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MATHEMATICAL MODEL AND SIMULATIONS OF RADIATION FLUXES FROM BURIED RADIONUCLIDES*

Ahmad Saat
Faculty of Applied Sciences
Universiti Teknologi MARA Pahang
26400 Bandar Jengka
Pahang D.M.
Tel: 09 4663323, Fax: 09 4663343
E-mail: asaat@phg.itm.edu.my

ABSTRACT

A mathematical model and a simple Monte Carlo simulations were developed to predict radiation fluxes from buried radionuclides. The model and simulations were applied to measured (experimental) data. The results of the mathematical model showed good acceptable order of magnitude agreement. A good agreement was also obtained between the simple simulations and the experimental results. Thus, knowing the radionuclide distribution profiles in soil from a core sample, it can be applied to the model or simulations to estimate the radiation fluxes emerging from the soil surface.

Key Words: Radiation flux; Buried radionuclides

1 Introduction

In-situ gamma-ray spectrometry is important in assessing emergencies that cause environmental contamination as has been shown following the Chernobyl nuclear accident[1]. However for studies of post-accident fallout distribution[2] and migration and deposition of radionuclides[3] in soils, laboratory measurements of soil and core samples were used to determine the nuclides depth distributions. Although in-situ gamma-ray spectrometry method has been shown to be a possible approach to determine radionuclides depth profiles in soils[4,5] the uncertainties that accompanied the method are considerably high[6]. In the present study we proposed a method which work in reverse order; determination of in-situ radionuclide fluxes using depth distribution profiles in soils obtained by laboratory measurements of core samples.

The objective of the study was to determine the fraction of radiation from a distribution of buried radionuclide, as measured at some distance above ground level. Two approaches were employed, mathematical modelling and simple Monte Carlo simulations. For the study it was assumed that the distribution was composed of layers of uniform nuclide activity, parallel with and buried at a certain distance from the soil surface. The radiation was measured at a certain height from ground level. Radiation fraction from each layer was summed to give the effective fraction from the bulk deposit. The approaches were then tested using experimental data.

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2 The study

The methods used in the study will be discussed in more details in the following sections. Table 1 gives the data used in getting the results of each method. For simplicity, soil was assumed to be composed of 75% SiO_2, 20% Al_2O_3 and 5% water. The values of mass absorption coefficients for soil, μ_s were calculated using mixture rule on absorption coefficient values obtained from table by Hubbell[7]. The corresponding valuẽs for air, μ_a were interpolated from the same table.

TABLE 1. The Input Data.

Soil density (gcm^{-3})	1.6	
Air density (gcm^{-3})	1.29×10^{-3}	
Energy (keV)	μ_s (cm^2g^{-1})	μ_a ($\times 10^{-2}$ cm^2g^{-1})
50	0.298	2.073
100	0.168	1.541
200	0.128	1.234
500	0.0894	0.871
662	0.0788	0.726
800	0.0725	0.708
1000	0.0650	0.636
1500	0.0529	0.518
2000	0.0456	0.445

3 Mathematical models

In a simple model, it was assumed that 50% of the radiation were directed parallel upward from the buried layer. The infinite extent layer of radionuclide was buried at a distance *z* from the surface, and the measurement was carried out at a distance *h* above the surface. Since the absorption of a parallel beam of radiation follows exponential trends, the fraction of the flux at the measurement level will be

$$F_d/F_o = \tfrac{1}{2} e^{-\rho_s\mu_s z-\rho_a\mu_a h} \quad \gamma s^{-1}cm^{-2}/Bqcm^{-2}. \qquad (1)$$

Here, F_o is the initial flux, and F_d is the radiation flux at the measurement level.

In real situations, radiation from the buried radionuclide was directed towards all direction in space. Therefore, the law of decreasing in flux with burial depth *z* and measuring height *h* will not be a simple exponential absorption of gamma photons as in equation (1). In these situations the geometrical effect of the extent of the layer of the buried radionuclide needs to be taken into account. The radiation therefore assumed a broad beam character.

As initially suggested by Condon[8], for a broad beam of radiation entering an infinite extent of absorbing medium, the intensity at a certain point in the medium is given by

$$I = I_o \int_0^{\pi/2} e^{-\rho\mu x \sec\theta} \sin\theta \, d\theta, \qquad (2)$$

where x is the vertical depth into the absorbing medium, and I_o the intensity at the entrance.

