Selection of Criticality Source Term & Modeling Parameters

by Wayne Davis (wayne.davis@wsms.com)

The following monologue is from my personal experience in modeling nuclear criticality accidents – evaluating criticalities involving racked fuel in fuel pools and solution criticalities. Other analysts may evaluate scenarios that are not applicable to this discussion. In other words (in the vernacular), your mileage may vary.

When estimating the source term for nuclear criticality event, it's first necessary to determine the size of the criticality event itself. This is measured in number of fissions. Each nuclear fission results in two or more fission products. These fission products may then decay into other isotopes. Some of these isotopes may be either gaseous or volatile. Depending upon where the criticality takes place (e.g., in a solution or a solid), some fraction of these fission products may be released. For example, fission products in a fuel assembly may be bound up in the fuel matrix and not readily released while those in a solution may easily come out of solution.

I've found that there can be quite a range in the number of fissions postulated for an event. As some analysts are more conservative than others, I've seen estimates that vary by several orders of magnitude. In general, I try to use the fission yield postulated in the Safety Basis documents (e.g., DSA). In some cases, I found their estimate to be too high and used a lower value, which I then justified in the EPHA.

DOE O 151.1C does not directly address the criticality *source term*. It addresses only the requirement that an accidental nuclear criticality be categorized as a non-classifiable Operational Emergency. However, this precludes a release that would drive classification. If an accidental criticality releases fission products to the environment sufficient to exceed Protective Action Criteria, it must be classified as would any other hazardous material release.

From DOE G 151.1-2 *Technical Planning Basis*:

> *2.4 Characterize the Hazardous Materials*
>
> … For criticality accidents, the "inventory" of interest is the total yield of gaseous and volatile fission products from the postulated criticality. Analyses of these postulated criticality events would generally be available in the facility SAR, SAD, or DSA.
>
> Where the material consists of a reactor core or irradiated fuel containing mixed fission products, relevant factors that define the radiotoxicity of the mixture (e.g., enrichment, burn-up, age) should be analyzed and the case that produces the largest impact selected. The actual isotopic composition of the mixture used for consequence calculations can then be included as an appendix and referenced.
>
> *2.6.1 Estimates of Source Terms*
>
> The source term (i.e., release to the environment) associated with MAR primary barrier failure mode, initiating event, and release condition should be calculated. For each possible failure mode of the primary barrier (for example, puncture, corrosion/ oxidation, explosive shattering), the release fraction or release rate from the DOE source term handbook, DOE-HDBK-3010-94. can be used to estimate the amount of radioactive material that would become airborne.

When I first started estimating consequences from criticalities, I used source terms from Authorization Basis documents (e.g., DSA). I soon found that these source terms varied

widely – not just from one facility to another, but from one DSA revision to another. An analyst would rerun a criticality code (e.g., ORIGEN) and come up with a different source term – often starting with the same yield. Then there's the issue of what source term to use from the criticality run. The source term (i.e., activity) varies dramatically as the time since criticality increases. I eventually decided to standardize on the source term from the DOE Handbook. The DOE Handbook criticality source term is taken from NRC Reg Guides that were withdrawn back in 1998. Though withdrawn, none have taken their place and they are still valid. Applicable tables from these Reg Guides are reproduced in the DOE Handbook.

HotSpot allows us to create a mixture file with all of the isotopes included in Handbook tables. A sample mixture file is included in the standard HotSpot installation; it's for a plutonium criticality. I found the uranium solution source term to bound the plutonium source term, so I use the same mixture file for all solution criticalities. I use a different mixture file for criticalities involving solids (e.g., fuel assemblies) as release fractions are smaller (due to holdup in the matrix).

Keep in mind that HotSpot includes a *mixture scale factor*. Steve Homann was kind enough to add this at my request. I got tired of having to re-create mixture files for every fission yield. This field allows the user to scale the number of fissions up or down in the input file without having to recalculate and input all of the source terms in the mixture file. For example, let's say I include source terms from the Handbook for 1E19 fissions in my mixture file. If I want to then perform a run for 1E18 fissions, I simply change the mixture scale factor from 1.0000E+00 to 1.0000E-01 (as with all HotSpot inputs, I suggest you keep format intact – don't omit any zeros).

