

EXTERNAL EXPOSURE TO RADIONUCLIDES ACCUMULATED IN SHORELINE SEDIMENTS WITH AN APPLICATION TO THE LOWER CLINCH RIVER

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Abstract—A simple analytical method was developed to estimate external doses from exposure to contaminated riverine shorelines. The method consists of deriving an adjustment factor that accounts for the geometry of the riverine shoreline; the adjustment factor is applied to the dose-rate coefficients already available for infinite contaminated surfaces. Such a geometry factor circumvents very complex radiation transport calculations which would otherwise be necessary to model exposures to a finite contaminated surface. For instance, for radionuclides emitting gamma rays of energies above 600 keV (e.g., ^{137}Cs), the published dose-rate coefficients must be reduced by 75%, 60%, 50%, and 33% for shoreline widths of 4, 10, 20, and 50 m, respectively. The geometry factor changes only mildly with the energy of the gamma radiation. This property allows for the geometry factor to be used for radionuclides emitting multiple gamma rays of various energies. If a quick analysis is desired, the geometry factors derived for ^{137}Cs can be used for all radionuclides. More refined analysis can be performed by deriving geometry factors for each radionuclide according to its gamma spectrum. Also, the mild variation with energy allows the geometry factors to be applied to the case when radionuclides are accumulated in layers under the soil surface, and not only to the case when radionuclides are deposited onto the soil surface. Empirical relationships between the geometry factor and the dimension of the shoreline were provided so that one can obtain values of the geometry factor for any shoreline width. These relationships can be easily used to account for the uncertainty in the dimension of the shoreline. The method was applied to derive similar adjustment factors for contaminated surfaces of other simple geometries (e.g., circular surfaces). An example of how this method can be applied to its full extent is presented for the case of external exposure to the shores of the lower Clinch River. This river received large amounts of ^{137}Cs , ^{60}Co , ^{106}Ru , ^{95}Zr , ^{95}Nb , ^{144}Ce , and ^{90}Sr released during 1944–1991 from the Oak Ridge Reservation in Oak Ridge, Tennessee.

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INTRODUCTION

WHEN RADIOACTIVE materials are released into river waters some radionuclides attach easily to particulate matter, and they may become deposited in the sediments, either on the bottom or on the shores of the river. An individual standing or walking on the banks of the river would be externally exposed to radiation emitted by radionuclides that have been deposited directly on the shoreline. The magnitude of exposure will depend on the amount and type of radioactivity deposited in the sediment, the depth of the contaminated sediment layer, and the size of the shore at the location where exposure takes place.

Radiation doses from external exposure to a contaminated ground surface are generally obtained using pre-calculated dose-rate factors. Such coefficients are available for an infinite plane (Sv s^{-1} per Bq m^{-2}) or an infinite contaminated slab (Sv s^{-1} per Bq kg^{-1}) (Kocher 1983; Kocher and Sjoreen 1985; Jacob et al. 1988a, b; Eckerman and Ryman 1993; Eckerman and Leggett 1996). Different properties of such dose-rate factors and, in general, of the doses from external exposure to gamma radiation have already been studied. O'Brien and Sanna (1977) discussed the effect of the male-female body size differences on the absorbed dose-rate distributions in humans. Jacob et al. (1988a, b) investigated the simple and non-trivial time dependencies of the organ dose equivalents after a single deposition of radionuclides. Chen (1991) analyzed the effect of soil density and soil thickness on the effective dose equivalents. Xu et al. (1995) studied the angular dependence of effective dose equivalent from broad parallel beams of photons of given energies.

This paper describes a simple method to estimate radiation doses from external exposure to finite contaminated areas, such as riverine shores. The method consists in using a multiplicative reduction factor that is applied to the dose-rate coefficients estimated for infinite contaminated surfaces. Similar reduction factors are given

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by Eckerman and Ryman (1993) based on U.S. NRC (1977) recommendations (0.1 for discharge canal banks, 0.2 for riverine shorelines, 0.3 for lake shores, and 0.5 for ocean shores). The reduction factors presented here can be applied for shores of any sizes, and they can be used to account for the uncertainty about the width of the shoreline, and the change of the width of the shoreline with time.

This method was applied to assess the effects of external exposure to the shores of the lower Clinch River, which has been contaminated with radioactive materials released from the Oak Ridge Reservation during 1944–1991. The Oak Ridge Reservation (ORR) was established in eastern Tennessee in 1942 as part of the Manhattan Project. One part of the ORR, the Clinton Laboratory (later known as the X-10 site and now as Oak Ridge National Laboratory; ORNL), was designed to operate for one year as a pilot plant for the Hanford, Washington, plutonium production facilities. All radioactive wastes generated from this facility were to be stored in large underground “gunite” tanks. The original plans changed and, in 1944, the first radioactive effluents from the X-10 site entered White Oak Creek and flowed into White Oak Lake, which served as a final settling basin. From White Oak Lake, large amounts of ^{137}Cs , ^{60}Co , ^{106}Ru , ^{95}Zr , ^{95}Nb , ^{144}Ce , and ^{90}Sr were eventually released into the Clinch River at Clinch River Mile 20.8 (CRM 20.8; see Fig. 1).

In July 1991, the State of Tennessee initiated the Oak Ridge Health Studies sponsored by the U.S. Department of Energy (DOE) with the focus on assessing any adverse health effects induced by hazardous materials released from ORR since its creation. One task of the study was the investigation of the potential health effects from radioactive materials released into the Clinch River, including external exposure to contaminated shoreline sediments. This paper summarizes the radiation doses and risks of cancer from external exposure obtained using the above method.

METHODS

To estimate the doses for individuals exposed to contaminated ground surfaces, the time-integrated radionuclide concentration in soil is multiplied by a dose-rate factor:

$$D_i = DRF_{ext} \times \int C_S(t) dt, \quad (1)$$

where

C_S = concentration of a radionuclide in the soil (Bq m^{-2}); and

DRF_{ext} = external dose-rate factor (Sv y^{-1} per Bq m^{-2}) defined as the dose received during the period of exposure by an individual standing on a contaminated area having a unit concentration.

