Scientific Notebook - Inventory Boosting Analysis

John Bradbury, 2010

DOE's three-dimensional model of radionuclide transport in the saturated zone did not simulate ingrowth of daughters from decay of parents along the flowpath (SNL, 2007). Instead a method termed "inventory boosting" was used to approximate the contribution of some daughters to radionuclide flux across the accessible environment boundary. Table 1 lists the parents and daughters, with their respective half lives that were modeled using the inventory boosting method.

Table 1	
Parent	**Boosted Daughter**
Am-241 (433 yrs)	Np-237 (2.14e6 yrs)
U-238 (4.47e9 yrs)	U-234 (2.46e5 yrs)
Pu-238 (87.7 yrs)	U-234 (2.46e5 yrs)
Am-243 (7,370 yrs)	Pu-239 (2.41e4 yrs)
Pu-240 (6,560 yrs)	U-236 (2.34e7 yrs)
Pu-242 (3.75e5 yrs)	U-238 (4.47e9 yrs)

Other parent daughter pairs included in DOE's License Application were modeled using the one-dimensional pipe element of GoldSim which can simulate ingrowth of daughters along the flowpath. See Scientific Notebook on Secular Equilibrium (Bradbury, 2010) for staff review of one-dimensional simulations.

In the year that the parent is introduced into the saturated zone, inventory boosting adds the amount of daughter that would be ingrown from the mass of introduced parent for the rest of the regulatory period. For example, if 1 g of Pu-242 is added to the saturated system in the first year, the amount of U-238 added in that year is the amount that the decay of Pu-242 would produce over the 1MY regulatory period. This is

$$\text{Mass U-238} = W(1\text{ g} - (1\text{ g})e^{-\lambda(1{,}000{,}000-t)}),$$

where t = the year the parent was introduced into the saturated zone, λ is the decay constant for Pu-242, and W is the ratio of the atomic weight of the daughter to that of the parent. At year zero, the mass of U-238 boosted is $1\text{ g} - (1\text{ g})e^{(-1.85\text{e-}6/\text{yr})(1{,}000{,}000\text{yr})} = 0.84$ g. At year 1,000,000, the mass of U-238 boosted is 0 g, because there is no time remaining for the Pu-242 to decay within the regulatory period.

In order to determine whether this method does not underestimate the flux from ingrown daughters, the staff performed one-dimensional simulations comparing the inventory boosted model with one that included ingrowth along the flowpath. Both models include advection, sorption and radioactive decay.

The GoldSim pipe element requires that mass input be expressed as cumulative. Consequently, if each year one gram is added to the pipe, then the cumulative quantity is 1g/yr*Time(yr). For the ingrown daughters, the cumulative input is derived from the definite integral of Equation 1.

$$Mass\ of\ Daughter/yr = W(1 - 1e^{-\lambda(1{,}000{,}000-t)}) = W - We^{(-1{,}000{,}000\lambda+\lambda t)} = W - W\frac{e^{\lambda t}}{e^{1{,}000{,}000\lambda}}$$

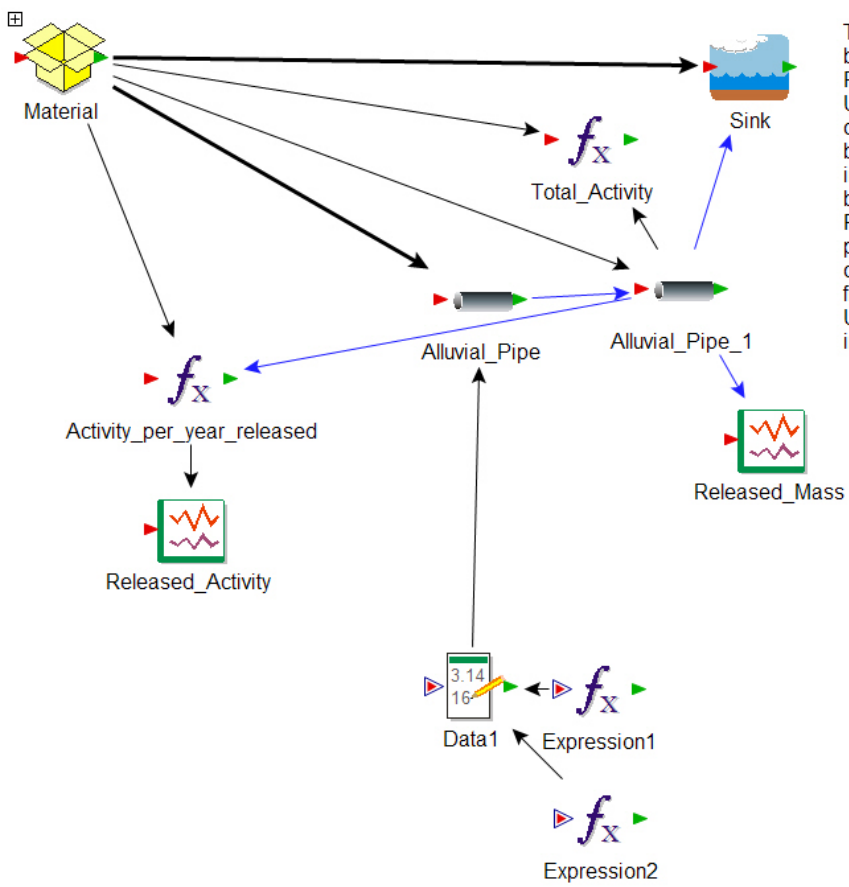
$$\int_0^t \frac{Mass\ of\ Daughter}{yr}dt = \int_0^t (W - W\frac{e^{\lambda t}}{e^{1{,}000{,}000\lambda}})dt$$

$$= \int_0^t W(g/yr)dt - W\frac{1}{e^{1{,}000{,}000\lambda}}\int_0^t e^{\lambda t}dt$$

$$= \left|{}_0^t W\left(\frac{g}{yr}\right)t - W\frac{1}{(e^{1{,}000{,}000\lambda})\lambda}\right|_0^t e^{\lambda t}$$