One of the problems encountered when attempting to best model the source term given in the Handbook is that the yield is results from: *an initial burst of 1E+18 fissions followed by 47 bursts of 1.9E+17 fissions at ten minute intervals for eight hours for a total of 1E+19 fissions*.

HotSpot will not allow a source term to be entered in this manner. As we know, increasing the duration of a release accounts for plume meander and thus reduces the consequence to the receptor. To try to find the best solution, I modeled the source term in multiple ways. I initially modeled each contribution over its particular duration, then summed the results. I found that modeling the entire 8-hr source term over a duration of 8 hrs (480) minutes was within 10% of the sum of each contribution over its release duration. Therefore I now model the source term as one release with a duration of 8 hrs.

The next consideration is one of groundshine dose. As we know, we must include 4 days of groundshine in the PAG dose. While the noble gases do not contribute to groundshine, the iodine does. HotSpot has long had the option to include 4 days of groundshine. The problem I encountered was that for some onsite receptors, if I included 4 days of groundshine, I could exceed the PAG. I had to turn off the 4-day groundshine dose and add in a groundshine dose over a more reasonable two hours. The upcoming version of HotSpot will allow the user to specify the duration of groundshine dose; that will alleviate that problem.

Another consideration is that the most recently published ICRP DCFs do not include values for all isotopes in the criticality source term. DCFs for Kr-89 and Xe-137 are missing. I contacted Dr. Keith Eckerman at ORNL. He is a primary author of ICRP and EPA

documents that provide the DCFs used in HotSpot (and NARAC). He sent new (unpublished) values which I then included in my mixture files. I've attached that email.

I won't go into much detail here, but modeling a release from a solid requires consideration of the lower release fractions involving the matrix of the solid (e.g., metal or oxide). If this is something you need to do, pay particular attention to subsection 6.3.2 of the Handbook. Only the portion of the solid that melts or softens releases its inventory. For example, if a fuel assembly goes critical, assume that only 10% (0.1 fraction – **not** 0.01 as the Handbook erroneously states) of the solid softens. Handbook Table 6-10 has ARF values for various fission products. Let's take Kr, a noble gas. It has an ARF of 0.5. If a fuel assembly has an inventory of 1000 Ci and goes critical, assume that 10% of the solid softens. The inventory released (e.g., to moderator) would be 1000 Ci x 0.1 (DR) x 0.5 (ARF) = 50 Ci.

If anyone would like more info on modeling releases from solids, such as spent fuel, and the effect of liquids on reduction of iodine (due to it going into solution), feel free to contact me.

"Eckerman, Keith F."
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11/13/2008 04:17 PM

To wayne.davis@srs.gov
cc
Subject RE: DCFs for Kr-89 & Xe-137

Wayne-

Attached are the requested DCFs. These were calculated in the manner of Federal Guidance Report 12 using nuclear decay data which will appear later this year (probably next year but as a 2008) publication of the ICRP.

A meeting was cancelled and I was working on the software package that creates these tables so you get them sooner than expected. Let me know if you see any problems with the files or have any questions.

Regards,
Keith

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Kr-89 External Dose Rate Coefficients (Sv/s)

