A Technical Basis for the Selection of Area vs. Volume Criteria for Contaminated Structures – 11003

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ABSTRACT

The selection of Derived Concentration Guideline Levels (DCGLs) is a necessary element in the planning and implementation of the decommissioning of nuclear facilities. For interior building surfaces, the RESRAD-Build code is often used to calculate the DCGLs based on the site-specific information for the facility. This code discusses the two prevalent options used when modeling the surfaces of the contaminated building structures. One option is the Area Source approach for which the DCGL is stated in units of radioactivity per unit area (i.e., pCi/m^2 or dpm/100 cm^2). The second option is the Volume Source approach for which the DCGL is stated in units of radioactivity per unit mass (i.e., pCi/g). In some cases, it is obvious which option should be used to fit the specific facility being decommissioned. However in many cases, it is not clear which option is appropriate. This presentation addresses that question: How far can the contamination penetrate into a surface before it becomes a Volume Source rather than an Area Source?

This paper presents a basic technical approach to establishing the basis for deciding which source option should be selected. Examples applying this method are provided for five radionuclides and one mixture to show cases that cover a range of different radiological emissions and to illustrate the conclusions that can be drawn from this technical approach. Based on these examples, a table is provided for the thicknesses of contamination that recommends a transition point between selecting the Area Source or Volume Source to calculate site-specific DCGLs. Conclusions are given that would help guide the reader in conducting their own development of a site-specific technical basis document to address this point.

INTRODUCTION

The RESRAD-Build computer code[1] is often used in the nuclear industry to develop site-specific DCGLs that are appropriate to be used as an element of a facility decommissioning process to demonstrate compliance with the dose criterion established by the regulatory agency. This computer code was developed under the joint sponsorship of the U.S. Department of Energy (USDOE) and the U.S. Nuclear Regulatory Commission (USNRC) for site-specific dose assessment of residual radioactivity on structural surfaces. An alternative conservative approach is to utilize the screening values provided in USNRC Decommissioning Guidance (NUREG-1757, Volume 2 Rev. 1, Appendix H, Table H.1).

If the decision is to use RESRAD-Build to develop site-specific DCGLs, the parameter selection choices for source geometry is whether the source is a “Volume”, “Area”, “Line” or “Point”. This paper is limited to a discussion of the “Volume” and “Area” options.

A Volume Source is entered by defining the geometry (area and thickness of the source) and the concentration (such as in pCi/g) of the radionuclides in the source. The code does allow different layers to be considered for the Source term. An Area Source is entered by defining the geometry (area of the source) and surficial concentration (such as in pCi/m^2) of the radionuclides in the source. No guidance is provided in the code regarding the decision on which selection is appropriate for the specific situation to be analyzed. In certain instances where the contaminated surface is porous, the contamination would be expected to have penetrated into the material. The RESRAD-Build user manual provides no specific guidance as to when to select the Volume or Area source term model over the other. The description of the two source configurations provided in the user manual is:

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[1] RESRAD-Build is a member of the RESRAD family of codes developed by the Environmental Science Division of Argonne National Laboratory. Information about and copies of the entire family of codes are available at http://web.ead.anl.gov/resrad/home2/index.cfm. (Most recently accessed on November 5, 2010)

- A contaminated area in the building should be considered a Volume Source if it can be clearly represented in a three-dimensional configuration. A segment of a wall in the building, contaminated with radioactive materials, is an example of a possible Volume Source.
- Definition of a Surface Source is considered in those cases of surface contamination in which the thickness of the contaminated layer is considerably smaller than the affected area exposed to open air.

USNRC Decommissioning Guidance (NUREG-1757, Volume 2 Rev. 1, Appendix H, Section H.2.2) states that for use of the area screening values provided in the document: "The residual radioactivity on building surfaces (e.g., walls, floors, ceilings) should be surficial and non-volumetric [e.g., ≤10 mm (0.39 in) of penetration]". No technical basis or further explanation is provided to justify this statement.

The standard ANSI/HPS N13.12-1999 (Reaffirmed 2010), "Surface and Volume Radioactivity Standards for Clearance" also provides some guidance. Under definitions (Section 2), the standard states:

- **Surface Contamination**: Radioactive contamination residing on or near the surface of an item. This contamination can be adequately quantified in units of activity per unit area. When an item has been exposed to neutrons (including structural components and shielding at nuclear reactors), or when an item could have cracks or interior surfaces allowing the distribution of radioactive contamination within the interior matrix, it is considered to be a volume contamination source.
- **Volume Contamination**: Radioactive contamination residing in or throughout the volume of an item. Volume contamination can result from neutron activation or from the penetration of radioactive contamination into cracks or interior matrix of an item.