The GoldSim model used to study inventory boosting is illustrated in Figure 1. Two pipe elements in series represent a heterogeneous flow path along which the radionuclides are transported. The pipes contain sorptive material with properties similar to those used in DOE's 1-D transport model.

Figure 1



This example compares inventory boosting of U-238 and U-234 daughters of Pu-242 and U-238 respectively. For the U-238 daughter the method is not conservative after 800Ky when the boosting flux is less than the flux when ingrowth in the pipe is modeled. Inventory boosting of U-234 from the decay of Pu-238 failed because the number produced in the calculation exceeded the capacity of the code. The flux of U234 from inventory boosting via the decay of U238 exceeds the flux at all times when ingrowth in the pipe is considered.

The expression elements calculate the cumulative input of daughters based on Equation 2 when one gram per year of parent is added to the flow path.

Figure 2 shows that the boosted U-238 exceeds the ingrown U-238 for the period up to 800 Kyr. From then on until 1 MY, the ingrown U-238 exceeds the boosted U-238. However, the amount is insignificant relative to the amount of U-238 introduced into the saturated zone via transport from the unsaturated zone.

Figure 2

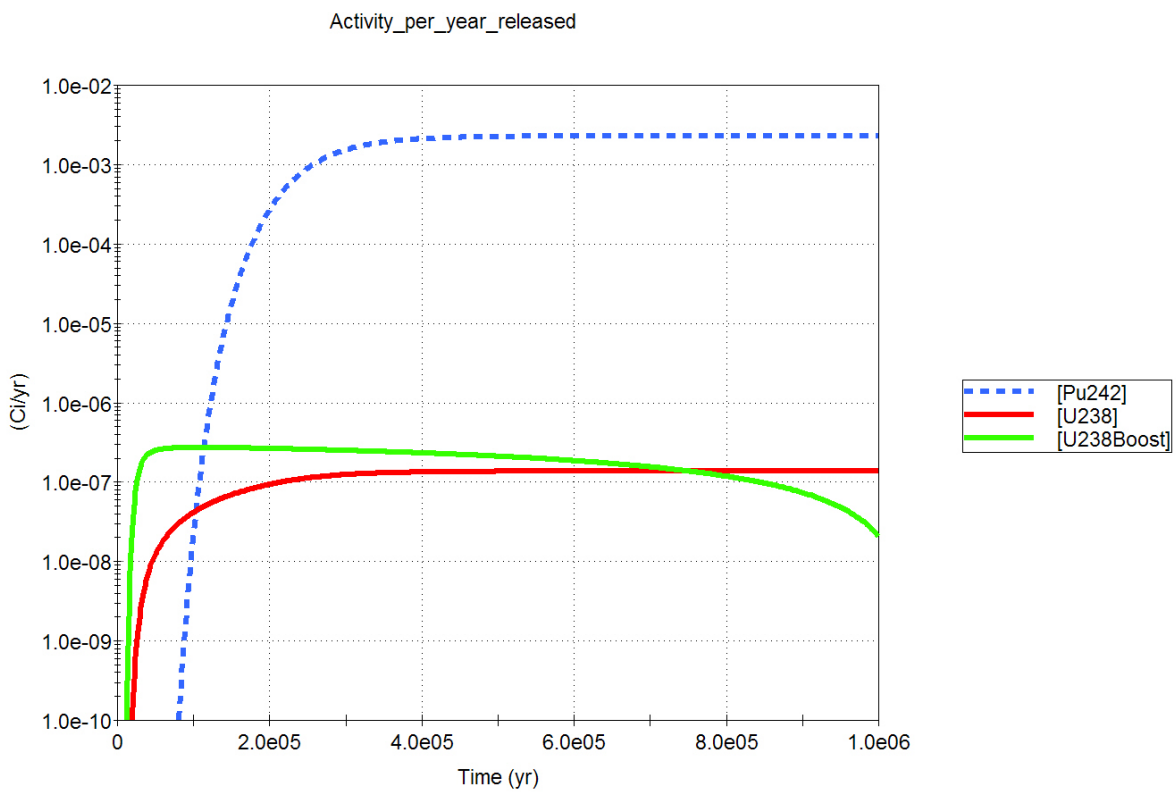
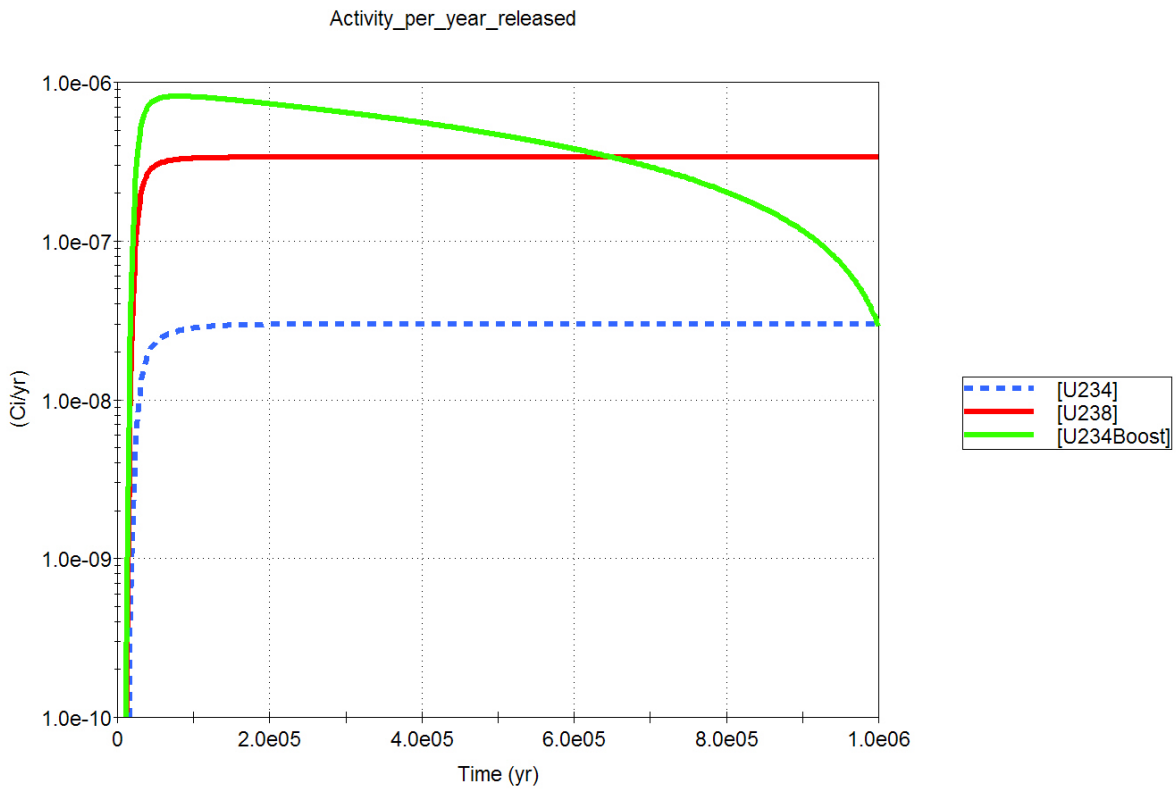