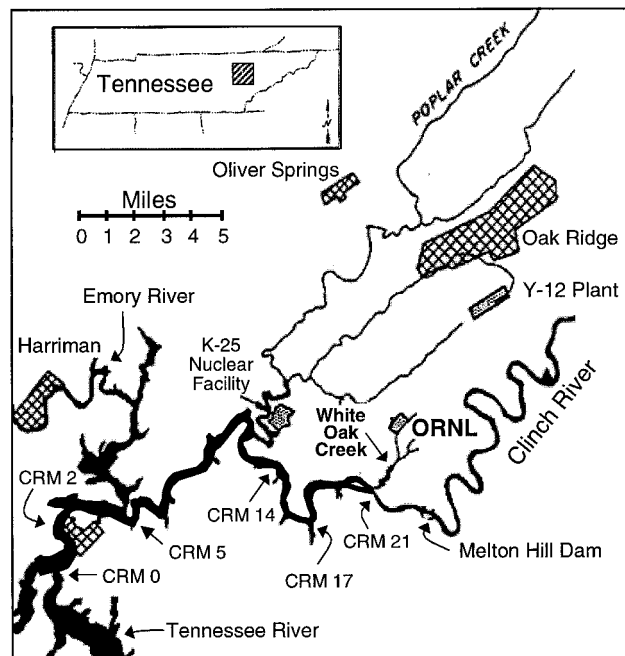


Fig. 1. Map of the area adjacent to the studied portion of Clinch River. For analysis purposes, the lower portion of Clinch River, starting from the release point to the confluence with Tennessee River, was divided into five segments or reaches: Clinch River Mile (CRM) 21 to CRM 17 (Jones Island area), CRM 17 to CRM 14 (Grassy Creek area), CRM 14 to CRM 5 (K-25 nuclear facility area), CRM 5 to CRM 2 (Kingston Steam Plant area), and CRM 2 to CRM 0 (City of Kingston area).

If the contaminated ground surface is finite, a multiplicative adjustment factor (G) can be introduced to account for the geometry of the contaminated area:

$$DRF_{ext} = DRF_{ext}^{inf} \times G, \quad (2)$$

where DRF_{ext}^{inf} = dose-rate factor for an infinite contaminated surface (Sv y^{-1} per Bq m^{-2}).

The dose-rate factors are available for exposure from an infinite plane or an infinite slab of contaminated ground surface (Kocher 1983; Kocher and Sjoreen 1985; Eckerman and Ryman 1993; Eckerman and Leggett 1996).

A riverine shore can be approximately described as a rectangular area. In most cases, the dimension parallel with the river of this area (shoreline length) is larger than three times the mean free path in air of the highest energy gamma radiation emitted by the contaminants (Table 1), and it can be, therefore, considered infinite for radiation transport purposes. On the other hand, the dimension perpendicular to the river, corresponding to shore width, usually does not exceed distances comparable with the photon mean free path in air.

In this study, a geometry adjustment factor (G) accounting for the finite geometry of the shore was calculated as the ratio of the dose to an individual standing in the center of the rectangular surface, to the dose to the same individual from an infinite surface, when surfaces

Table 1. Mean free path of gamma radiation and the empirically derived fit coefficients (a and b) for the Berger formulation for the radiation buildup in air (Chilton et al. 1984).

Energy (MeV)	Attenuation coefficient (μ) (m^{-1})	Mean free path in air ($1/\mu^a$) (m)	Coefficients for the buildup factor ^a	
			a	b
0.1	1.78×10^{-2}	56.1	5.93	0.113
0.661 ^b	9.67×10^{-3}	103.4	1.815	0.049
0.75 ^c	8.79×10^{-3}	113.8	1.795	0.0483
1.252 ^d	6.93×10^{-3}	144.2	1.38	0.028
3.0	4.30×10^{-3}	232.7	0.75	0.005

^a The buildup factor is defined using Berger formulation (eqn 5).

^b Energy of the gamma radiation emitted by $^{137}\text{Cs}/^{137}\text{Ba}$.

^c Energy representative of the gamma radiation emitted by $^{95}\text{Zr}/^{95}\text{Nb}$.

^d Average energy of the gamma radiation emitted by ^{60}Co .

are contaminated at the same concentration. For a photon of a given energy, this factor can be approximated by the ratio of the "free-field" gamma ray fluxes:

$$G = \frac{Dose_{finite\ surface}}{Dose_{infinite\ surface}} \cong \frac{Flux_{finite\ surface}}{Flux_{infinite\ surface}}, \quad (3)$$

where $Flux$ = the free-field flux from a finite and an infinite surface, respectively (cm^{-2}).

The fluxes are calculated at a point 1 m above ground in a gamma-ray "free-field." The contamination of the surface is assumed to be uniform (S_A , Bq m^{-2}). Attenuation in air and "buildup"[§] due to the photons scattered by air are considered. Similar point-kernel methods for estimation of fluxes have been employed by Kocher (1983) and by Kocher and Sjoreen (1985).

For a given photon energy, the differential flux at the point of interest is given by

$$dF = \frac{B(\mu r)e^{-\mu r}}{4\pi r^2} \times S_A dA, \quad (4)$$

where

dF = differential flux (Bq m^{-2});

$B(\mu r)$ = empirically derived buildup factor (unitless);

$S_A dA$ = differential plane source term, considered as a point source (Bq);

μ = attenuation coefficient (m^{-1} ; Table 1);

r = the distance from any differential source to the point of interest P (m);

$r = \sqrt{x^2 + y^2 + h^2}$, where x , y are the Cartesian coordinates of a point source; and

$h = 1$ m above the ground surface.

The Berger formulation was used to describe the buildup factor:

$$B(\mu r) = 1 + a\mu r e^{b\mu r}, \quad (5)$$

[§] Buildup = an increase in the flux of photons at a given location because of the contributions of the photons scattered by the air.

where a and b are empirically derived, unitless parameters which depend on the photon energy (Table 1; Chilton et al. 1984).

The radiation flux at the point of interest is computed as follows:

$$Flux = 4 \int_0^\infty \int_0^{\frac{w}{2}} dF \times dx \times dy, \quad (6)$$

where w is the width of the area, and the factor of four arises because the integral is over one-fourth of the shoreline. For the infinite plane, an analytical expression of the flux was used (eqn 7; Chilton et al. 1984):

$$Flux = 2\pi S_A \left[E_1(\mu h) + \frac{a}{1-b} \cdot e^{-(1-b)\mu h} \right]. \quad (7)$$

In eqn (7), $E_1(x)$ is the exponential integral function of order 1 and all other variables have been defined above. For the finite rectangular area, the integration was performed numerically in the Cartesian coordinate system using the Mathematica^{||} software package (Wolfram 1993.) Similar geometry factors can be derived for contaminated areas of shapes other than rectangular.