Further, in Section 4.3, "Surface and volumetric measurements", of the standard it states:

- Volumetric measurements for clearance ***shall*** be used when volumetric radioactive materials are known or potentially present.
- Surface screening levels ***shall*** be used when an item's size or shape reasonably allows direct radiological surveys for surface radioactive contamination.

When a concrete surface has been wetted for an extended period of time, it is expected that the surface contamination will penetrate into the structure and experience has shown that the penetration into the concrete may be substantial. Shallow surface removal techniques have been found insufficient to remove all of the contamination from the surface of the structure. The practical implication is whether a Final Status Survey should be completed using surface measurements with a portable survey instrument or whether volumetric measurements are more appropriate. Volumetric measurements can be made either in-situ or by laboratory analysis of removed material. Volumetric measurements are more time consuming and expensive compared to portable survey instrument measurements.

This paper presents a basic technical approach to establishing the basis for deciding which source option should be selected. Examples applying this method are provided for five radionuclides and one mixture to show cases that cover a range of different radiological emissions and to illustrate the conclusions that can be drawn from this technical approach. Based on these examples, a table is provided for the thicknesses of contamination that recommends a transition point between selecting the Area Source or Volume Source to calculate site-specific DCGLs. Conclusions are given that would help guide the reader in conducting their own development of a site-specific technical basis document to address this point.

PROCEDURE TO ESTABLISH EQUIVALENT BASIS FOR COMPARISON

It is not readily apparent when a Volume Source term in units of activity per unit mass (e.g. pCi/g) would be equivalent to an Area Source term in units of activity per unit area (e.g. pCi/m^2). For purposes of this paper, two sources would be the radiological equivalent if the two sources provide the same dose rate (e.g. mRem/yr) and the two sources have the same area and same total inventory of radioactive material. This point of equivalence for a Volume and Area source can be established by following these steps:

1. Select a specific room model with a source term of a specific area size.
2. Calculate the Surface DCGL (e.g. in units of pCi/m^2) using RESRAD-Build and calculate the total inventory of radioactivity associated with the source at the DCGL level.

3. Calculate the Volume DCGL (e.g. in units of pCi/g) using RESRAD-Build for a series of source thicknesses and calculate the total inventory of radioactivity associated with the source at the DCGL level for each source thickness.
4. Plot the results of Step 3 for the total inventory against the source thickness.
5. The "point of equivalency" is the point on the plot from Step 4 for the thickness of the Volume Source where the inventory on the curve equals the inventory calculated in Step 2 for the Area Source.

The "point of equivalency" is then the thickness of the Volume Source that results in the same dose as an area source of equal areal size. For smaller Volume Source thicknesses, the Area Source model results in a larger allowed inventory of radioactive material at the same dose. For larger volume source thicknesses, the Volume Source model results in a larger allowed inventory of radioactive material at the same dose. Knowledge of this thickness would thus provide a rational technical basis for when to utilize DCGLs calculated by the Volume Source model vs. those calculated by the Area Source model.

RESRAD-BUILD PARAMETERS SELECTED

The RESRAD-Build input parameters were taken as the standard default parameters for simplification of the calculations. No effort has been made to utilize the probabilistic capabilities that are incorporated into this code. Table I lists the primary input parameters for the calculations of the two models considered.

Table I. Primary input parameters for RESRAD-Build code calculations

Parameter	Volume Source Model	Area Source Model
Source Area	36 m^2 (Circular)	36 m^2 (Circular)
Source Thickness	Varied from $1X10^{-7}$ to 1,000 cm, one layer	Not Applicable
Radionuclide	Individual radionuclides from Table 1 and mixture from Table 2	Individual radionuclides from Table 1 and mixture from Table 2
Radionuclide concentration	1 pCi/g	1 pCi/m^2
Source density	2.4 g/cc	Not Applicable
Number of source layers	1	Not Applicable

INDIVIDUAL RADIONUCLIDES CONSIDERED – SET #1

For illustration purposes, the set of radionuclides given in Table II have been used as input to the RESRAD-Build code calculations.