Figure 3 illustrates that boosting of U-234 from the decay of U-238 exceeds at all times the flux from the model that considers ingrowth of U-234 (blue dashed curve) in the pipe.

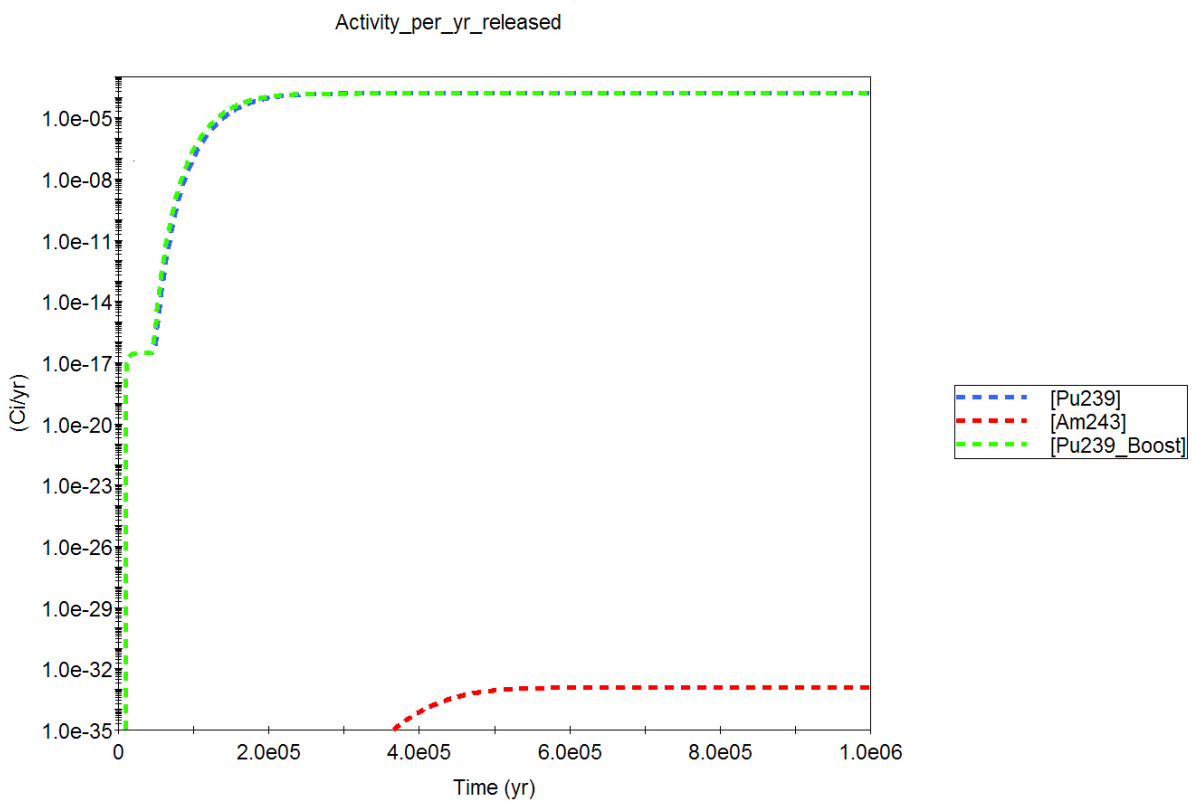
Figure 3



Pu-238 to U-234 cannot be calculated because the decay constant of Pu-238 is so great that $e^{\lambda t}$ is greater than can be handled in GoldSim. Furthermore, DOE's model indicated no Pu-238 reached the saturated zone within 1MY.

Figure 4 illustrates that boosted Pu-239 is essentially the same as the ingrown Pu-239 generated from the decay of Am-243 in the pipe. The low flux of Am-243 is a result of its short half life and its large sorption coefficient (1000 ml/g).