In the modified model, we assumed the initial flux of the layer F_o is proportional to I_o and 50% of the radiation were directed 'upwards'. Thus, the flux at surface of the ground will be

$$F_l = \frac{1}{2} F_o \int_0^{\pi/2} e^{-\rho_s \mu_s z \sec(\theta)} \sin\theta \, d\theta. \tag{3}$$

To take into account the attenuation in air before reaching the measurement level, the model becomes

$$F_l = \frac{1}{2} F_o \int_0^{\pi/2} e^{-\rho_s \mu_s z \sec(\theta)} \sin\theta \, d\theta \, . \int_0^{\pi/2} e^{-\rho_a \mu_a h \sec(\theta_l)} \sin\theta_l \, d\theta_l. \quad \ldots\ldots\ldots\ldots \tag{4}$$

To evaluate the integration of Equation (4), let $u = \sec\theta$ and $u_l = \sec\theta_l$. Therefore we get $du = \sec\theta \tan\theta \, d\theta$, and $du_l = \sec\theta_l \tan\theta_l \, d\theta_l$. Substituting these into Equation (4), with the integration limits change to from 1 to ∞, we get

$$F_d/F_o = \frac{1}{2} \int_1^{\infty} u^{-2} e^{-\rho_s \mu_s z u} du \, . \int_1^{\infty} u_l^{-2} e^{-\rho_a \mu_a h u_l} du_l. \tag{5}$$

The integrals of Equation (5) known as 'Gold integrals' can be written in general form as

$$\int_1^{\infty} x^{-n} e^{-tx} dx = H_n. \tag{6}$$

The values of the integral, H_n for different t and n = 1, 2 and 3, has been tabulated[9]. Using the table, substituting n = 2, $t = \rho_s \mu_s z$ for the first integral, and $t = \rho_a \mu_a h$ for the second integral of Equation (5), F_d/F_o can be calculated by interpolation.

4 Simple Monte Carlo simulations

In the simulation code, gamma photon source points of the buried radionuclide were assumed to be distributed uniformly in a layer of radius *a* cm, positioned at *z* cm from soil surface. Since most gamma photon from radioelements cannot penetrate more than 50 cm of overburden[10], in the present study the deposition geometry is limited to that order of magnitude. Each random point position is described by three variables, *r*, θ and *z*. The length *r* is measured from the central axis of the layer, and randomised between 0 and *a*. The angle θ in the in the plane of the layer is measured from the positive x-axis, and its value is randomised between 0 and 2π[11].

Since radiation from the buried radionuclide was measured at a fixed level *h* cm from ground level, the radiation direction from each source point is described by two variables j and ϕ. The radial length j is measured from the extended symmetrical axis of the layer, while ϕ in the same manner as θ. Assuming the gamma photon travels in straight line from its source point to the detection point, the ratio of the detected flux to the initial activity per unit area will be[11]

$$(F_d/F_o)_i = e^{-\rho_s \mu_s r_{si} - \rho_a \mu_a r_{ai}}, \qquad (7)$$

where μ_s and μ_a are defined earlier. The lengths r_{si} and r_{ai} are the distances covered by the i-th simulated gamma photons from its source point through soil and air respectively. The values of r_{si} and r_{ai} were computed by simple trigonometry using the three randomised variables (r, θ and ϕ) described earlier, as well as the fixed variables z, j, and h.[11]

In the simulations, for every j value, 50,000 gamma photon source points were randomised. For every source point the flux ratio that reaches the detection level was calculated using Equation (7). They were then summed and averaged, to give the average flux ratio at distance j from the extended axis. By this way we can compute the average flux ratio at any radial distance and measuring level. The output of the simulation will list out the flux ratio at different radial distances. The present simulation code was written in FORTRAN 90.

5 Results and Discussions

5.1. Comparison between methods

The models and the simulation were applied on thin layers of buried radionuclide in soil using the input data of Table 1. Results of the study by the three methods are compared in graphs of Figure 1. In the figure the effects of burial depth on the flux ratio were plotted for different gamma-ray energies (50, 100, 200, 500, 800, 1000 and 1500 kev).