RESULTS

The geometry factors were derived for a number of photon energies and for shore widths ranging from 2 m up to 600 m (Table 2). Fig. 2 displays the geometry factors for a width up to 100 m, which is more representative as an upper bound for a riverine shore. The lowest energy (0.1 MeV) and the highest energy (3.0 MeV) represent the lower and upper limits for the energy of the gamma rays emitted by most of radionuclides of interest. As shown in Table 2 and Fig. 2, the geometry factor is rather insensitive to photon energy. For energies over 0.6 MeV, the geometry factors for a given width of the contaminated area are practically the same. For lower energies, differences are noticeable. The geometry factor for an energy 0.1 MeV is lower than the geometry factor for 1.3 MeV by 20% to 30% for shore widths of 2 to 10 m, by 10% to 20% for shore widths of 10 to 30 m, and by 1% to 10% for shore widths of 30 to 75 m. As a general observation, for shore widths less than 75 m, the lower the energy the lower the geometry factor. On the other hand, for shore widths larger than 100 m, the geometry factor is higher for lower energies (Table 2). The lower the energy, the lower the shore width for which the dose-rate factors for an infinite surface are reached.

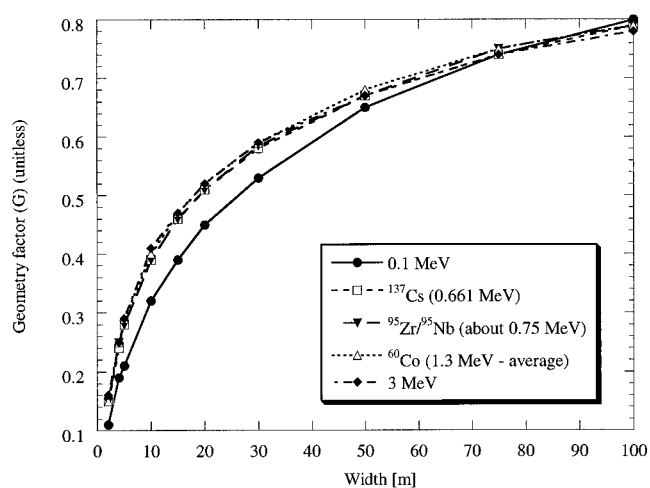
To obtain the geometry factor for shore widths other than the ones reported in Table 2, curve fitting or interpolation techniques can be used. Cubic spline interpolation gives a very precise reproduction of the curves in Fig. 2. Linear interpolation in the logarithmic space

^{||} Mathematica, Wolfram Research, Inc., 100 Trade Center Dr., Champaign, IL 61820-7237, USA.

Table 2. Geometry factors to be applied to the dose-rate factor for infinite extent as a function of shoreline width, for different gamma energies.

Width (m)	Radionuclide and representative energy (MeV)					
	0.1 MeV	¹⁴⁴ Ce/ ¹⁴⁴ Pr — ^a	¹³⁷ Cs, ⁹⁰ Sr, ¹⁰⁶ Ru/ ¹⁰⁶ Rh 0.661 MeV	⁹⁵ Zr/ ⁹⁵ Nb 0.75 MeV	⁶⁰ Co 1.252 MeV	3.0 MeV
2	0.11	0.14	0.15	0.15	0.15	0.16
4	0.19	0.22	0.24	0.25	0.25	0.25
5	0.21	0.26	0.28	0.28	0.29	0.29
10	0.32	0.37	0.39	0.39	0.40	0.41
15	0.39	0.44	0.46	0.46	0.47	0.47
20	0.45	0.49	0.51	0.51	0.52	0.52
30	0.53	0.56	0.58	0.58	0.59	0.59
50	0.65	0.66	0.67	0.67	0.68	0.67
75	0.74	0.74	0.74	0.75	0.75	0.74
100	0.80	0.79	0.79	0.79	0.79	0.78
150	0.88	0.87	0.86	0.85	0.85	0.83
200	0.93	0.91	0.90	0.90	0.89	0.87
300	0.97	0.95	0.94	0.94	0.93	0.91
400	0.99	0.97	0.97	0.97	0.96	0.94
500	0.995	0.98	0.98	0.98	0.97	0.96
600	0.998	0.99	0.99	0.99	0.98	0.97

^a 0.1 MeV (for ¹⁴⁴Ce) and 0.661 MeV (for ¹⁴⁴Pr); see text for details.

**Fig. 2.** Geometry correction factor as a function of width of the shoreline for selected gamma energies.

between the data points in Table 2 provides sufficient precision for practical applications, and is also easy to implement. For convenience, a curve fit of the data in

Table 2 is presented below. The data were fitted in the logarithmic space by a third order polynomial (eqn 8):

$$G = \exp\{m_0 + m_1[\ln(x)] + m_2[\ln(x)]^2 + m_3[\ln(x)]^3\}. \quad (8)$$

The coefficients of the fitted equation are presented in Table 3. Using eqn (8), the geometry factors presented in Table 2 can be reproduced to within 3%. However, a precision of more than 2 digits for the estimated geometry factors is not warranted for a number of reasons. First, the method used to derive them (eqn 3) is an approximation, being based on a point-kernel estimation of flux at 1 m height in air. Secondly, there are other much larger sources of uncertainty that affect the dose from external exposure such as the shape and dimensions of the contaminated area, the concentration of the radionuclide in the soil, the depth distribution of radionuclide in the soil, or the movement of the individual across the contaminated area. Thirdly, most radionuclides emit a combination of gamma rays of various energies. Thus, the geometry factor for such a radionuclide is actually a combination of the geometry factors for each gamma ray. Given the mild variation of the geometry factor with the energy of the gamma radiation, one can use a representative geometry factor (e.g., the geometry factor for an

Table 3. Coefficients^a of the fit equation (eqn 8) on the geometry factors presented in Table 2.

	0.1 MeV	0.661 MeV	0.75 MeV	1.252 MeV	3.0 MeV
m_0	-2.737	-2.4311	-2.4239	-2.4606	-2.3828
m_1	0.83273	0.85867	0.86463	0.92658	0.87616
m_2	-0.060051	-0.10347	-0.10663	-0.12581	-0.1164
m_3	-0.00049551	0.0044753	0.0047988	0.0064037	0.0057982
R	0.99983	0.9998	0.99971	0.99973	0.99988

^a The coefficients are given with all the precision obtained from the fit procedure. However, the geometry factor estimated using eqn (8) should be rounded to two significant digits.

energy of 1.25 MeV for ^{60}Co , which emits two gamma rays of 1.173 MeV and 1.332 MeV).

The geometry factors for estimation of external doses for an individual standing in the center of a circular or of a rectangular contaminated area are provided in Tables 4 and 5.