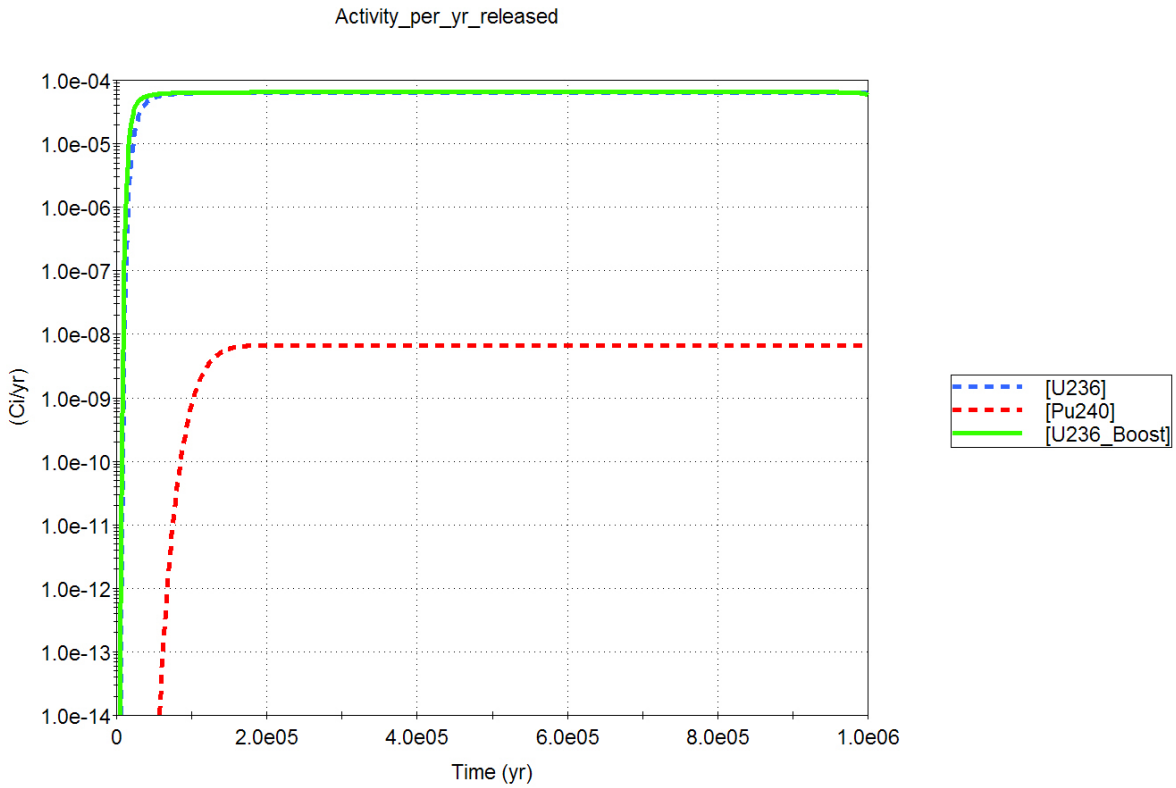
Figure 4



From the Postclosure Ground Water Impact Analysis of the Supplemental Environmental Impact Statement (Sandia, 2009), the cumulative mass of Am 241 introduced into the saturated zone is only 0.165g for 1MY. The cumulative mass of Np-237 is 1.02E6 g for 1MY. Consequently, boosting Np-237 by the decay of Am-241 is insignificant and a separate calculation to compare the two methods was not performed.

Figure 5 compares the flux of U-236 from the inventory boosted method and the ingrowth method in the flow path. The two fluxes are essentially coincident. The lower flux of Pu-240 is due to its short half life. Most of it decays to U-236 before exiting the pipe.