Organ	Air Submersion (Bq/m3)	Water Immersion (Bq/m3)	-------- Surface (/Bq/m2)	--------- Ground 1 cm	Ground Volume 5 cm	--------- Source 15 cm	--------- (Bq/m3) Infinite
R_Marrow	9.76E-14	2.11E-16	1.74E-15	1.15E-17	3.32E-17	5.41E-17	6.64E-17
Adrenals	8.32E-14	1.80E-16	1.51E-15	1.04E-17	2.98E-17	4.80E-17	5.87E-17
Bone_Sur	1.42E-13	3.10E-16	2.36E-15	1.53E-17	4.55E-17	7.60E-17	9.41E-17
Brain	1.05E-13	2.27E-16	1.61E-15	1.12E-17	3.31E-17	5.40E-17	6.62E-17
Breasts	1.09E-13	2.37E-16	1.75E-15	1.18E-17	3.52E-17	5.84E-17	7.23E-17
Gb_Wall	8.48E-14	1.84E-16	1.54E-15	1.00E-17	2.87E-17	4.74E-17	5.82E-17
Esophagu	8.53E-14	1.85E-16	1.48E-15	9.81E-18	2.88E-17	4.65E-17	5.65E-17
St_Wall	8.94E-14	1.94E-16	1.63E-15	1.05E-17	3.05E-17	4.95E-17	6.05E-17
Si_Wall	8.24E-14	1.79E-16	1.62E-15	1.03E-17	2.93E-17	4.72E-17	5.76E-17
Uli_Wall	8.45E-14	1.83E-16	1.63E-15	1.04E-17	2.97E-17	4.79E-17	5.85E-17
Lli_Wall	8.36E-14	1.81E-16	1.66E-15	1.05E-17	3.00E-17	4.84E-17	5.91E-17
Ht_Wall	8.90E-14	1.93E-16	1.60E-15	1.06E-17	3.04E-17	4.91E-17	5.98E-17
Kidneys	8.94E-14	1.94E-16	1.65E-15	1.07E-17	3.11E-17	5.06E-17	6.19E-17
Liver	9.03E-14	1.96E-16	1.63E-15	1.07E-17	3.08E-17	5.01E-17	6.13E-17
Lng_Tiss	9.84E-14	2.13E-16	1.70E-15	1.13E-17	3.31E-17	5.38E-17	6.60E-17
Ovaries	8.35E-14	1.81E-16	1.58E-15	1.05E-17	2.97E-17	4.70E-17	5.70E-17
Pancreas	8.19E-14	1.77E-16	1.55E-15	1.00E-17	2.86E-17	4.62E-17	5.64E-17
Skin	2.06E-13	3.43E-16	1.47E-14	3.13E-17	5.61E-17	8.08E-17	9.58E-17
Spleen	9.05E-14	1.96E-16	1.63E-15	1.06E-17	3.10E-17	5.06E-17	6.19E-17
Testes	9.74E-14	2.11E-16	1.83E-15	1.17E-17	3.47E-17	5.78E-17	7.18E-17
Thymus	9.42E-14	2.04E-16	1.62E-15	1.08E-17	3.18E-17	5.17E-17	6.31E-17
Thyroid	1.01E-13	2.18E-16	1.66E-15	1.14E-17	3.18E-17	5.13E-17	6.24E-17
Ub_Wall	8.49E-14	1.84E-16	1.72E-15	1.05E-17	3.03E-17	4.93E-17	6.07E-17
Uterus	8.11E-14	1.76E-16	1.60E-15	1.01E-17	2.88E-17	4.65E-17	5.70E-17
Muscle	9.58E-14	2.08E-16	1.82E-15	1.15E-17	3.31E-17	5.43E-17	6.68E-17
H Sub R	9.57E-14	2.07E-16	1.80E-15	1.14E-17	3.30E-17	5.39E-17	6.64E-17
E	9.56E-14	2.06E-16	1.84E-15	1.13E-17	3.26E-17	5.33E-17	6.54E-17

Xe-137 External Dose Rate Coefficients (Sv/s)