APPLICATION OF THE METHOD TO ESTIMATE EFFECTS OF EXPOSURE TO CLINCH RIVER SHORELINES

The method presented above was applied to assess the effects of external exposure to the shorelines of the lower Clinch River, which has been contaminated with radioactive materials released from the Oak Ridge Reservation during 1944–1991. The Grassy Creek area (CRM 17 to CRM 14; Fig. 1) is used to illustrate the application of the geometry factors in estimating the doses and risks. However, the discussion applies (with small differences) to the other segments of the river.

Of the radionuclides released to the Clinch River from ORNL between 1944 and 1991, ^{137}Cs , ^{60}Co , ^{106}Ru , ^{95}Zr , ^{95}Nb , ^{144}Ce , and ^{90}Sr were identified as important for the external exposure pathway during an initial screening phase (Thiessen et al. 1998). For these radionuclides, the amounts released (95% confidence intervals) were estimated at 44–74 TBq of ^{137}Cs ($T_{1/2} = 30$ y), 15–21 TBq of ^{60}Co ($T_{1/2} = 5.3$ y), 260–300 TBq of ^{106}Ru ($T_{1/2} = 1.0$ y), 31–52 TBq of ^{95}Zr ($T_{1/2} = 64.0$ d), 22–74 TBq of ^{95}Nb ($T_{1/2} = 35.0$ d), 17–27 TBq of ^{144}Ce ($T_{1/2} = 284.6$ d), and 63–81 TBq of ^{90}Sr ($T_{1/2} = 29.1$ y). Most of the releases occurred in the first two decades of the 1944–1991 release period (Thiessen et al. 1998). These radionuclides emit both beta and gamma radiation. For ^{137}Cs , ^{60}Co , ^{95}Zr , and ^{95}Nb , the energy from photon emissions

dominates the total emitted energy. For ^{106}Ru and ^{144}Ce , beta radiation makes a sizeable contribution, while ^{90}Sr is a pure beta emitter.

The excess lifetime risk of cancer from exposure to a radionuclide of concern was estimated for 11 types of cancer. The excess lifetime risk (ELR_i) of cancer is defined as the probability that an individual exposed to a radiation dose D_i in a given year (i) will acquire a radiation-induced cancer over the remaining lifetime of the individual. If the individual was exposed for more than 1 y, the excess lifetime risk of cancer is summed over the number of exposure years (N) to obtain a total excess lifetime risk ($TEL R$). Eqn (9) applies to each individual type of cancer:

$$TEL R = \sum_{i=1}^N ELR_i = \sum_{i=1}^N RF_i \times D_i, \quad (9)$$

where

RF_i = organ-specific risk factor, or the excess lifetime risk of cancer for an organ per unit dose from exposure during year “ i ” (Sv^{-1}); and

D_i = dose delivered to the organ from an exposure in year “ i ” (Sv).

The risk factors (RF_i) are calculated as the product of the age-adjusted excess relative risk per Sv ($ERR_{1\text{ Sv}}$) for cancer incidence obtained from the Japanese A-bomb survivors studies [Thompson et al. (1994) for all solid tumors and UNSCEAR (1994) for leukemia] and the age-adjusted annual background incidence rate of cancer in Tennessee[†] for a given cancer type, assuming an average 70-y lifetime. The $ERR_{1\text{ Sv}}$ values were adjusted for biases due to random and systematic errors in the doses estimated for the Japanese A-bomb survivors, and for biases in the extrapolation of the $ERR_{1\text{ Sv}}$ values from the Japanese population to the U.S. population (NCRP 1997; Thiessen et al. 1998).

The risks estimated using eqn (9) rely on the use of a linear non-threshold dose-response relationship. This type of relationship is currently accepted by the most prominent radiation protection organizations and consensus committees (i.e., National Council on Radiation Protection and Measurements, International Commission on Radiological Protection, United Nations Scientific Committee on the Effects of Radiation, National Radiological Protection Board). As part of the application of the linear model to exposures to radionuclides accumulated in the sediments of Clinch River, the risks were reduced by a dose and dose-rate reduction factor (DDREF) ranging from 2 to 10 for lung, 1 to 7 for leukemia, and 1 to 5 for all other cancers.

Table 4. Geometry factors for estimation of external doses to an individual standing in the center of a circular contaminated area. The values are obtained for a photon energy of 0.661 MeV (^{137}Cs).

For a given radius			
Radius (m)	G (unitless)	Radius (m)	G (unitless)
2	0.13	75	0.77
4	0.24	100	0.82
5	0.27	150	0.89
10	0.39	200	0.93
15	0.46	300	0.97
20	0.51	400	0.99
30	0.59	500	0.99
50	0.69	600	1.00
For a given area			G (unitless)
Area (m ²)	Radius of the circular area (m)		
10	1.8		0.12
100	5.6		0.29
1,000	17.8		0.49
10,000	56.4		0.71
100,000	178.4		0.92
1,000,000	564.2		1.00

[†] Tennessee Cancer Reporting System. Cancer incidence data for the State of Tennessee provided to the authors by the Tennessee Department of Health, Nashville, TN; 1996.

Table 5. Geometry factors for estimation of external doses to an individual standing in the center of a rectangular contaminated area. The values are obtained for a photon energy of 0.661 MeV (^{137}Cs).

Side of the rectangle (m)	2	4	6	8	10	30	50	70	100	200	400	600
2	0.07	0.10	0.11	0.12	0.13	0.14	0.15	0.15	0.15	0.15	0.15	0.15
4		0.15	0.18	0.19	0.20	0.23	0.24	0.24	0.24	0.24	0.24	0.24
6			0.21	0.23	0.24	0.29	0.29	0.30	0.30	0.31	0.31	0.31
8				0.26	0.27	0.33	0.34	0.34	0.35	0.35	0.35	0.36
10					0.29	0.36	0.37	0.38	0.38	0.39	0.39	0.39
30						0.48	0.52	0.54	0.56	0.57	0.58	0.58
50							0.58	0.61	0.63	0.66	0.67	0.67
70								0.64	0.67	0.71	0.73	0.73
100									0.71	0.76	0.79	0.79
200										0.84	0.88	0.89
400											0.94	0.96
600												0.98

The doses for individuals exposed to contaminated Clinch River shoreline sediments were estimated as

$$D_i = \sum_k C_{S,i,k} \times DRF_{ext,k} \times EF \times \Delta t, \quad (10)$$

where

- $C_{S,i,k}$ = concentration of radionuclide k in the shoreline sediment (Bq kg^{-1}) in year “ i ”;
- $DRF_{ext,k}$ = external dose-rate factor (Sv y^{-1} per Bq kg^{-1}), defined as the dose received during the period of exposure by an individual standing on a shore with a unit sediment contamination;
- EF = exposure frequency (unitless), obtained from the number of trips to the river per year and the number of hours per trip (Table 6); and
- Δt = exposure duration (1 y).