Figure 5



References

Bradbury, J. 2010, Scientific Notebook – Secular Equilibrium Assumption Analysis. ML110820653

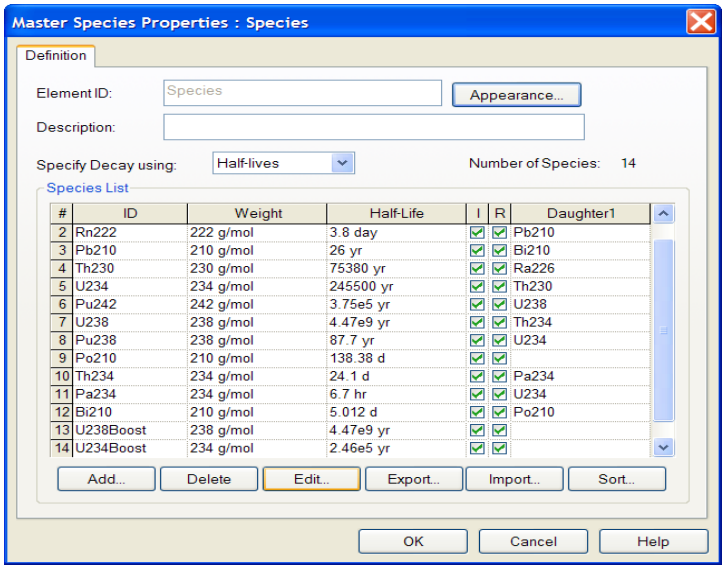
Sandia, 2009, Inputs to Jason Associates Corporation in Support of the Postclosure Repository Supplemental Environmental Impact Statement. LSA-AR-037 Rev 00. Las Vegas, Nevada. DOE, Office of Civilian Radioactive Waste Management. [included in DOE, 2009c, ML1111082A018]

SNL, 2007. Site-Scale Saturated Zone Transport. MDL-NBS-HS-000010 REV 03 ADD 01. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20080121.0003. ML090770725

Appendix

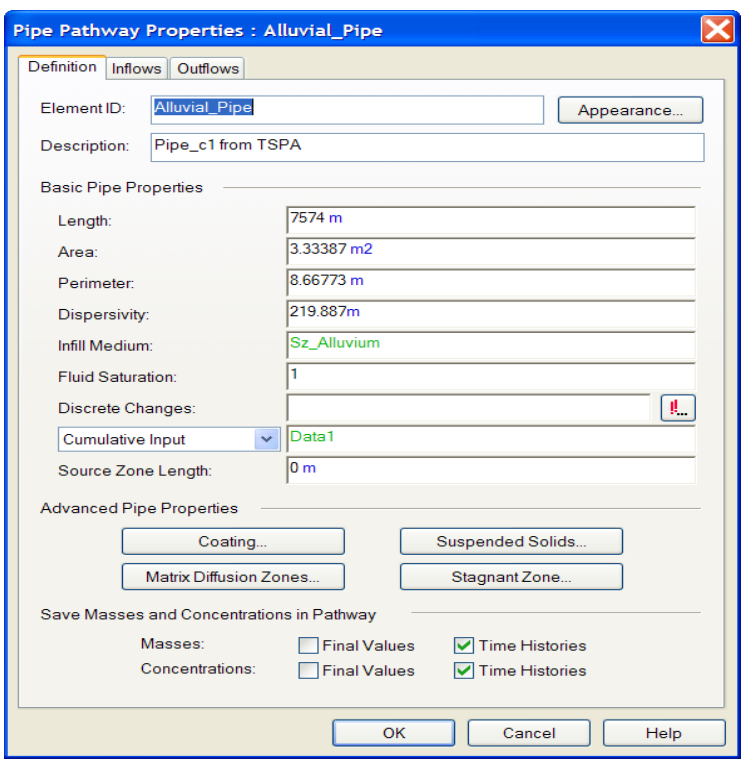
The figures in this Appendix provide details of the GoldSim elements used in the models.