Organ	Air Submersion (Bq/m3)	Water Immersion (Bq/m3)	-------- Surface (/Bq/m2)	--------- Ground 1 cm	Ground Volume 5 cm	--------- Source 15 cm	--------- (Bq/m3) Infinite
R_Marrow	9.32E-15	2.01E-17	1.95E-16	1.26E-18	3.57E-18	5.50E-18	6.19E-18
Adrenals	8.05E-15	1.73E-17	1.70E-16	1.13E-18	3.14E-18	4.80E-18	5.40E-18
Bone_Sur	1.69E-14	3.67E-17	3.01E-16	1.92E-18	5.72E-18	9.21E-18	1.04E-17
Brain	1.02E-14	2.21E-17	1.80E-16	1.24E-18	3.56E-18	5.50E-18	6.18E-18
Breasts	1.11E-14	2.39E-17	2.03E-16	1.35E-18	3.98E-18	6.28E-18	7.10E-18
Gb_Wall	7.92E-15	1.71E-17	1.72E-16	1.04E-18	3.00E-18	4.62E-18	5.21E-18
Esophagu	7.98E-15	1.72E-17	1.63E-16	1.06E-18	3.05E-18	4.61E-18	5.15E-18
St_Wall	8.60E-15	1.85E-17	1.81E-16	1.15E-18	3.26E-18	5.02E-18	5.63E-18
Si_Wall	7.70E-15	1.66E-17	1.77E-16	1.10E-18	3.07E-18	4.68E-18	5.24E-18
Uli_Wall	8.01E-15	1.72E-17	1.80E-16	1.13E-18	3.15E-18	4.80E-18	5.37E-18
Lli_Wall	7.83E-15	1.68E-17	1.83E-16	1.13E-18	3.16E-18	4.84E-18	5.43E-18
Ht_Wall	8.51E-15	1.83E-17	1.79E-16	1.17E-18	3.25E-18	4.97E-18	5.57E-18
Kidneys	8.65E-15	1.86E-17	1.83E-16	1.18E-18	3.36E-18	5.20E-18	5.84E-18
Liver	8.70E-15	1.87E-17	1.82E-16	1.18E-18	3.33E-18	5.13E-18	5.75E-18
Lng_Tiss	9.64E-15	2.08E-17	1.93E-16	1.27E-18	3.61E-18	5.57E-18	6.25E-18
Ovaries	7.44E-15	1.60E-17	1.88E-16	1.11E-18	3.03E-18	4.62E-18	5.17E-18
Pancreas	7.65E-15	1.64E-17	1.68E-16	1.07E-18	3.01E-18	4.59E-18	5.14E-18
Skin	1.29E-13	1.51E-16	1.55E-14	2.61E-17	2.89E-17	3.14E-17	3.22E-17
Spleen	8.75E-15	1.88E-17	1.82E-16	1.18E-18	3.35E-18	5.16E-18	5.78E-18
Testes	9.70E-15	2.09E-17	2.11E-16	1.32E-18	3.87E-18	6.14E-18	6.96E-18
Thymus	9.03E-15	1.95E-17	1.84E-16	1.21E-18	3.45E-18	5.33E-18	5.99E-18
Thyroid	9.89E-15	2.13E-17	1.98E-16	1.29E-18	3.44E-18	5.23E-18	5.86E-18
Ub_Wall	8.11E-15	1.75E-17	1.87E-16	1.13E-18	3.21E-18	4.96E-18	5.59E-18
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Muscle	9.42E-15	2.03E-17	2.08E-16	1.28E-18	3.64E-18	5.65E-18	6.37E-18
H Sub R	9.39E-15	2.03E-17	2.05E-16	1.27E-18	3.61E-18	5.60E-18	6.31E-18
E	1.04E-14	2.12E-17	3.48E-16	1.49E-18	3.78E-18	5.73E-18	6.42E-18

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Authorized Derivative Classifier

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ABSTRACT

Deposition velocity is a parameter used in atmospheric transport models to specify the amount an atmospheric constituent transferred from the atmosphere to the surface of the earth. The material may deposit on the surface of soil, water, or vegetation. The deposition may be the result of rainfall or diffusion. A method for the calculation of deposition velocity based upon the decrease in deposition with distance from a point source is presented. The method does not require a knowledge of the time over which the deposition occurs or the concentration of the material in the atmosphere. However, the method does assume that the deposition rate is proportional to the air concentration. The sensitivity to errors resulting from certain errors in the measurements and from violation of some of the assumptions of the model underlining the calculations are also be discussed.The method has been used to estimate the deposition velocity of I-129. Two sets of I-129 deposition data at various distances from the center of SRS were used in the calculations. The results indicate that the deposition velocity is near a value of 0.2 cm/s. This is consistent with the processes that are known to control iodine deposition.

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ESTIMATING DEPOSITION VELOCITY FROM ATMOSPHERIC RELEASESOF IODINE

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INTRODUCTION

Deposition velocity is a parameter used in atmospheric transport models to specify the amount an atmospheric constituent that is transferred from the atmosphere to the surface of the earth. The material may deposit on thesurface of soil, water, or vegetation. The deposition may be the result of rainfall or diffusion. The deposition velocity can be specified as an average of all these processes or can be specified for certain conditions, i.e. the dry deposition velocity. The need for integrated, average deposition velocity or process specific deposition velocity depends on the type of model used. Deposition velocities for materials are often determined by field measurements using the meteorological methods of gradient or eddy flux or by measuring the accumulation of material on the surface over fixed periods of time. Deposition velocity is also calculated by summing the results of laboratory measurements of the uptake of the material by the individual elements of the surface, e.g. plant leaves and stems, soils, etc.