Table II. List of radionuclides considered and their primary emissions for Set #1

Radionuclide	Emissions	Comment
Fe-55	Electron Capture, emission of low energy photons	Hard to detect radionuclide due to the low energy of the photons
Sr-90	2 betas, average energies 195.8 keV and 934.8 keV	Pure high energy Beta emitter, includes the Y-90 progeny
Cs-137	Gamma, 661.65 keV Beta, average energy 415.2 keV	Primarily a Gamma emitter with a beta emission, includes the Ba-137m progeny
Am-241	Alpha, 5.4857 MeV (85.2%)	Primarily an Alpha emitter with some low energy photon emissions

SUMMARY OF RESULTS FOR SET #1

Figures 1 through 4 show the plotted results for the calculations of the four individual radionuclides considered in Set #1.

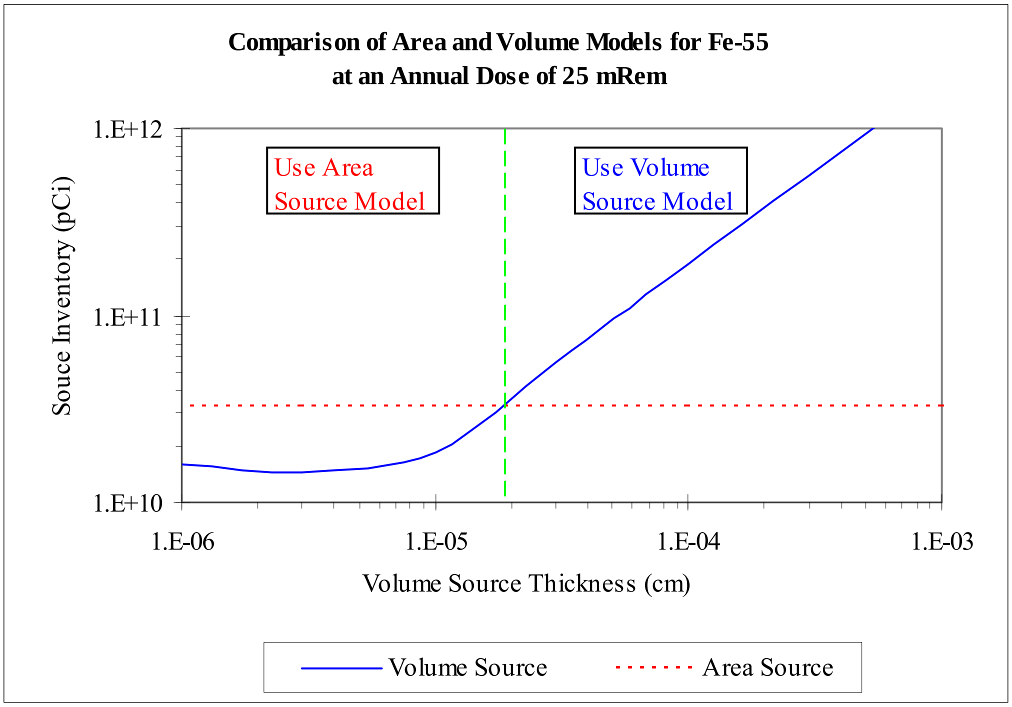

Figure 1. Comparison of Area and Volume Source Models for Fe-55

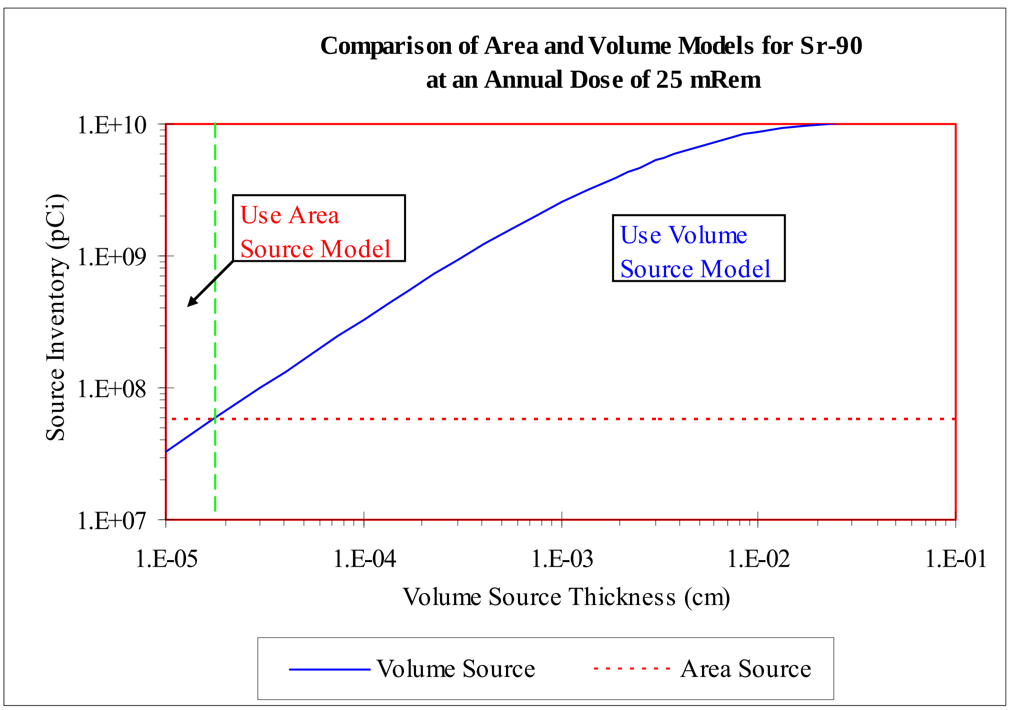

Figure 2. Comparison of Area and Volume Source Models for Sr-90

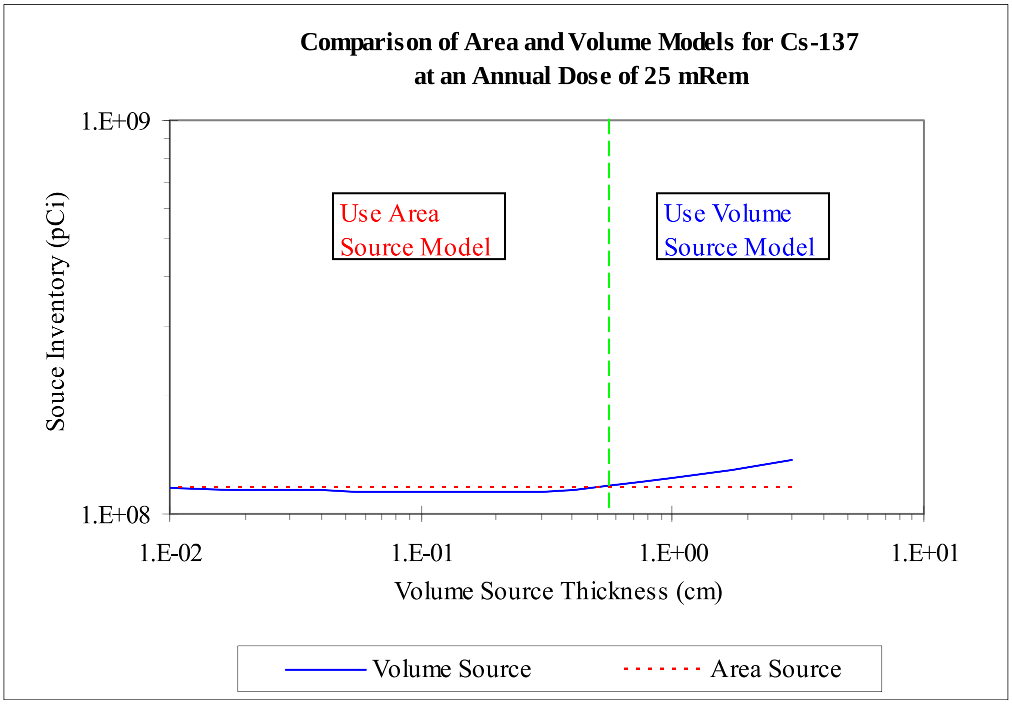

Figure 3. Comparison of Area and Volume Source Models for Cs-137

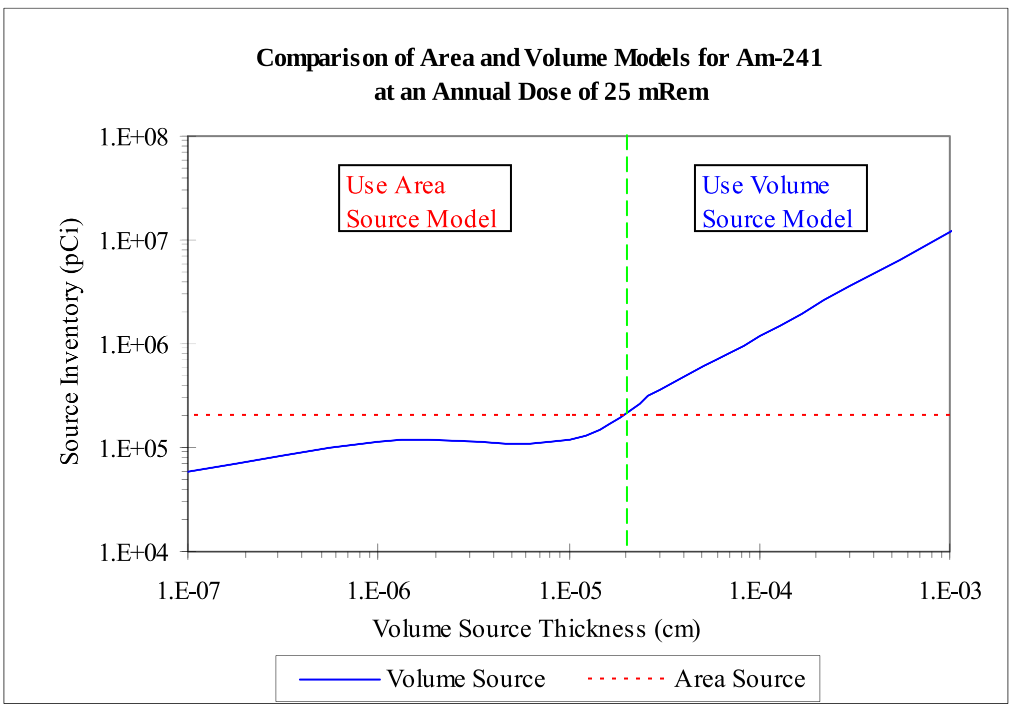