Furthermore,

$$DRF_{ext,k} = DRF_{ext,k}^{inf}(z) \times G(w) \times H, \quad (11)$$

where

- $DRF_{ext,k}^{inf}$ = dose-rate factor for an infinite surface contaminated with radionuclide k , as a function of z ;
- z = the thickness of the contaminated sediment slab (m) (Table 6);
- G = the geometry factor (unitless) that depends on the width of the shore (w); and
- H = an uncertain correction factor (unitless) that accounts for the nonhomogeneity of the contamination across the shore and for the movement of the individual on the contaminated surface (Table 6).

The doses and risks for each of the 11 cancer types were calculated for a combined exposure to all radionuclides (eqn 10), and for each radionuclide separately (eqn 10 without the summation). The total risk for all types of cancers combined was also estimated, using a method described in Thiessen et al. (1998).

The contaminated shore was idealized as a strip of land of finite width, but of infinite length (compared to the mean free path of the radiation) along the side of the river. It was shown that exposure to photons emitted from flowing surface water, deep sediments, or the opposite shores were small and can be neglected (Thiessen et al. 1998). For each reach of the river, the surface contamination of the shore was assumed to be uniform along the width and length of the shoreline.

The contamination of the sediment varies from one year to another, as predicted by the water transport model (HEC-6-R described by Thiessen et al. 1998). The estimated concentrations in the shoreline sediment of the most important radionuclides are presented in Fig. 3 for the Grassy Creek area (CRM 14.0). The uncertainty in the estimated concentrations in sediment (expressed as a geometric standard deviation, GSD) is about 1.7 for all radionuclides for years of release past 1955. The uncertainty in the concentration in sediment is larger in the early years. This is due to the lack of documentation and the lack of adequate measurements of the amounts released into the Clinch River in the early years. For all radionuclides, except ^{95}Nb , the GSD for the early years can be as large as 3.0; for ^{95}Nb the GSD reaches values of 5.8 and 6.0 in the early years.

The radionuclides released into the Clinch River are accumulated in the first few centimeters of sediment. Thus, the source of radiation is a volume source, rather than a surface source. However, it is always possible to perform a volume-to-surface source transformation (i.e., to estimate the flow of radiation from the surface). The gamma radiation coming out from the surface will have an energy spectrum different from the energy of the gamma radiation emitted by the radionuclide. However, given the mild variation with energy (Fig. 2, Table 2), the geometry factors derived for the surface source (eqn 4) can be used for a volume source. The dose-rate factors for various contamination depths ($DRF_{ext,k}^{inf}$) published by Eckerman and Leggett (1996) were used.

For these radionuclides, the geometry factors were selected as follows: For $^{137}\text{Cs}/^{137\text{m}}\text{Ba}$, the single gamma emission (0.661 MeV) was used for estimating the

Table 6. Parameters used for the analysis of doses and risks from external exposure to radionuclides accumulated in the Grassy Creek area (CRM 17–CRM 14) of the Clinch River shoreline sediments.

Parameter/values				Explanation	
Exposure frequency for the Grassy Creek area					
Low water season		High water season		Units	In the Grassy Creek area, the north side of the river was not accessible to members of the public because the land was part of the Oak Ridge Reservation. However, exposure to shoreline sediments was possible on the south side of the Clinch River for tenants fishing and children playing near the water's edge. Activities on the bank of the river depended on the season, and on the level of the water (Thiessen et al. 1998.)
Winter	Spring	Summer	Fall		
1–2	1–2	4–12	4–12	trips mo ⁻¹	
0.5–5	0.5–8	0.5–8	0.5–5	h per trip	
	80–450			h y ⁻¹	

The thickness of the contaminated sediment layer (z)Triangular PDF^a.

min = 2 cm, mode = 7 cm, max = 15 cm

Spatial and temporal nonhomogeneity of contamination (H)Uniform PDF^a

min = 0.95, max = 1.05

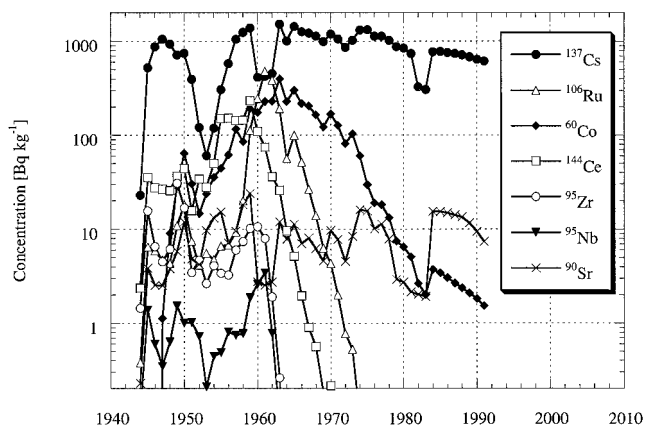
The average thickness of the contaminated sediment layer is 7 cm. At least 2 cm of sediment is present at any time. In some cases, additional sediment from the hill-slopes of the shores is deposited on the shoreline by run-off during rain, and it mixes with the contaminated sediment deposited by the water, increasing the amount of sediment deposited. However, it is unlikely that the thickness of the contaminated slab would exceed 15 cm. On the other hand, during scouring events such as floods, the sediment is removed faster than it is deposited. Because direct measurements of the thickness of the contaminated shoreline sediment at each location and in each exposure year were not available, the thickness of the sediment slab was treated as an uncertain variable (Thiessen et al. 1998.)

The doses and risks are calculated based on the assumption that the ground surface is uniformly contaminated. However, the concentration in the shoreline sediment could change from the water line to the upper edge of the shore. Also, annual average concentrations in sediment are used and there is uncertainty about the exact value of the average shoreline concentration. The daily variation of shoreline contaminant concentrations would largely be the result of variations in deposition and scouring patterns on a daily scale. These sources of uncertainty were appreciated to be less than 5% (Thiessen et al. 1998.)

^a PDF = probability distribution function (used to describe the uncertainty in the parameter values).

geometry factor. For ⁶⁰Co, the average energy (1.252 MeV) of the two photons (1.173 MeV and 1.332 MeV) was chosen as the representative energy for ⁶⁰Co. Three gamma rays of practically the same energies (0.72, 0.76, 0.77 MeV) are emitted by ⁹⁵Zr and its decay product ⁹⁵Nb. A geometry factor was estimated for these radionuclides using a representative gamma energy of 0.75 MeV.