This report will discuss a method for the calculation of deposition velocity based upon the decrease in deposition with distance from a point source. The method does not require a knowledge of the time over which the deposition occurs or the concentration of the material in the atmosphere. However, the method does assume that the deposition rate is proportional to the air concentration. The sensitivity to errors resulting from certain errors in the measurements and from violation of some of the assumptions of the model underlining the calculations are also be discussed.The method has been used to estimate the deposition velocity of I-129. Two sets of I-129 deposition data at various distances from the center of SRS were used in the calculations. The results indicate that the deposition velocity is near a value of 0.2 cm/s. This is consistent with the processes that are known to control iodine deposition.

DISCUSSION

The model used in the calculation of deposition velocity is based on the assumption that, over a long period of time, and at some distance removed from the source, the dispersion of a material from a point source can be described as radial advection through an average, fixed layer of the atmosphere. Deposition from the atmosphere is proportional to the atmospheric concentration and the surface area at any radial distance from the source. The mass balance of material in the atmosphere, under these assumptions leads to

$$dC/C = [1.0/r + 2\pi\ v_d/(uL)]dr \qquad 1)$$

This can be solved for concentration as a function of distance

$$C/C_0 = (r/r_0)^{-n} \exp[-2 \pi v_d(r-r_0)/(uL)] \qquad 2)$$

where

n=1.0
C= concentration (pCi/cu m) or deposition (pCi/sq m)
r= radial distance from the source
v_d= deposition velocity
u= wind speed
L= mixing depth of the atmosphere
$_0$ = subscript indicating at a reference point.

Equation 2) specifies that concentration (or deposition which is proportional to concentration) decrease with distance is a straight line when plotted on logarithmic coordinates. This has been shown to be a reasonable approximation of decrease in concentration for a number of radionuclides monitored at SRS. However, the slope of the concentration with distance (logarithmic) usually is not equal to negative one, as would be expected for a material which remains in the atmosphere (is not deposited) as it disperses from SRS.

The Effect of Deposition Velocity

Figure 1. illustrates the effect of deposition on the decrease in concentration with distance from a source. The effect is to decrease the concentration more than that caused by the dilution from radial dispersion into a larger volume of air. The deviation from the case with no deposition will depend on the rate of deposition, i.e. the deposition velocity and the mixed depth of the atmosphere. If deposition is present, the decrease in concentration is not described by a straight line. The increasing surface area at greater distances from the source causes an increasing effect of deposition with distance from the source.However, short segments of the curve of decreasing concentration with distance can be approximated as straight lines with slope less than negative 1.0. This explains why field data often appears to plot as a straight line on logarithmic paper but with a slope other than negative 1.0.

The Effect of Background

The term background is used here as the concentration due to releases from other sources that are evenly distribute in the vicinity of the location where measurements are made. Figure 2. illustrates the effect of a background that is half the concentration at the point furthest from the source. The effect of the background is greater at lower concentration, i.e. further from the source. This will cause the curve describing the concentration decrease with distance to bend upward. A straight line fit to approximate this curve will have a slope greater than negative 1.0. Thus the effect of an uncorrected background will be in the opposite direction of the effect of deposition from the atmosphere.

