.2668    | 4.3115       | 3.4123   | 3.0100   |
| 3             | 6.8579    | 7.0722       | 5.0695   | 4.3212   |
| 4             | 10.0980   | 10.5900      | 7.0860   | 5.8102   |
| 6             | 18.5940   | 19.9630      | 11.9990  | 9.3174   |
| 7             | 23.8680   | 25.8480      | 14.9330  | 11.3350  |
| 8             | 29.8230   | 32.5530      | 18.1900  | 13.5290  |
| 9             | 36.4490   | 40.0940      | 21.7730  | 15.8980  |
| 10            | 43.7250   | 48.4870      | 25.6820  | 18.4430  |

*Diff(%) is calculated by \( \frac{(\text{Capo} - \text{G-P})}{\text{G-P}} \times 100% \)

### III.2 The effect of double layered shield

In the QAD series, there is only one buildup factor of medium could be selected in each calculation, but most practical problems encountered involve more than one shielding material. The behaviors of buildup factors of two layered shield were investigated by the previous studies(17,18), except air. In the point view of the dose estimation on the radwaste installation site boundary, the effect of air buildup is significant due to the large distance between source and detector. Hence, the effects of two-layer buildup factors, such as air followed by concrete and concrete followed by air, were investigated in this study. Point isotropic buildup factors at 1.25 MeV for single- and double-layered shields of air, concrete, air-concrete, and concrete-air were calculated by MCNP and plotted in Fig. 3. As shown in Fig. 3, the buildup factors of the double-layered shields of concrete followed by air show an excess over the corresponding buildup factor of the air shield, and those of the shields of air followed by concrete fall below the concrete data.

The replace of the buildup factor of the single layered shield of air by the double layered shield of concrete followed by air will underestimate the dose, particularly for far-field detection point. It is an inherent defect of applying the QAD series code on the dose estimation of the radwaste installation site boundary. Therefore the correction factors for double layered shield of concrete followed by air at several energies calculated by MCNP. Fig.4 shows the correction factors for the double layered shield of concrete followed by air, the total thickness was 25 mfp and 1st-layer thickness was 2, 5, 8, 10, and 15 mfp, at 1.25 MeV.

### III.3 The Verification of QADCG/INER-3

The dose for the reference problem of ANS-6.6.1 I.1 and II.1 were calculated by MCNP and QADCG/INER-
3. The results are plotted in Fig. 5 and Fig. 6 together with the data points calculated by other codes in ANS-6.6.1, correspondingly. As indicated in Fig. 5 and Fig. 6, the results of QADCG/INER-3 and MCNP calculations agreed each other very well and fall among data points of other calculations. The buildup factor of air was applied in these calculations of QADCG/INER-3.

Fig. 3 Point isotropic buildup factors at 1.25 MeV for single- and double-layered shields of air and concrete

Fig. 4 Correction factors for double layered shield of concrete followed by air

Fig. 5 MCNP and QADCG/INER-3 results for ANS-6.6.1 reference problem I.1
IV. The Procedure of the Estimation of Skyshine Dose

The dedicated codes for calculation of skyshine dose rate, e.g. SKYSHINE-III, SKYDOSE, McSKY, were designed in dealing with point sources, but the volume sources were encountered in most practical engineering problem. Hence it is necessary to convert the volume sources into the equivalent point source before the dedicated codes were applied. Accord to the previous study, the accuracy of the skyshine dose estimation could be improved by representing the equivalent source as those emitting photons from the surface of the volume source, and the energy and angular distribution of this source should be considered. The methodology of the equivalent point source conversion and the modified codes with capability of dealing with anisotropic and multiple energy point source, e.g. McSKY and SKYDOSE, are adopted in this report.

The skyshine doses for the reference problem of ANS-6.6.1 I.2 were calculated by MCNP, modified McSKY and SKYDOSE. The results are shown in Fig. 7 together with the data points calculated by other codes in ANS-6.6.1. As indicated in Fig. 7 that the results of SKYDOSE and McSKY calculations agreed each other very well. However, they were somewhat lower than the values of the MCNP calculation and the discrepancy grew with an increase of the distance. This discrepancy is caused by the fact that the Line Beam Response Function (LBRFs) adopted in the SKYDOSE and McSKY codes were calculated by the single-scattering method. The LBRFs become underestimated with an increase of the distance from the source as revealed by Harima et al. (19).

In routine skyshine dose rate analysis one encounters problems with volume sources instead of point source. An integral investigation of skyshine dose rates for the volume source problem in ANS-6.6.1 II.2 was, therefore, performed. The MCNP code was employed to calculate the photons escaping from the upper surface of the volume source, which were treated as the equivalent point source with energy and angular distribution positioned...
at the center of the upper surface of the tank. The revised version of the SKYDOSE code was then applied for the calculation of skyshine dose rates. Meanwhile, calculations were also made for replacing the equivalent point source with an isotropic point source of the same intensity. On the other hand, the reference problem was also fully simulated by using the MCNP code for the calculation of the skyshine dose rates.