¹⁰⁶Ru and its decay product ¹⁰⁶Rh emit a large number of photons, which have energies ranging from 0.428 MeV to 2.406 MeV. The lowest energy gamma ray with a large nuclear yield has an energy of 0.512 MeV. For this range of energies, the geometry factors remain approximately the same for a given shoreline width (Fig. 2). Therefore, the correction factor for any of the intermediate energies can be assumed to be a good representation of the real correction factor for ¹⁰⁶Ru/¹⁰⁶Rh. For the Clinch River assessment, the geometry factors obtained for ¹³⁷Cs/¹³⁷Ba were also applied to ¹⁰⁶Ru/¹⁰⁶Rh.

**Fig. 3.** Concentration of the radionuclides of interest in the shoreline sediments of the Clinch River for Grassy Creek area (CRM 17 to CRM 14).

A geometry factor for $^{144}\text{Ce}/^{144}\text{Pr}$ is more difficult to assess due to the large spectrum of energies emitted by these isotopes. ^{144}Ce emits photons in a range from 0.033 MeV to 0.133 MeV (ICRP 1983). The most probable photon emission (10.8%) is the most energetic one, 0.133 MeV. ^{144}Pr , the decay product of ^{144}Ce , emits a photon of 0.697 MeV energy in 1.5% of its decays. The other listed photon emissions (ICRP 1983) have energies up to 2.186 MeV, but very low probabilities of occurrence. For most organs, the contribution to the external dose from ^{144}Pr is about twice as high as the contribution from ^{144}Ce . A representative reduction factor for ^{144}Ce (which emits low energy photons) can be obtained by using the energy of 0.1 MeV (Table 2). On the other hand, for ^{144}Pr the reduction factor obtained for 0.661 MeV is appropriate. Therefore, a reduction factor for $^{144}\text{Ce}/^{144}\text{Pr}$ (Table 2) was determined as a weighted combination of the reduction factor for ^{144}Ce (one-third) and the reduction factor for ^{144}Pr (two-thirds).

The internal organ dose rates for $^{90}\text{Sr}/^{90}\text{Y}$ (which are beta emitters only) are due to photons arising as the emitted beta particles lose their kinetic energy within the ground and air (bremsstrahlung radiation). Only a small fraction of the kinetic energy of the beta radiation is converted to photons; as a result the numerical value of the dose-rate factor for these radionuclides is one to two orders of magnitude lower than for other radionuclides. However, in the screening phase of this project, $^{90}\text{Sr}/^{90}\text{Y}$ came very close (a lifetime risk of 9×10^{-6}) to the decision limit (a lifetime risk of 10^{-5}) used to select radionuclides important for the external irradiation pathway. ^{90}Sr was kept in the final analysis, but no particular efforts were made to derive the external dose-rate factors for exposure to finite contaminated surfaces. For simplicity, the dose-rate factors for infinite surfaces were reduced using similar geometry factor as for the other radionuclides. The energy of the bremsstrahlung radiation produced by $^{90}\text{Sr}/^{90}\text{Y}$ ranges from zero to the maximum energy of the emitted beta radiation (about 2.3 MeV). The geometry factors for a 0.661 MeV energy were used for $^{90}\text{Sr}/^{90}\text{Y}$. This choice is conservative because, for the size of Clinch River shoreline widths, the geometry factors are lower for lower energies where there is more attenuation.

The above approach and discussion apply to all organs other than skin. For external irradiation from contaminated area sources, the dose is given by the contribution of both electrons and gamma rays emitted by each radionuclide. For all internal organs, the dose is given by photons because electrons are stopped shortly after penetrating the skin. For skin, however, electrons can dominate the dose from radionuclides such as $^{90}\text{Sr}/^{90}\text{Y}$, $^{106}\text{Ru}/^{106}\text{Rh}$, and $^{144}\text{Ce}/^{144}\text{Pr}$ (Kocher and Eckerman 1981). Transport of electrons through air from the contaminated ground to the body surface is different from the transport of gamma radiation. Practically, a small contaminated area can produce the same electron dose to the skin as a very large (infinite) contaminated area. Thus, for $^{90}\text{Sr}/^{90}\text{Y}$, $^{106}\text{Ru}/^{106}\text{Rh}$, and $^{144}\text{Ce}/^{144}\text{Pr}$ and for

skin tissue only, no adjustment for the geometry of the shoreline is necessary. That is, for these radionuclides, the doses to skin can be calculated based solely on the dose-rate factors for an infinite area. For ^{137}Cs , ^{60}Co , and $^{95}\text{Zr}/^{95}\text{Nb}$ (mainly gamma emitters), the dose-rate factors for skin are adjusted using the calculated geometry factor as for all other organs.

The uncertainties in the radiation doses and the risks were estimated by propagating the uncertainties through eqns (9) and (10). Each parameter of the equation was described by a probability distribution function (PDF), as described below. The Monte-Carlo method using the Latin Hypercube Sampling technique (Iman and Shortencarier 1984; Morgan and Henrion 1990) was employed to generate 400 samples for each parameter. A probability distribution function for each dose and risk estimate was obtained using eqns (9) and (10) starting from the set of samples for each parameter. The 95% confidence interval of the resulting PDF was used to describe the uncertainty in the estimated doses and risks. The median of the resulting PDF was reported as the best estimate of the quantity of interest. Four hundred Latin Hypercube samples are enough to insure stability of the limits of the 95% confidence interval.

Thickness of the contaminated sediment layer

Radionuclides accumulated by sedimentation along the shorelines of the Clinch River are found in the top few centimeters of the sediment bed. Dose-rate factors are available (Eckerman and Leggett 1996) as a function of the thickness (z) of a contaminated slab, for $z = 1$ cm, 5 cm, 15 cm, and for infinite depth (defined as four times the mean free path of emitted photons.)

To calculate the dose-rate factor for any given thickness (z), an empirical observation was used. It can be shown that the dose-rate factors vary with the thickness (z) of the contaminated slab as $[1 - \beta^* \exp(-z/mfp_{soil})]$, where β is a constant, and mfp_{soil} is the mean free path of the radiation in soil. This property can be illustrated by plotting the dose-rate factors as a function of $\exp(-z/mfp_{soil})$ (Fig. 4).