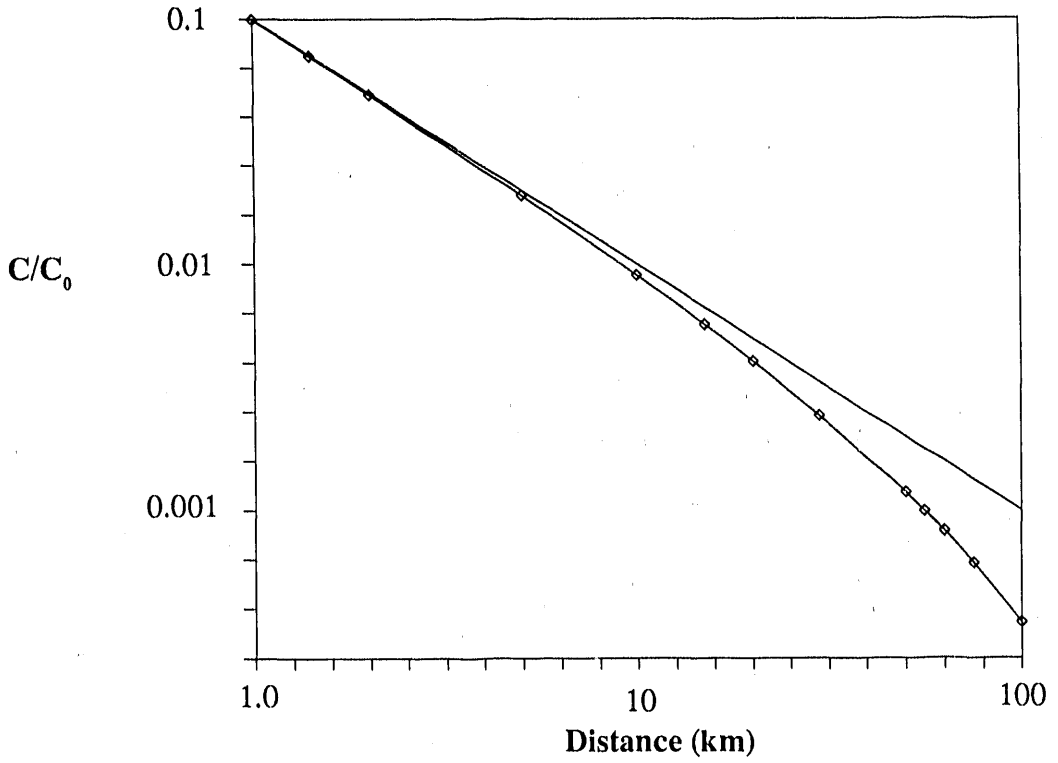

Figure 1. The effect of deposition on concentration versus distance from a point source.

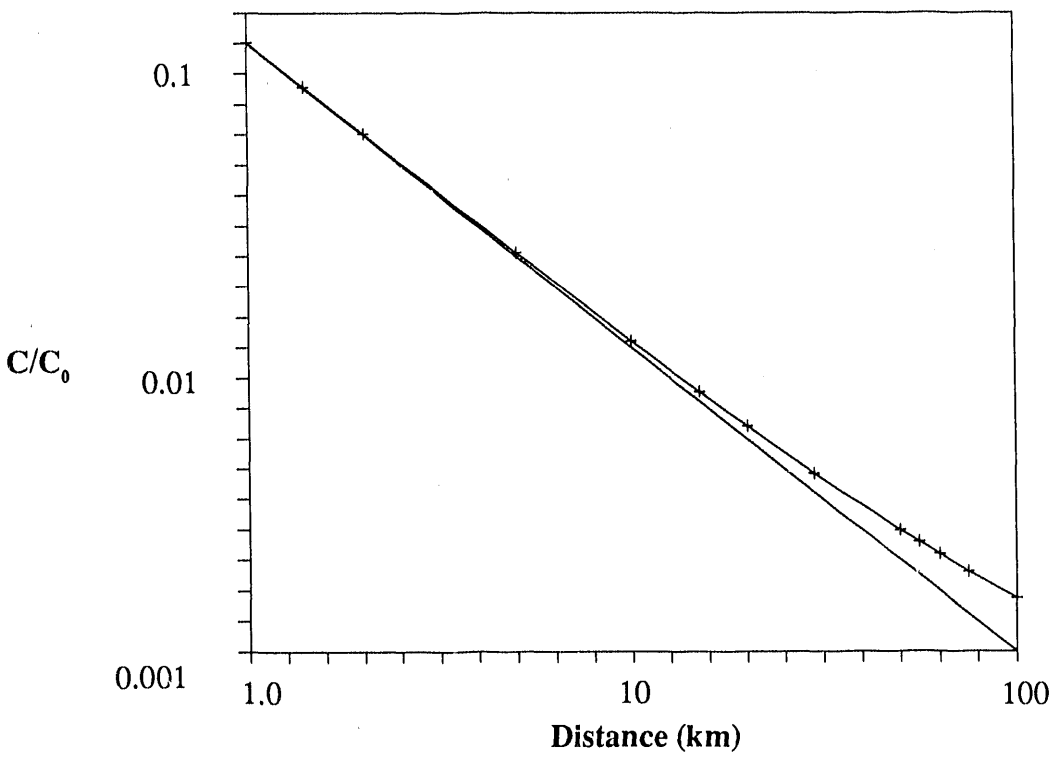

Figure 2. The effect of background on concentration versus distance from a point source.