Figure 4. Comparison of Area and Volume Source Models for Am-241

EXAMPLE - DETAILS OF CALCULATIONS FOR Cs^{137}

Table III provides additional details of the results for Cs-137 as an example of the input and calculations performed. The RESRAD-Build code was used in a deterministic mode to calculate the first year dose (mRem/yr). This value is divided into 25 mRem/yr in order to calculate the DCGL appropriate for the model in units of either pCi/m^2 or pCi/g. The value of 25 mRem/yr is the USNRC dose criterion for unrestricted release at time of license termination. Table III gives the values obtained for the DCGLs. Using the calculated DCGL for each case the total inventory of the source is calculated based on the source geometry and density. The calculated values of source inventory were then plotted to give Figure 3.

Table III. Calculated Results for Cs-137

Model	Source Concentration	Source Thickness (cm)	Source Area (m^2)	Calculated DCGL	Calculated Source Inventory (pCi)
Area	1 pCi/m^2	Not Applicable	36	3,280,000 pCi/m^2	1.18E+08
Volume	1 pCi/g	3	36	53 pCi/g	1.36E+08
Volume	1 pCi/g	1	36	144 pCi/g	1.24E+08
Volume	1 pCi/g	0.3	36	441 pCi/g	1.14E+08
Volume	1 pCi/g	0.1	36	1,309 pCi/g	1.13E+08
Volume	1 pCi/g	0.03	36	4,448 pCi/g	1.15E+08
Volume	1 pCi/g	0.01	36	13,508 pCi/g	1.17E+08

DISCUSSION OF RESULTS FOR SET #1

The “Point of Equivalency” (where the two lines cross as shown in Figures 1 through 4) provides the decision point between the Volume Model and the Area Model in order to determine the appropriate DCGL in a specific case. These are example calculations and the use of site-specific parameters would be expected to change the figures. In each figure, when the contamination source thickness is less than the point where the lines cross it would be appropriate to use the Area source model to derive the DCGL. When the contamination source thickness is greater than the point where the lines cross it would be appropriate to use the Volume Source model to derive the DCGL. The design of the Final Status Survey would need to reflect an appropriate measurement method to demonstrate compliance with the DCGL selected as appropriate for the situation.