Thus, to obtain the dose-rate factors for any given thickness, a linear fit to the curves shown in Fig. 4 was used. This approximation is good for all internal organs because the dose is dominated by gamma radiation emitted by the radionuclides (Fig. 4a). Also, it holds for skin tissue (Fig. 4b), for which the dose can be dominated by either gamma radiation (^{137}Cs , ^{60}Co , ^{95}Zr , ^{95}Nb) or by emitted electrons (^{90}Sr , ^{106}Ru , ^{144}Ce). A similar method for dealing with the thickness of the contaminated soil is described by Chen (1991).

The thickness of the contaminated sediment layer (Table 6) was defined as a probability distribution function (PDF). For each value of the thickness sampled from the PDF, a dose-rate factor was estimated using relationships similar to those in Fig. 4 derived for all organs and each radionuclide. This dose-rate factor is representative for an infinite soil slab of the sampled thickness. Then, this dose-rate factor was multiplied by the geometry factor to account for the finite shoreline

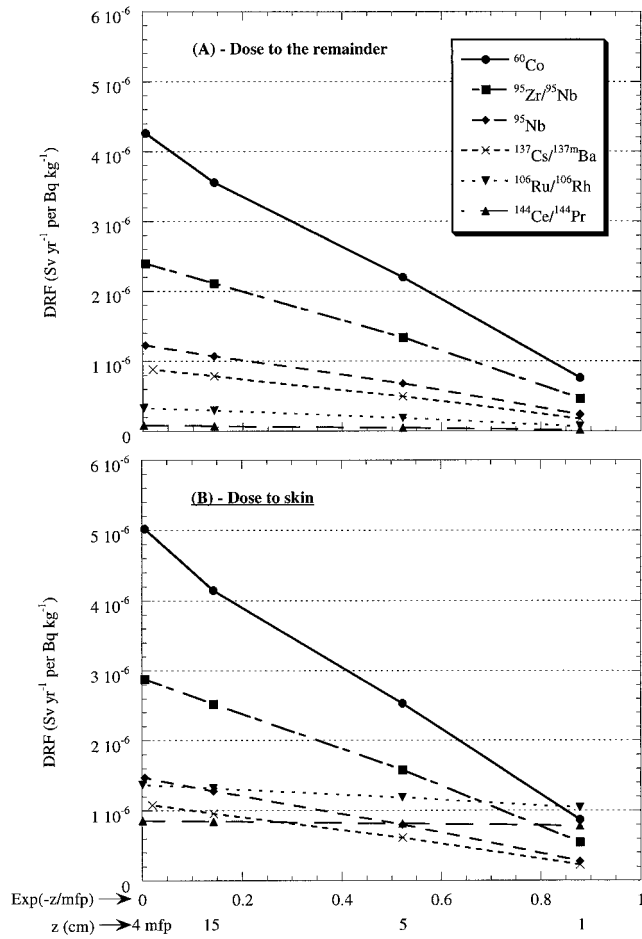


Fig. 4. Dependence of the external dose-rate factor (Eckerman and Leggett 1996) on the depth (z) of an infinite uniform contaminated slab of soil expressed in terms of the mean free path (mfp) of ^{137}Cs photons in soil.

width (eqn 11). The same thickness of the contaminated sediment was applied for all locations and all years. In contrast, the width of the shoreline was derived for each location of interest and for each year.

Determination of the shoreline width

The width of the riverine shores is subject to seasonal and annual variations. The width of the Clinch River shores was obtained using the daily river width estimated by the HEC-6R model and the slope of the shores at the location of interest available from the measurements of the river profile (Thiessen et al. 1998). The measurements indicated that one shore on the river is very steep, while the opposite one is rather flat and accessible to people. To express the uncertainty introduced by the daily variation in the shoreline width the following procedure was used:

1. The maximum and the average shoreline width in a given year were estimated (Fig. 5);
2. A minimum shoreline width of 2 m was assumed for all years;

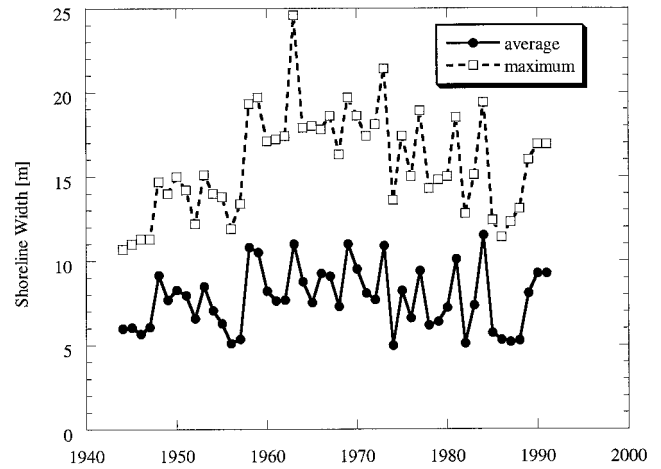


Fig. 5. Estimated shoreline width for the Grassy Creek area of the Clinch River.

3. The uncertainty in the width of the shoreline during a year was expressed as a triangular probability distribution function with a minimum of 2 m, a mode equal to the estimated average shoreline width in each year and a maximum given by the estimated maximum shoreline width in the same year; and
4. The probability distributions for the shoreline width were sampled independently for each year and each location.

Based on the shore width, a geometry adjustment factor was obtained using the relationship presented in Fig. 2. The result of this procedure is a set of probability distributions that describes the uncertainty introduced by the geometry of the Clinch River shoreline (Fig. 6). There is one distribution for each exposure year (48 y), for each radionuclide of interest (7 radionuclides), and

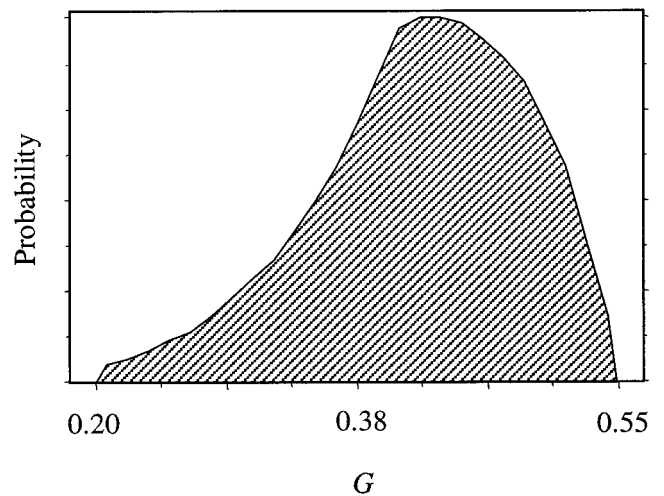


Fig. 6. Example of the estimated geometry adjustment factor (G) for ^{137}Cs at for the Clinch River shore in the Grassy Creek area in 1963.

for each location of interest (4 locations). These distributions are applied to the dose-rate factors for each specific organ. The dose-rate factors available for areas contaminated to an infinite extent were then modified using the geometry factor (eqn 11).