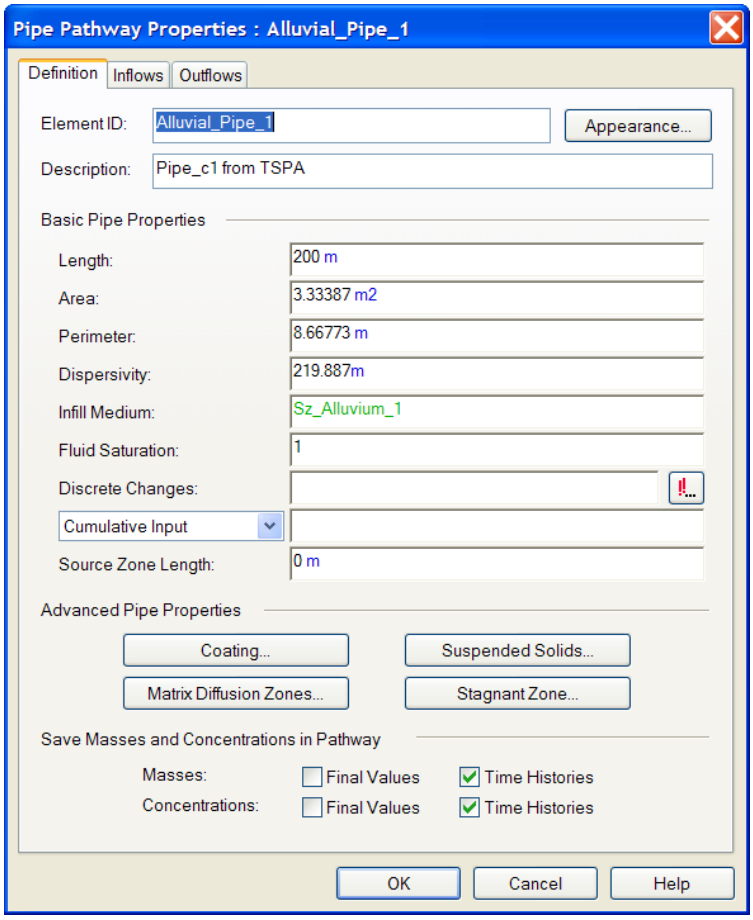
Uranium Series GoldSim Elements

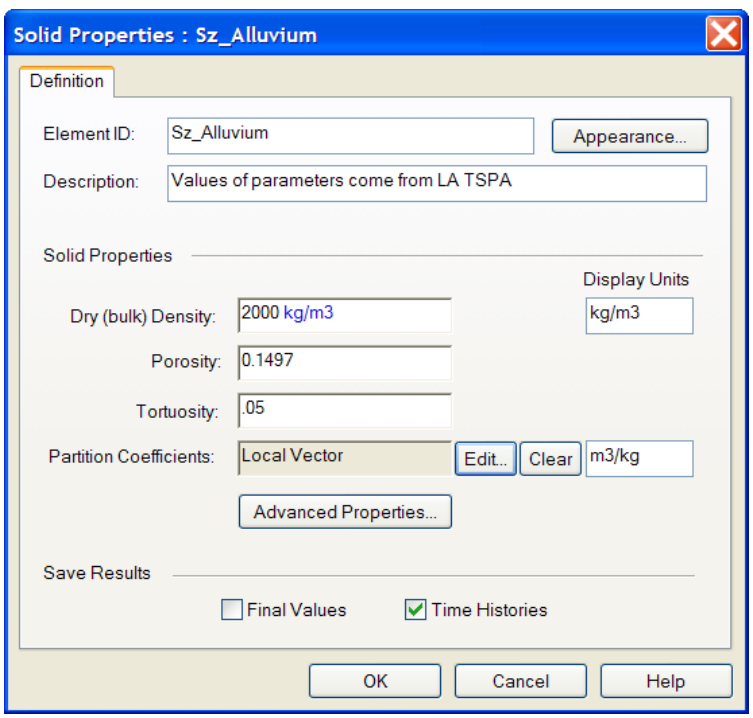


#	ID	Weight	Half-Life	I	R	Daughter1
2	Rn222	222 g/mol	3.8 day	✓	✓	Pb210
3	Pb210	210 g/mol	26 yr	✓	✓	Bi210
4	Th230	230 g/mol	75380 yr	✓	✓	Ra226
5	U234	234 g/mol	245500 yr	✓	✓	Th230
6	Pu242	242 g/mol	3.75e5 yr	✓	✓	U238
7	U238	238 g/mol	4.47e9 yr	✓	✓	Th234
8	Pu238	238 g/mol	87.7 yr	✓	✓	U234
9	Po210	210 g/mol	138.38 d	✓	✓	
10	Th234	234 g/mol	24.1 d	✓	✓	Pa234
11	Pa234	234 g/mol	6.7 hr	✓	✓	U234
12	Bi210	210 g/mol	5.012 d	✓	✓	Po210
13	U238Boost	238 g/mol	4.47e9 yr	✓	✓	
14	U234Boost	234 g/mol	2.46e5 yr	✓	✓	

The figure above does not show the first species, which is Ra-226. It has a half life of 1600 yrs and decays to Rn-222.




Pipe Pathway Properties : Alluvial_Pipe_1
Definition
Inflows
Outflows
Element ID:
Alluvial_Pipe_1
Appearance...
Description:
Pipe_c1 from TSPA
Basic Pipe Properties
Length:
200 m
Area:
3.33387 m2
Perimeter:
8.66773 m
Dispersivity:
219.887m
Infill Medium:
Sz_Alluvium_1
Fluid Saturation:
1
Discrete Changes:
Cumulative Input
Source Zone Length:
0 m
Advanced Pipe Properties
Coating...
Suspended Solids...
Matrix Diffusion Zones...
Stagnant Zone...
Save Masses and Concentrations in Pathway
Masses:
Final Values
Time Histories
Concentrations:
Final Values
Time Histories
OK
Cancel
Help