The Effect of Vertical Mixing

If the assumption of complete mixing of the material in the atmosphere is not met, the concentration at the surface (and thus the deposition) will be great close to the source than the simple radial dispersion model would predict. In this case the slope of the line approximating the decrease in concentration with distance would be more negative than expected from radial dispersion. In this case, the effect of vertical mixing will be the same as the effect of deposition. If the source is from a stack or other elevated location, the material will not reach the ground for some distance away from the stack. In this case the concentration will increase with distance from the stack, finally reaching a maximum, and then decreasing with distance. The effect on the slope of a straight line approximation of concentration decrease with distance can be either to make the slope less negative near the source and more negative away from the source. This is illustrated in figure 3.

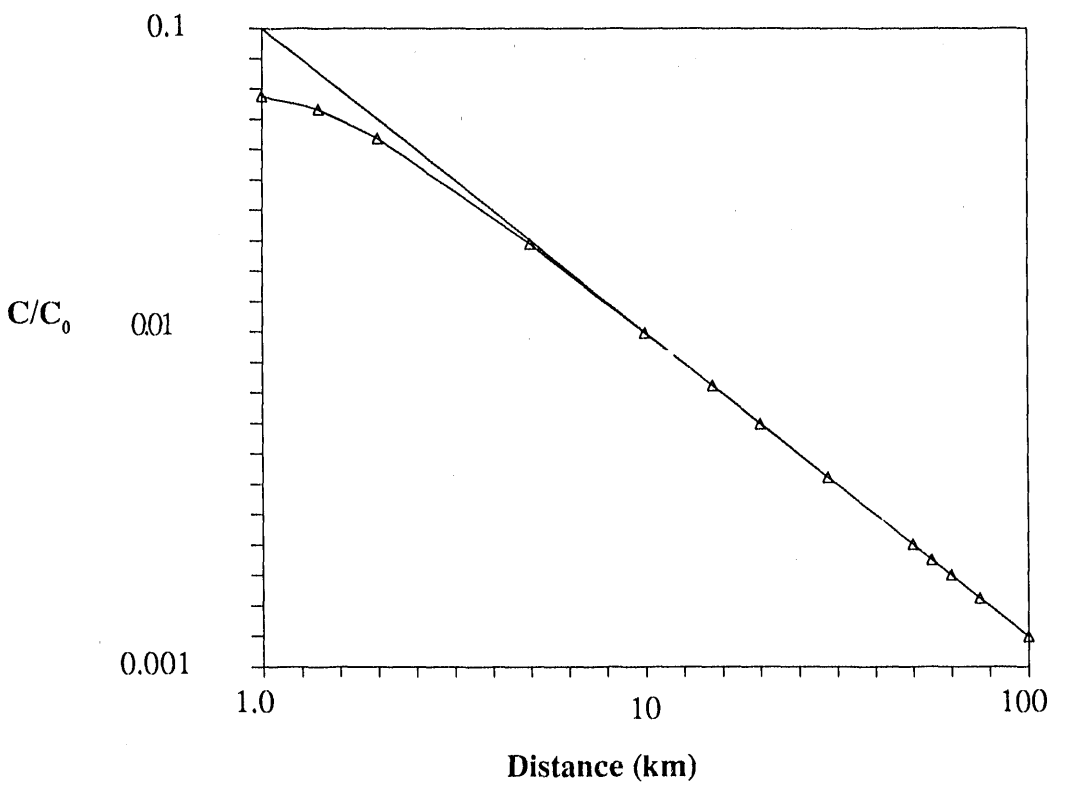

Figure 1. The effect of vertical mixing on concentration versus distance from a point source.