For high energy gamma emitters such as Cs-137, the cross over point is about 0.5 cm or 5 mm. This value is consistent with the USNRC guidance that for use of the area screening values provided in NUREG-1757, Volume 2, Appendix H: “The residual radioactivity on building surfaces (e.g., walls, floors, ceilings) should be surficial and non-volumetric [e.g., ≤10 mm (0.39 in) of penetration].” The curve in Figure 3 is relatively flat so the decision point where it would be appropriate to use the Volume Source Model would require that the contamination had penetrated deep into the surface. There does not seem to a strong preference of one model over the other in the case of hard gamma emitters.

For the other three radionuclides (Fe-55, Sr-90 and Am-241) the cross over point is about 0.00002 to 0.00003 cm. In practicality, this means that for surfaces other than a hard non-porous surface such as metal, it would be appropriate to use the Volume Source Model to calculate the appropriate DCGL.

INDIVIDUAL RADIONUCLIDES CONSIDERED – SET #2

For illustration purposes, the radionuclide given in Table IV has been used as input to the RESRAD-Build code calculations. Figure 5 shows the plotted results for the calculations for Tritium considered in Set #2.

Table IV. Radionuclide considered for Set #2

Radionuclide	Emissions	Comment
H-3 (Tritium)	Beta, average energy 5.7 keV	Pure low energy Beta emitter

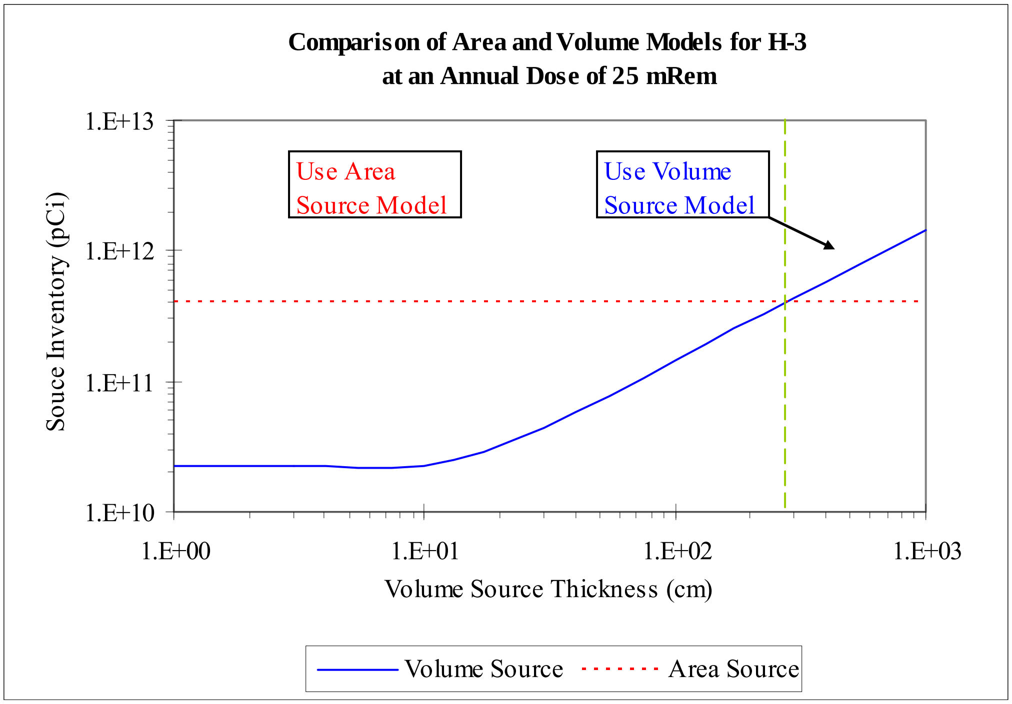

Figure 5. Comparison of Area and Volume Source Models for H-3

DISCUSSION OF RESULTS FOR SET #2

Tritium (H-3) is a case unto itself in that even for great depths of penetration, the Area Model is the limiting case and should be used to derive the site specific DCGL. The RESRAD-Build code treats H-3 differently than other radionuclides and uses a specific tritium-transport model for the Volume Source which incorporates a diffusion process. Tritium contamination requires special consideration, because in addition to erosion, tritium, which most often is in the chemical form of tritiated water (HTO), can vaporize and diffuse out of the building material and reach the indoor air. There are different parameters that need be selected when tritium is a contaminant. Since this paper is based primarily on using the default values given in the code, it is not known how representative those values are with respect to site-specific models.

MIXTURE OF RADIONUCLIDES CONSIDERED – SET #3

For illustration purposes, a mixture of a selection of the radionuclides given in Table V has been used as input to the RESRAD-Build code calculations. This mixture selection is based on experience as being reasonably representative of possible conditions.