Solid Properties : Sz_Alluvium
Definition
Element ID:
Sz_Alluvium
Appearance...
Description:
Values of parameters come from LA TSPA
Solid Properties
Display Units
Dry (bulk) Density:
2000 kg/m3
kg/m3
Porosity:
0.1497
Tortuosity:
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Partition Coefficients:
Local Vector
Edit...
Clear
m3/kg
Advanced Properties...
Save Results
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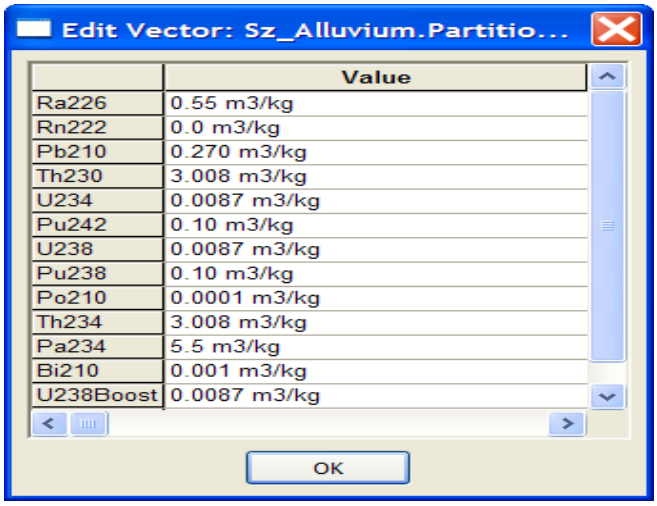
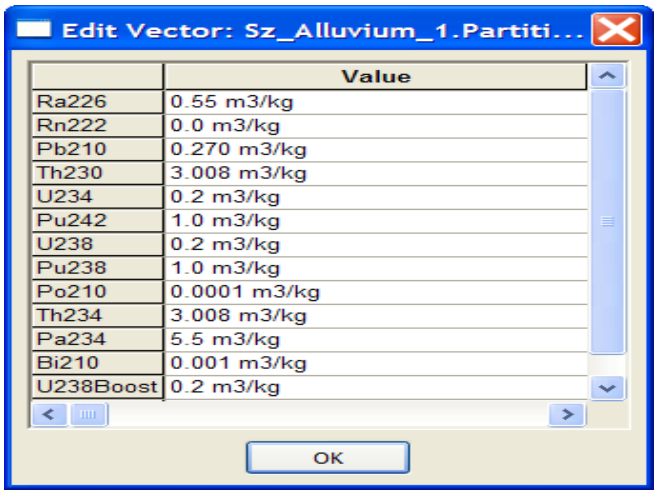
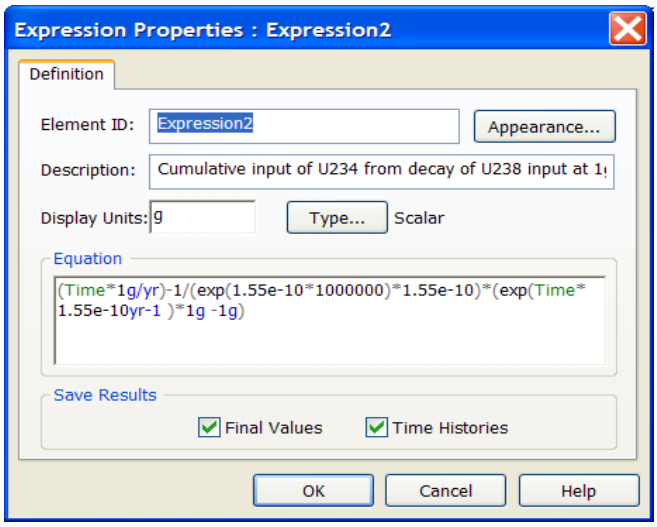
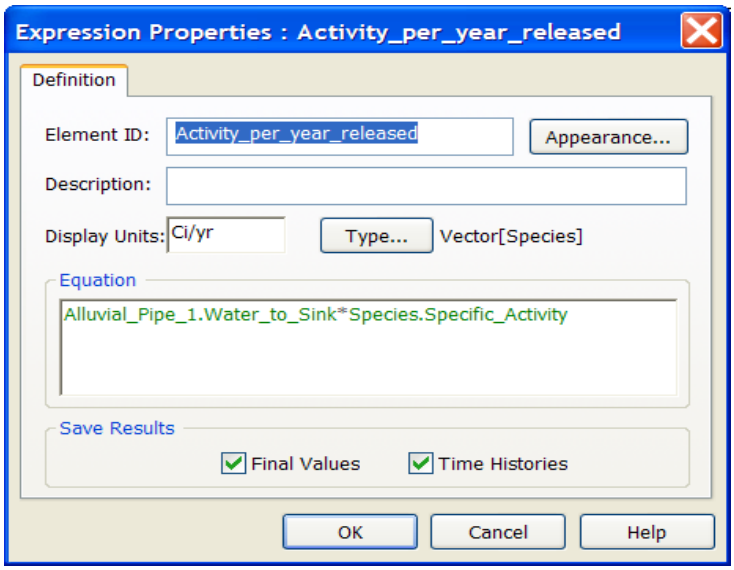


The Solid Properties of Sz_Alluvium_1 is the same as Sz_Alluvium.

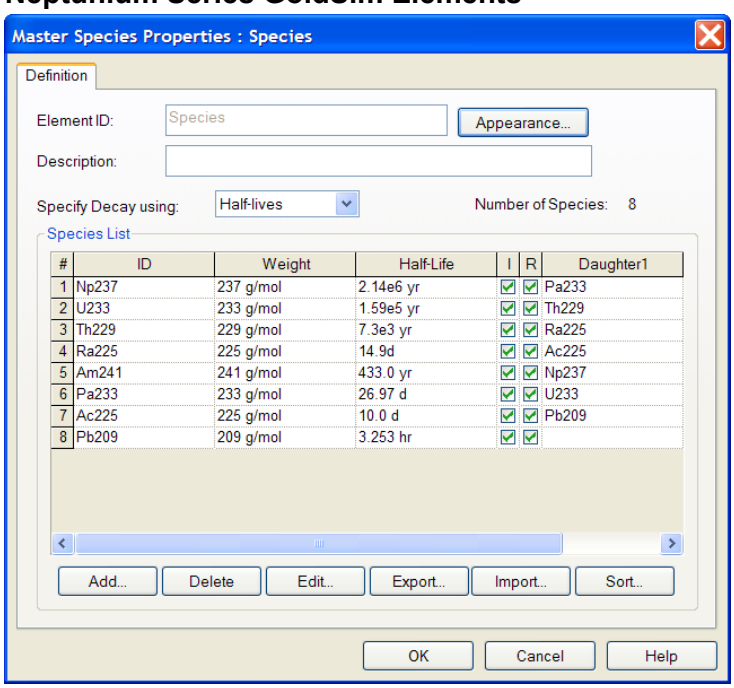




Note that the equation in Expression 2 does not include the ratio of the atomic weight of the daughter to that of the parent. This factor decreases the daughter mass by only 2%. Consequently, it was decided not to rerun the simulations with the corrected equation because the results would not have changed significantly.