Organ masses

The published dose-rate factors (Eckerman and Leggett 1996) are calculated for the ICRP reference individual. Differences between the prescribed values of organ masses for the reference individual and those for real individuals exposed along the Clinch River can arise from natural variability and from sex-specific factors. The uncertainties in the specifications of the target organ mass were estimated (Thiessen et al. 1998) and then used to obtain the total dose to each organ. To account for the uncertainty in the organ masses, the dose-rate factors were multiplied by the deterministic values of the organ masses for the reference individual (ICRP 1992) and then divided by the probability distribution functions representing the uncertainty in the estimated organ masses.

Dose and risk estimates from external exposure to radionuclides accumulated in the Clinch River shoreline sediments

Radiation doses and risks of radiogenic cancer from external exposure to Clinch River shorelines were estimated for reference adult individuals. Doses were estimated for 21 organs, for each radionuclide, for each segment of the river and for all years of exposure (1944–1991). The excess lifetime risk of cancer was estimated for 11 cancer types (breast, leukemia, lung, colon, bladder, ovaries, thyroid, kidneys, stomach, liver, and pancreas) for which both dose-response relationships and background cancer incidence rates were available. The dose and risk estimates varied little from one location to the next. The estimates obtained for the Grassy Creek area will be discussed here.

The highest estimated doses from external exposure to radionuclides released to Clinch River were received by (in order) bone surface (0.047 cSv; 95% C.I. 0.0082–0.22 cSv), skin (0.040 cSv; 95% C.I. 0.0088–0.16 cSv), breast (0.034 cSv; 95% C.I. 0.0070–0.15 cSv), bone marrow (0.029 cSv; 95% C.I. 0.0052–0.13 cSv), and colon (0.027 cSv; 95% C.I. 0.0056–0.14 cSv). The estimated doses vary among all organs by less than a factor of 3. The relative low variation among organs is characteristic to external exposure pathway for which the irradiation of the whole body is quite uniform. Exposure to ^{60}Co and ^{137}Cs dominated the doses for all organs (including skin).

The estimated excess lifetime risk varied among cancer types by about two orders of magnitude. The highest risks were estimated for breast cancer (95% C.I. of 1.1×10^{-6} to 5.9×10^{-5}), leukemia (95% C.I. of 2.4×10^{-7} to 1.3×10^{-5}), lung cancer (95% C.I. of 2.1×10^{-7} to 8.9×10^{-6}) and colon cancer (95% C.I. of 1.9×10^{-7} to 1.0×10^{-5}).

The uncertainty in the dose estimates was practically uniform among organs, with a geometric standard deviation (GSD) of about 2.2. The uncertainty in the risk estimates varied from a GSD of 2.6 for lung cancer to a GSD of 4.9 for stomach or liver cancer. The additional uncertainty is introduced solely by the uncertainty in the dose-response relationship.

By using no geometry correction, the dose estimates would have been overestimated by a factor of about 2.8 (Fig. 7). On the other hand, by using the reduction factor (equal to 0.2) listed by Eckerman and Ryman (1993) the dose estimates would have been underestimated by a factor of 1.8. For a given year or a given radionuclide the uncertainty in the annual dose estimate is dominated by the uncertainty in the concentration in sediment, and by the uncertainty in the exposure frequency. The uncertainty in the geometry factor has a contribution to the uncertainty in the annual dose that depends on the variation in the shoreline width in a given year, at the location of interest. Typical values for this contribution are about 10%. The contribution of the uncertainty in the geometry factor to the total dose (summed over all years of exposure and over all radionuclides) is reduced because of the summation to values lower than 10%.

CONCLUSION

A simple analytical method has been developed to estimate external doses from exposure to contaminated riverine shorelines. The method consists of deriving an adjustment factor that accounts for the geometry of the riverine shoreline; this adjustment factor is applied to the dose-rate coefficients already available for infinite contaminated surfaces. Such a geometry factor circumvents very complex radiation transport calculations which

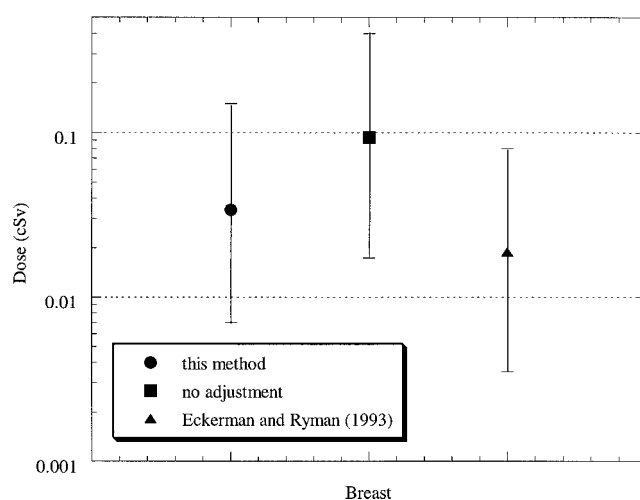


Fig. 7. Comparison of the estimates of total dose to the breast tissue obtained using the geometry factors derived here, using no adjustments for the finite size of the shorelines, and using the reduction factor (equal to 0.2) recommended by Eckerman and Ryman (1993). Similar differences between dose estimates are observed for other organs.

would otherwise be necessary to model exposures to a finite contaminated surface. The method was applied to derive similar adjustment factors for contaminated surfaces of other simple geometries (e.g., circular surfaces).

The geometry factor changes mildly with the energy of the gamma radiation. This property is extremely powerful because it allows for the geometry factor to be used for radionuclides emitting multiple gamma rays of various energies. If a quick analysis is desired, one should use the geometry factors derived for ^{137}Cs (Table 2) for all radionuclides. More refined analysis can be performed by deriving geometry factors for each radionuclide according to its gamma spectrum. Also, the mild variation with energy allows the geometry factors to be applied to the case when radionuclides are accumulated in layers under the soil surface, and not only to the case when radionuclides are deposited onto the soil surface.

Empirical relationships between the geometry factor and the dimension of the shoreline were provided, so that one can obtain values of the geometry factor for any shoreline width. These relationships can be easily used to account for the uncertainty in the dimension of the shoreline.

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