Observation on Figure 1 indicated that the simple mathematical model tends to give higher values of activity ratio than the other two methods. For each photon energy studied, the difference in values obtained by the simple mathematical model shows somewhat a constant factor, for the range of burial depth studied. This might indicates the presence of systematic error in the simple model. The higher value in the simple model is expected, since the assumption that gamma photons were directed parallel upwards would effectively give rise to a shorter average path length in the soil. Thus less absorption will take place.

Results of the simple Monte Carlo simulations are quite agreeable to those obtained by the modified model. However, at higher energy (>800 kev) deviations of a few percent are observed for smaller burial depths (<8 cm).

The three models predict that for radionuclide buried 5 cm underground the flux of low energy (50 keV) gamma photons would be reduced down to less 5% of the original. The same effect for 1000 keV and higher gamma photons would only occur if the radionuclide was buried about 25 cm underground.

In Figure 2, graphs of dependence of flux ratio detected against energy at different burial depths are shown. Plots of the results from the three methods are shown on the same graph for comparison purposes.

As mentioned earlier, it is clear from the figure that the simple mathematical model gives higher values of F_d/F_o. This is attributed to the same reason discussed above. And, again here it can be seen that the results of Monte Carlo simulations are in good agreement with those of the modified model.

Another observation on Figure 2 shows that for gamma-ray over 500 keV the change in flux ratio becomes diminutive as the energy increases, irrespective of the burial depth. However, for shallow burial depth the flux ratio is higher. This indicates that the penetration property of higher energy gamma photons in soils differs only marginally.

5.2 Application of methods on measured data

For in-situ surveys, the buried radioactive sources were assumed to be large and homogeneous. If the sources of interest are small and located away from the detector, the detector response may not indicate the true activity sought. Another point of important implication is that the area of search is increased if the detector is placed as high as possible above the ground[10]. As a consequence of this, for shallowly buried distribution the detector response would increases progressively as the detector position is increased from the soil surface.

Since no local data were available, in the study three locations in the north-west England were considered. They are South Glen, Kippford on Solway estuary, Askam marsh on Duddon estuary, and Foulney marsh, Morcambe Bay. In-situ data used were supplied by the British Geological Survey. The measurements were carried out using a 3"x3" NaI detector placed on the sediments surface, facing it. The detector was calibrated for natural series radionuclides using concrete calibration pads to give the activity ($Bqkg^{-1}$) of the measured radionuclide. Buried activity distributions were determined using estimations of the core samples data of Oldfield *et al.*[3]. The results for the respective activities at the three different locations for ^{137}Cs at 662 keV gamma photons are registered in Table 2. The fluxes ($Bqcm^{-2}$) shown in the brackets were obtained using the relation

$$\text{flux} = \text{Activity}/\mu_s, \qquad (8)$$

where μ_s is the mass absorption coefficient of the soil.

The core activity at each depth was assumed to be the initial activity of a layer of the buried radionuclide at that depth. The activities were used on the three methods described earlier. The flux from each layer measured at the soil surface was calculated, the results of which are given in Table 3. The last row of the table gives the sum of the fluxes from each layer, which is the effective flux from the distribution. This would be equivalent to the value of the in-situ measurements of Table 2.

TABLE 2. Cores and In-situ Data for ^{137}Cs of the Three Locations Considered.

	Activity, $Bqkg^{-1}$ (Flux, $Bqcm^{-2}$)					
Location	South Glen		Askam marsh		Foulney marsh	
Core depth (cm)						
2	1769	(22.463)	964	(12.241)	1098	(13.943)
6	2731	(34.679)	1779	(22.591)	2762	(35.073)
10	4731	(60.076)	3289	(41.765)	3732	(47.391)
14	4367	(55.454)	4479	(56.876)	7334	(93.130)
18	4906	(62.298)	4939	(62.718)	4166	(52.902)
22	5315	(67.492)	5796	(73.600)	985	(12.508)
26	5008	(63.594)	4393	(55.784)	969	(12.305)
30	2346	(29.791)	-	-	-	-
36	731	(9.283)	-	-	-	-
In-situ	836	(10.615)	577	(7.323)	624	(7.918)

TABLE 3. Fluxes of ^{137}Cs from Different Layers Measured at Soil Surface for Three Study Locations. (SM - simple model, MM - modified model, S - simulations).