Table V. Mixture of radionuclides considered

Radionuclide	Percent of total activity
Sr-90	37%
Cs-137	62%
Am-241	1%

Figure 6 shows the plotted results for the calculations of the mixture of radionuclides considered as Set #3.

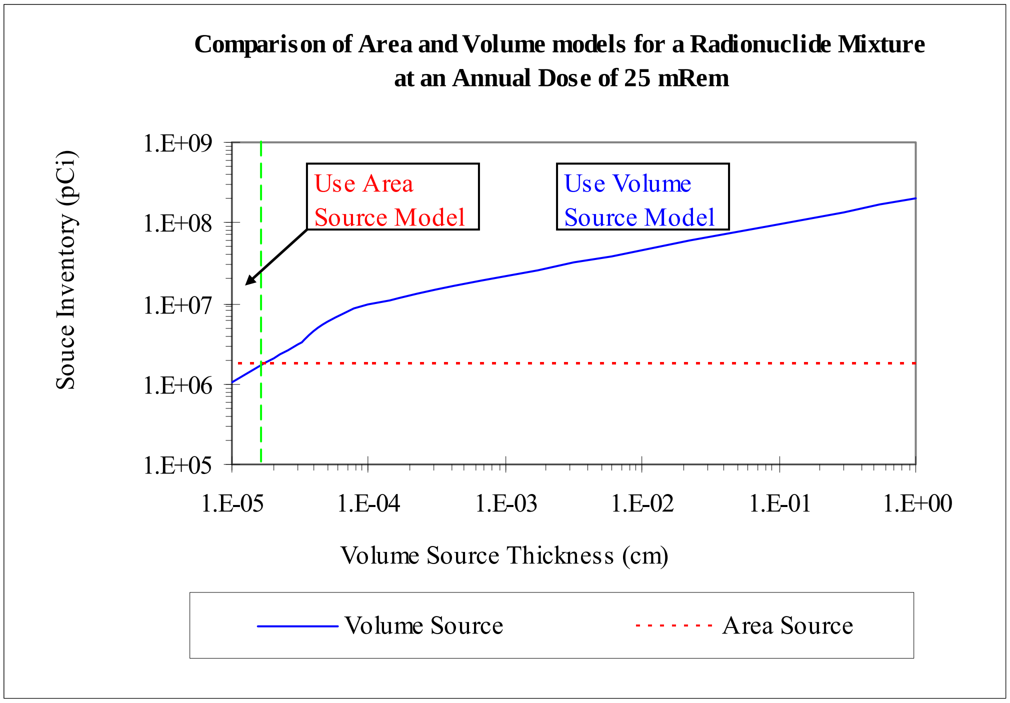

Figure 6. Comparison of Area and Volume Source Models for Mixture

DISCUSSION OF RESULTS FOR SET #3

Consideration of the results for the typical fission product mixture chosen as an example is also instructive. Table V gives the mixture chosen with Cs-137 as the primary radionuclide with respect to percentage of activity and Am-241 as a minor contributor to the total activity. Figure 7 gives the percent contribution to dose for each of the three radionuclides in the mixture as a function of the source thickness. For source thicknesses greater than about 0.1 cm, Cs-137 contributes over 90% of the dose. As the source thickness decreases contribution from Am-241 becomes more important and eventually contributes essentially the entire dose. However where Cs-137 had a “Point of Equivalency” at about 0.5 cm but for the mixture the “Point of Equivalency” is about 0.00002 cm which is the same value as for the Am-241 case calculation. From this it is clear that an appropriate mixture of radionuclides must be considered when making the determination as to whether use of the Volume or Area source models is appropriate to derive the site specific DCGLs for the particular situation.

