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B. J. Schrader (INEEL)
D. R. Wenzel (DR Wenzel Consulting)

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# Evaluating the Dose Consequences of Progeny Ingrowth and Decay During Transport in the Environment - Counting Atoms

Bradley J. Schrader

Idaho National Engineering and Environmental Laboratory
Idaho Falls, Idaho 83415-2503
208-526-3399
bschrade@inel.gov

D.R. Wenzel DR Wenzel Consulting Idaho Falls, Idaho 83402 drwenzel@cableone.net

#### SUMMARY

The evaluation of a release of radioactive material to the environment is a complex problem. The evaluation of the dose consequences from such a release adds issues that must be considered in order to determine the actual radiation dose to an individual. One of the key issues is the ingrowth of the progeny from the initial inventory during the transport through the environment.

The evaluation of an acute release from a criticality accident and a chronic release of transuranics to the soil are the two extremes that demonstrate the spectrum of issues that must be addressed in order to determine the dose to a receptor at some distance and time from the release point.

The evaluation of this can be accomplished by integration of the buildup and decay differential equations. The differential equations are difficult to evaluate for a source term that contains numerous short-lived radionuclides in decay chains. The evaluation is simplified by a method of evaluation called "counting atoms". The source term is evaluated on an atom basis during production and transport. The life cycle of the atoms of each radionuclide are evaluated individually from the generation during the a criticality event and during

transport through the facility and environment to the downwind receptor.

## **BACKGROUND**

Criticality accidents, the most complicated type of environmental release, will be evaluated here. This evaluation method can be applied to any environmental release of radioactive material.

For simplicity, only the fission products will be evaluated. The method of evaluation can also be applied to the activation products and the actinides.

#### FISSION PRODUCT INVENTORY CALCULATION

Calculation of the buildup and decay of fission products during a criticality event can determined by the following set of simultaneous equations.

$$\begin{split} \frac{dQ_1}{dt} &= S_1 - \mu_1 \ Q_1 \\ \frac{dQ_2}{dt} &= S_2 - \mu_2 \ Q_2 + \lambda_1 \ Q_1 \\ \frac{dQ_i}{dt} &= S_i - \mu_i \ Q_i + \lambda_{i-1} \ Q_{i-1} \end{split}$$

where

Q<sub>i</sub> = the number of atoms for the I<sup>th</sup> radionuclide

t = time of the fissions (s)

S<sub>i</sub> = the source rate at which atoms are being produced by fission of the I<sup>th</sup> radionuclide (atoms/sec)  $\begin{array}{ll} \lambda_{i} & = & \text{the decay constant for the I}^{\text{th}} \\ & & \text{radionuclide (s$^{-1}$)} \\ \mu_{i} & = & \lambda_{i} + \sigma_{ci} \; \varphi \\ & & \sigma_{ci} = & \text{neutron capture cross} \\ & & & \text{section for the I}^{\text{th}} \end{array}$ 

section for the I<sup>th</sup> radionuclide (cm<sup>-2</sup>)  $\varphi$  = neutron flux (n/cm<sup>2</sup>-s).

The general solution as formulated by Rubinson (1949) for the contribution to the n<sup>th</sup> radionuclide from the m<sup>th</sup> radionuclide precursor is shown in the following equation.

$$Q_{n}(t) = S_{m} \begin{pmatrix} n-1 \\ \prod \\ i=m \end{pmatrix} (\lambda_{i}) \sum_{i=m}^{n} \begin{bmatrix} \frac{1-e^{-\lambda_{i}t}}{n} \\ \lambda_{i} \prod (\mu_{j} - \mu_{i}) \\ j=m \\ j \neq i \end{bmatrix}$$

By summing all the precursors of a particular nuclide, the general solution for the total number of atoms for the i<sup>th</sup> radionuclide of the decay chain is obtained.

$$Q_{k}(t) = \sum_{m=1}^{k} \left( S_{m} \prod_{i=m}^{k-1} (\lambda_{i}) \right) \sum_{i=m}^{k} \left[ \frac{1 - e^{-\lambda_{i}t}}{k} \frac{1}{\lambda_{i} \prod (\mu_{j} - \mu_{i})} \right]$$