	Expected Flux ($Bqcm^{-2}$) at Soil Surface								
Depth (cm)	SM	MM	S	SM	MM	S	SM	MM	S
	South Glen			Askam Marsh			Foulney Marsh		
2	8.737	5.803	3.327	4.762	3.162	1.816	5.422	3.149	2.057
6	8.152	3.759	3.873	8.775	2.451	2.514	8.241	2.451	2.514
10	8.521	3.302	4.787	5.930	2.286	3.327	6.730	2.603	3.771
14	4.762	1.829	2.959	4.419	1.879	3.035	7.987	3.073	4.952
18	3.225	1.092	2.184	3.251	1.092	2.197	2.743	0.927	1.702
22	2.108	0.508	1.549	2.311	0.546	1.702	0.381	0.089	0.292
26	1.206	0.241	1.003	1.054	0.203	0.838	0.229	0.051	0.178
30	0.343	0.064	0.279	-	-	-	-	-	-
36	0.051	0.013	0.051	-	-	-	-	-	-
Total	37.105	16.610	20.013	30.302	11.619	15.429	31.733	12.343	15.467

The total fluxes predicted by the modified mathematical model agree reasonably well with those obtained by the simulations. Comparing the predicted effective fluxes by the three approaches of Table 3 to the in-situ flux measurements of Table 2, shows order of magnitude agreements. However, the fluxes of in-situ measurements show consistently lower values than those obtained by the three methods. The average ratios of in-situ fluxes to predicted fluxes are 0.26, 0.64 and 0.51 respectively for the simple model, modified model and simulations. The ratios are almost independent of location. This indicates a systematic deviation between the models and simulations compared to is-situ measurements. The assumption that the density of soil is uniform (1.6 gcm^{-3} used in the study) throughout the distribution may contribute to this deviation. In the real situation this may not be true. The core samples were dried before their activity were measured. While for in-situ activity measurements, the soil was in its natural state.

Another possible source of this deviation is the data used in the study. It was assumed that the radionuclide layer distributions are homogeneous throughout the location studied. Though the in-situ and cores data were taken on the same location, the exact spots were different. It is well known that due to various physical and chemical processes in the soil, the homogeneous assumption rarely appropriate.

On top of the systematic deviations mentioned earlier, uncertainty in determination in cores activity also contributes to the error in the establishment of the expected flux using the three approaches. The core data were secondary in nature, extracted from graphs of ^{137}Cs distribution profiles in a publication by Oldfield *et al.*[3]. The uncertainty comes from both the values of distribution depths and activities extracted. Since the fluxes were related exponentially to the distribution depths, variation of 1 cm in the value of distribution depth would contribute an error of more than 10% to the expected fluxes calculated. Another 5 to 10% error is expected in the values of the radiation activities. However, despite the less favourable results obtained when the approaches were applied to experimental data, the methods were useful for estimating the expected activity due to buried radionuclides. The modified mathematical model and the simulation agree reasonably well to one another.

6 Conclusions

The study has shown that the modified mathematical model and the simulations can be used to make an initial prediction of the fluxes emerging from buried radionuclides. However, the depth distribution profile of the buried nuclied need to be predetermined.