The effects of background and vertical mixing place some restraints on the use of equation 2) to estimate deposition. Either the background concentration of the material of interest must be very low, or well defined so that it can be subtracted from the measurements. Measurements should be made as far away from the source as possible to minimize the effect of vertical mixing and release height. To some extent these, these two restraints work against each other. The further measurements are made from the source, the more likely the background will become a significant fraction of the measurement.Table 1. lists the results of calculations of the slope of concentration with distance made from the curves in figure 1. through 3. The illustration used a mixing depth of 2000 meters, a wind speed of 3 m/s, a stack height of 60 meters, a deposition velocity of 1 m/s, and neutral atmospheric temperature stratification.

Table 1. The Effect of the Atmospheric and Measurement Conditions on the Apparent Slope of the Relationship between Concentration and Distance from the Source.

	Ideal	Background	Deposition	Mixing Depth
Distance (km)				
10-20	-1.00	-0.93	-1.15	-0.98
20-50	-1.00	-0.86	-1.34	-0.99
50-100	-1.00	-0.74	-1.76	-1.00

Estimation of I-129 Deposition Velocity

The deposition of I-129 in soils and vegetation has been made in the vicinity of SRS. The low specific activity of I-129 has made routine measurements difficult. However, the long half-life has meant that I-129 has accumulated, at low levels, in the soil around SRS. This material is recycled through the vegetation. Thus, the concentration of I-129 in soil and vegetation is a measure of the long_term deposition. The deposition velocity calculated from this data should be the integrated average for all processes involved in the exchange between the atmosphere and the surface. However, because of the relatively low amount of time involved with wet deposition (rainfall), the deposition velocity should be weighted towards the dry deposition velocity.The deposition velocity can be determined by rearranging equation 2).

$$v_d = - \ln [C/C_0/(r/r_0)^{-n}]/[2\pi(r-r_0)/(uL)] \qquad 2)$$

If the left hand side of equation 3) is plotted against the right hand side, the deposition velocity will be the slope of the straight line through the data points.

Table 2. shows the results of two measurements of I-129 at various distances from the center of SRS. Hochel's (1976) results are the average of a number of measurements taken at the indicated distances. Kantelo et al. (1981) results are two points taken along a straight line though the data plotted in logarithmic coordinates. Hochel's data is reported in units of concentration while Kantelo's in units of deposition.

The resulting deposition velocities are shown in Table 3. An average wind speed of 3 m/s and an average mixed depth o f the atmosphere of 540 meters was used in the calculations. The average wind speed (to the nearest m/s) was taken from Laurinat (1987). The average mixing depth was calculated from the environmental monitoring data, as summarized in Murphy et al. (1990), using the model described in this report. The resulting equation is:

$$L = F/(2\pi r u C) \qquad 3)$$

where F is the rate of tritium released during the period when the average tritium concentration, C was measured. The plant perimeter monitoring stations were used in the calculation. Notice that the estimate of wind speed and mixed layer are linked in the same way in equations 2) and 3). This means that any error in estimate of the wind speed will be compensated by the estimate mixing depth.

Table 2. I-129 Concentrations in the Vicinity of SRS

(Hochel, 1976)

Soil

r (km)	C (pCi/gm)	C/C_0	r/r_0	$(C/C_0)(r/r_0)$
20	0.021	1	1	1
40	0.005	2	0.238	0.476
160	0.001	8	0.047	0.381

Vegetation

r (km)	C (pCi/gm)	C/C_0	r/r_0	$(C/C_0)(r/r_0)$
20	0.009	1	1	1
40	0.004	2	0.444	0.888

(Kantelo et al. 1981)

r (km)	C (pCi/ sq m)	C/C_0	r/r_0	$(C/C_0)(r/r_0)$
20	109	1	1	1
40	48	2	0.440	0.881

SUMMARY

The results are suggest a mean deposition velocity of 0.2 cm/s. This is somewhat lower than the average deposition velocity for water vapor, which is around 0.4-0.8 cm/s. However, the molecular diffusivity of iodine vapor is about 5 time less than that of water vapor. When this is taken into account the results suggests that iodine is deposited by the same processes as determine the deposition of many other gases such as water vapor or SO_2, i.e vapor diffusion and deposition by solution inside vegetation and soil.

Table 3. I-129 Deposition Velocities Estimated from SRS Data.

	slope (eq. 3) km^{-1}	deposition velocity cm/s
Hochel (1976)		
soil	-0.00689	0.18
vegetation	-0.00588	0.15
Kantello et al. (1981)		
soil	-0.00634	0.16

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