For ease in evaluation of the integral, a linear operator is defined as

$$E_k = \sum_{m=1}^k \left( S_m \prod_{i=m}^{k-1} (\lambda_i) \right) \sum_{i=m}^k \left[ \frac{1}{\prod\limits_{\substack{j=m \\ j \neq i}} (\mu_j - \mu_i)} \right]$$

This allows the equation to be simplified to

$$Q_k(t) = E_k \left( \frac{1 - e^{-\lambda_i t}}{\lambda_i} \right)$$

### RADIONUCLIDE DECAY

The differential equations for the decay rate are similar to those for fission product buildup, except the source rate is considered to be zero. The general equations is

$$N_k(t) = E_k(e^{-\lambda_i t})$$

where

 $N_k(t)$  = the total number of atoms for the  $k^{th}$  radionuclide of the decay chain.

$$E_k(e^{-\lambda_i t}) = \sum_{m=1}^k Q_m \begin{bmatrix} k-1 \\ \Pi \\ i=m \end{bmatrix} \sum_{i=m}^k \begin{bmatrix} e^{-\lambda_i t} \\ \hline k \\ \Pi \\ j=m \\ j \neq i \end{bmatrix}$$

Q<sub>m</sub> = the total number of atoms for the m<sup>th</sup> radionuclide immediately following the cessation of fissioning

t = the decay time (s)

## TRANSPORT EQUATIONS

Atmospheric diffusion at ground level for a continuous point source can be expressed using the time-integrated form of the universal diffusion equation (Slade 1986) as follows:

$$\frac{\chi}{Q}(x,y,0) = \frac{1}{\pi \overline{u} \sigma_y \sigma_z} \exp \left[ -\frac{1}{2} \left( \frac{y^2}{\sigma_y^2} + \frac{h^2}{\sigma_z^2} \right) \right]$$

where

 $\chi/Q(x,y,0)$  = ground-level atmospheric diffusion relative to the initial point of release

x = distance downwind (m)

y = horizontal distance from plume centerline (m)

u = average windspeed at the release level (m/s)

σ<sub>y</sub>,σ<sub>z</sub> = standard deviations of effluent concentration of the plume in the horizontal and vertical directions (m)
 h = elevation of the point of release above the ground plane (m).

Further development of the atmospheric diffusion equations is left for other documents. For this discussion it is enough to say that the diffusion equation has a time dependency for a downwind receptor that is based on the wind speed and downwind transport distance.

This time dependency is why the ingrowth of progeny must be determined as a function of downwind distance. If equilibrium is assumed from the point of release the dose to the receptor may be significantly overestimated for distances less than about a 30 min transport time. In particular the distances out to 15 min, it is very important to allow for parent/progeny decay.

## **RESULTS**

Table 1 shows the difference in the concentrations for 1E18 fissions. The time dependence of Cs/Ba, Sr/Y and the halogens is shown over a time of 1 to 30 min.

Table 1
Time Dependence of Progeny Ingrowth (curies)

<u>Radio</u>			
nuclide	1min	15min	<u>30min</u>
Cs-137	5.34E-6	3.49E-5	3.73E-5
Ba-137m	7.27E-3	1.92E-4	3.77E-5
S-90	4.40E-6	3.43E-5	3.56E-5
Y-90	5.66E-6	5.72E-6	5.82E-6
I-131	6.71E-5	1.69E-3	4.73E-3
I-132	1.42E-2	1.82E-2	2.31E-2
I-133	2.52E-2	2.37E-2	3.53E-2
I-135	1.42E-0	1.47E-0	1.43E-0

Typical transport times to 1,600 meters (1 mile) range from 10 to 30 minutes. It is vital that the population exposure adjacent to the release account for the ingrowth of the progeny during transport. This can be done by watching the ingrowth of the progeny on a per atom basis between two points based on the transport time.

### CONCLUSION

It is very clear that unless the ingrowth of the progeny is accounted for in the transport of the release through the environment, the dose to the receptor can be significantly overestimated by assuming equilibrium. When the progney ingrowth is ignored, the dose to the receptor can be significantly underestimated.

A method for evaluation of the ingrowth of the progeny can be demonstrated by counting the atoms in the release and following each atoms buildup and decay during transport.

This method can be numerically integrated and is currently available and used by the atmospheric dispersion and transport code, RSAC (Schrader 2001).

#### REFERENCES

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