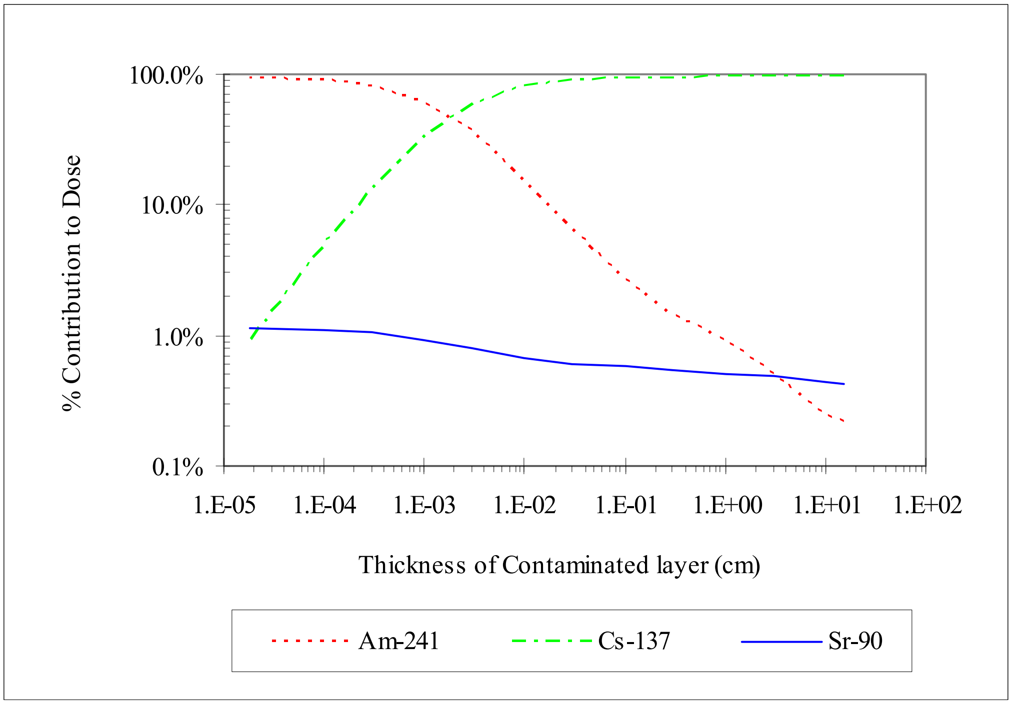

Figure 7. Percent contribution to dose of the radionuclides in the mixture case

Table VI provides the percent contribution to dose for each radionuclide as calculated by both the Volume and Area models at the Volume Source thickness of 0.00001786 cm. This is the point at which it was determined the two lines cross over in Figure 6 for the selected mixture. This table demonstrates that the RESRAD-Build computer code appears to be internally consistent in treatment of dose calculations in the two models compared in this paper at the "Point of Equivalency" as defined in this paper.

Table VI. Percentage contribution to dose for the mixture using the two models at the "Point of Equivalency

Radionuclide	**Percent contribution to dose for the Volume Model**	**Percent contribution to dose for the Area Model**
Sr-90	1.1%	1.1%
Cs-137	1.0%	1.0%
Am-241	97.9%	97.9%

CONCLUSIONS

Table VII provides a summary of the Volume Source thickness that is the "Point of Equivalency" for the six example cases calculated in this paper.

Table VII: Summary of Volume Source thickness at "Point of Equivalency"

Case Identity	Volume Source Thickness at "Point of Equivalency" (cm)
H-3	2.5 x 10+2
Fe-55	2 x 10-5
Sr-90	3 x 10-5
Cs-137	5 x 10-1
Am-241	2 x 10-5
Mixture	2 x 10-5

These conclusions should be considered preliminary because they are based on calculations that used the RESRAD-Build code with the default parameters rather than a site-specific case. At this point no effort has been made to quantify the effect of changing various code parameters on the cross over point between the Volume and Area models.

1. If the only radionuclide present as contamination is a high energy gamma emitter then it would probably be appropriate to use the DCGL calculated using the Area Model to demonstrate compliance with the regulatory criteria unless the penetration into the surface is greater than about 5 mm.
2. For radionuclide contamination that involves alpha emitters, pure beta emitters, electron capture decay and low energy photon emitters it appears to be more appropriate to utilize DCGLs derived using the Volume Model unless the surface is such that essentially no penetration is possible.
3. Tritium contamination of a surface appears to be a different case. For this radionuclide the use of the DCGL derived using the Area Model appears to be limiting even for deep penetration of the H-3 into the surface. Consideration of the practical problems associated with performing surface measurements for H-3 may lead to other considerations. It may be more appropriate to perform volumetric measurements in specific circumstances.
4. When there is a mixture of radionuclides present, it would be necessary to consider the appropriate mixture in the determination of which model to use for the specific situation.
5. The appropriate measurement technique for Final Status Surveys will have to be consistent with the models chosen to calculate the DCGLs.