Figure 8 shows the calculated skyshine dose rates as a function of distance. Also shown in Fig. 8 are data by using other codes(13) in ANS-6.6.1 As can be seen in Fig. 8 that the results of SKYDOSE and MCNP agreed each other very well and fell among data of other codes.

V. Integrated Technique for Assessing Environmental Dose of Radioactive Waste Storage Installation

There are two components involved in the integrated technique for assessing environmental dose of radioactive waste storage installation proposed in this report. One is the estimation of direct dose, and the other is the estimation of skyshine dose.

V.1 The Estimation of Direct Dose for Assessing Environmental Dose of Radioactive Waste Storage Installation

Fig. 9 shows the components involved in the estimation of direct dose of radioactive waste storage installation, includes the preparations of the parameters for geometry configurations, compositions of materials, detector arrangement, characteristic of source, the option of buildup factors, and the correction factors for double layered shields.

![Diagram of the procedure of assessing the direct dose of the radwaste storage installation](image)
(a) The preparation of the parameters for geometry configurations
The parameters should be prepared in this section, involves the arrangements and geometry configurations of source, shielding buildings, and detection points. The arbitrary three dimensional spatial setup could be described easily with the powerful CG(combinatorial geometry) subroutine built-in the QADCG code series, QADCG/INER-3 included.

(b) The preparation of the parameters for the characteristic of source
The parameters should be prepared in this section, involves the spatial distribution, energy spectrum and strength. The energy spectrum and strength could be imported from the newly developed SPECTRUM-506 code directly.

(c) The preparation of the parameters for the compositions of materials
The composition and type of materials should be assigned for each zone defined in the section geometry configuration. There is only one composition and type of medium could be assigned for one zone. The database of mass attenuation coefficients of photon was derived form DLC-136/PHOTX.

(d) The determination of buildup factor and correction factor
There is only one buildup factor of shielding material could be selected in each calculation, but there are two or more shielding materials involved commonly. According to the behavior of the multi-layered shield buildup factor, the selection of buildup factor depends on the distance between source and detector. In the case for the dose estimation at site boundary, the buildup factor of concrete is selected, if the distance between detector and source does is below 2-mfp of air. A replacement of buildup factor of air by buildup factor of concrete will overestimate the dose. But the buildup factor of air should be selected if the distance between detector and source has excess over 2-mfp of air. Besides, applying the correction factors for double layered shield is also recommended.

V.2 The Estimation of Skyshine Dose for Assessing Environmental Dose of Radioactive Waste Storage Installation
Fig. 10 shows the procedure involved in the estimation of skyshine dose of radioactive waste storage installation. It could be considered in two components, includes the conversion of the equivalent point source from volume source and the application of the dedicated skyshine codes, such as McSKY and SKYDOSE.

Fig. 10 The procedure of assessing the skyshine dose of the radwaste storage installation

(a) The conversion of the equivalent point source
The methodology of conversion of the equivalent point source from volume source adopted in this report is to represent the equivalent point source in terms of the photon emission rate out of the surface of the volume source. This equivalent source, in consequence of transport of the source region, shall bear some energy and angular distribution, which could be obtained by using Monte Carlo or discrete ordinate transport codes, such as EGS4/MCNP, or ANISN/DORT/TORT, etc.

(b) The application of the dedicated skyshine codes
The modified McSKY and SKYDOSE with the capabilities of dealing with anisotropics and multi-energetic source are applied in this report for the estimation of skyshine dose. The equivalent point source applied in McSKY or SKYDOSE could be obtained by using Monte Carlo or discrete ordinate transport codes.
CONCLUDING REMARKS

Owing to the requirement of more realistic evaluation of assessing environmental dose work for radwaste storage installation, those simplified and dedicated codes, e.g. QADCG/INER-2 and SKYSHINE-III applied in the routine design work are updated and accompanied with adequate methodology in this paper. These works are concluded as follows:

1. SPECTUM, which is used for calculating the spectrum and strength of the source, is updated to SPECTRUM-506 with 506 isotopes in the library.
2. QADCG/INER-2, which is used for calculating the direct dose, is updated to QADCG/INER3 with G-P buildup factors.
3. The effects of the double layered shield were also investigated in this paper. The underestimation caused by the replacement of the buildup factors of double layered shield of concrete followed by air by those of single layered shield could be corrected by introducing the correction factors proposed in this paper.
4. The methodology of the conversion of the equivalent point source and the modified codes, e.g. McSKY and SKYDOSE, are adopted for evaluating the skyshine dose in this paper.

REFERENCE