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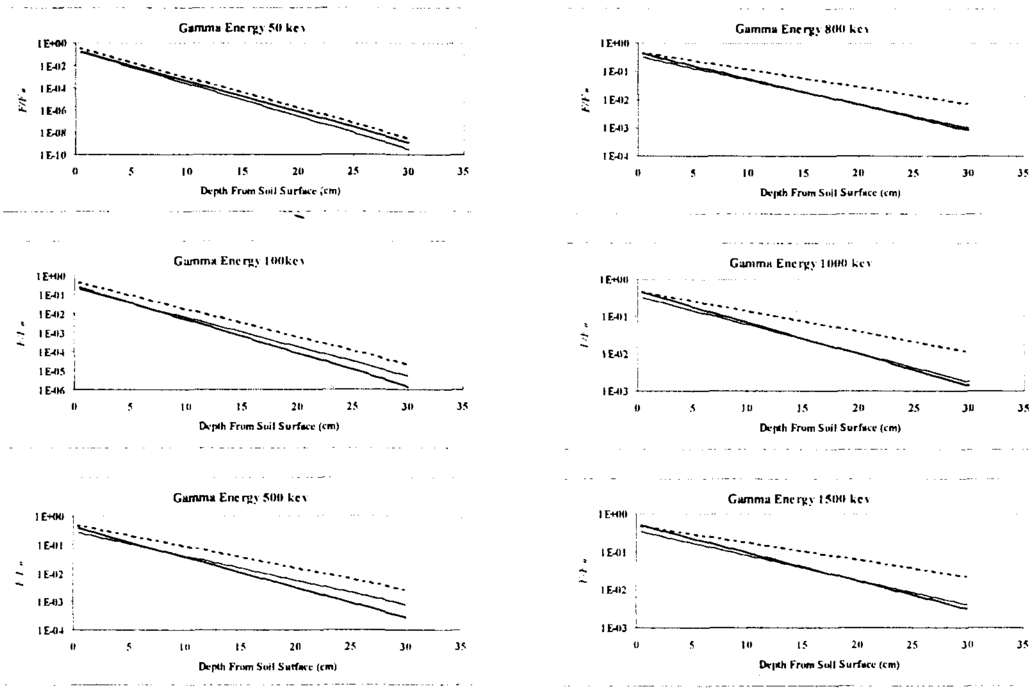

FIGURE 1. Ratio of Radiation Flux at 100 cm from Soil Surface, to the Flux of a Layer of Radionuclide at Different Burial Depths, for Different Photon Energies. Dotted Lines by Simple Model, Solid Lines by Simulations and Thin Lines by Modified Mathematical Model.

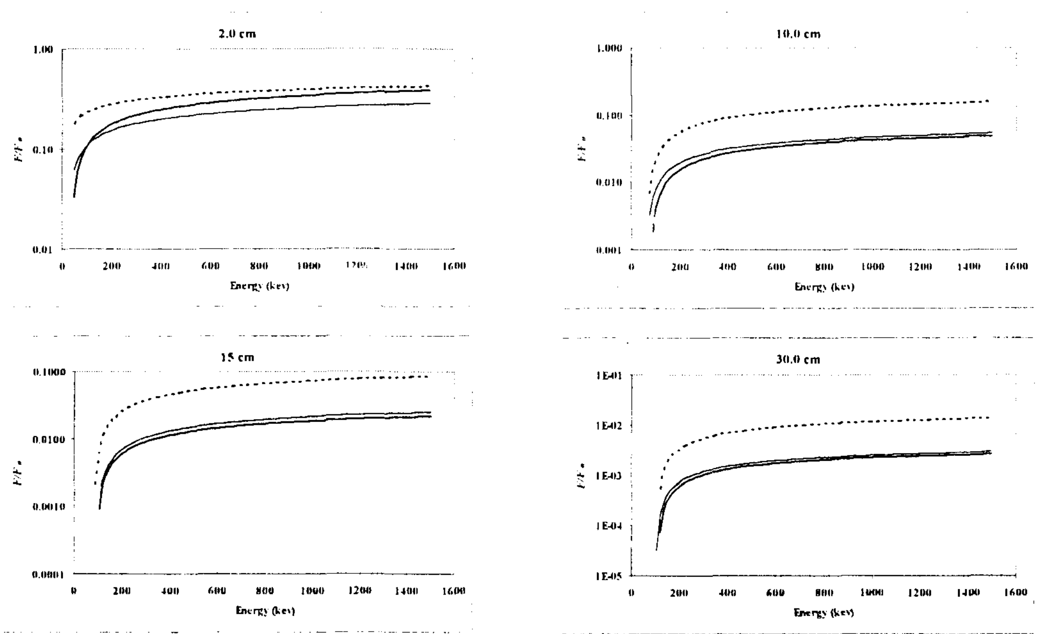

FIGURE 2. Ratio of Radiation Flux at 100 cm from Soil Surface, to the Flux of a Layer of Radionuclide at Different Photon Energies, for Different Burial Depths. Dotted Lines by Simple Model, Solid Lines by Simulations and Thin Lines by Modified Mathematical Model.