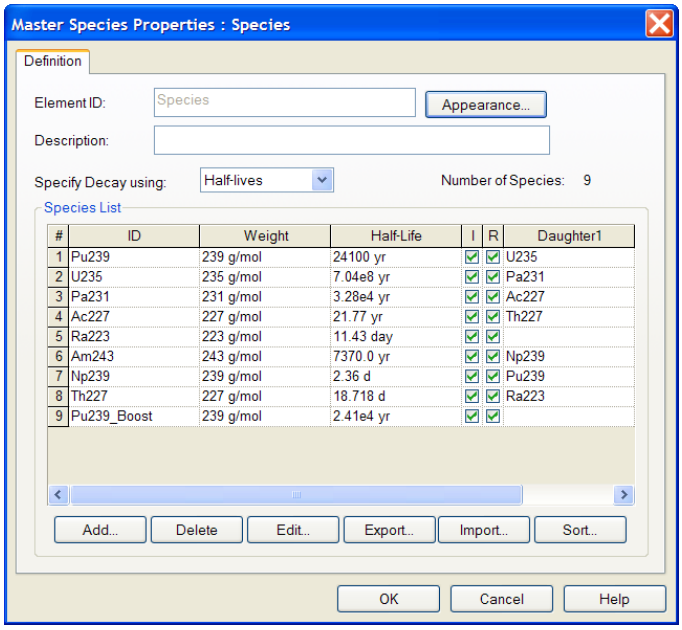
Neptunium Series GoldSim Elements



#	ID	Weight	Half-Life	I	R	Daughter1
1	Np237	237 g/mol	2.14e6 yr	✓	✓	Pa233
2	U233	233 g/mol	1.59e5 yr	✓	✓	Th229
3	Th229	229 g/mol	7.3e3 yr	✓	✓	Ra225
4	Ra225	225 g/mol	14.9d	✓	✓	Ac225
5	Am241	241 g/mol	433.0 yr	✓	✓	Np237
6	Pa233	233 g/mol	26.97 d	✓	✓	U233
7	Ac225	225 g/mol	10.0 d	✓	✓	Pb209
8	Pb209	209 g/mol	3.253 hr	✓	✓	

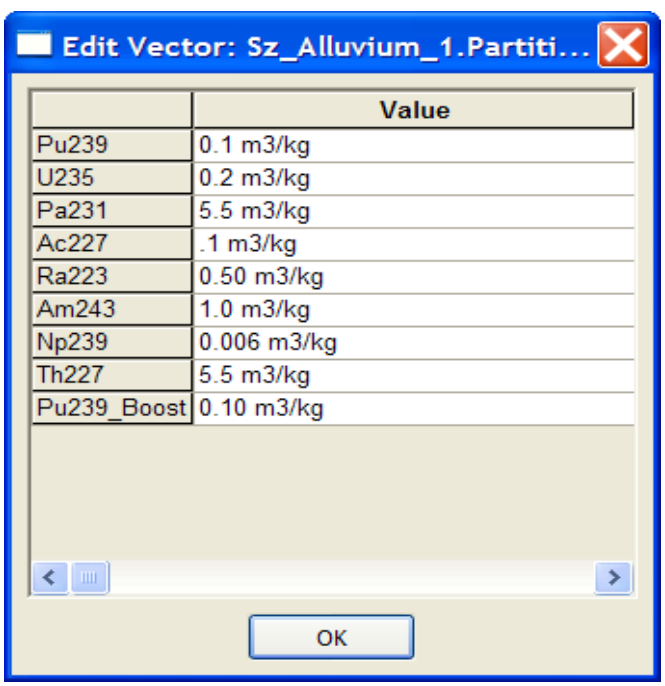
The model for the boosting of Np-237 by the decay of Am-241 was not developed because DOE demonstrated that the total mass of Am-241 added to the saturated zone over the regulatory period was only 0.165 g. In comparison, DOE suggests 1 million grams of Np-237 is added to the saturated zone.

Actinium Series GoldSim Elements



#	ID	Weight	Half-Life	I	R	Daughter1
1	Pu239	239 g/mol	24100 yr	✓	✓	U235
2	U235	235 g/mol	7.04e8 yr	✓	✓	Pa231
3	Pa231	231 g/mol	3.28e4 yr	✓	✓	Ac227
4	Ac227	227 g/mol	21.77 yr	✓	✓	Th227
5	Ra223	223 g/mol	11.43 day	✓	✓	
6	Am243	243 g/mol	7370.0 yr	✓	✓	Np239
7	Np239	239 g/mol	2.36 d	✓	✓	Pu239
8	Th227	227 g/mol	18.718 d	✓	✓	Ra223
9	Pu239_Boost	239 g/mol	2.41e4 yr	✓	✓	

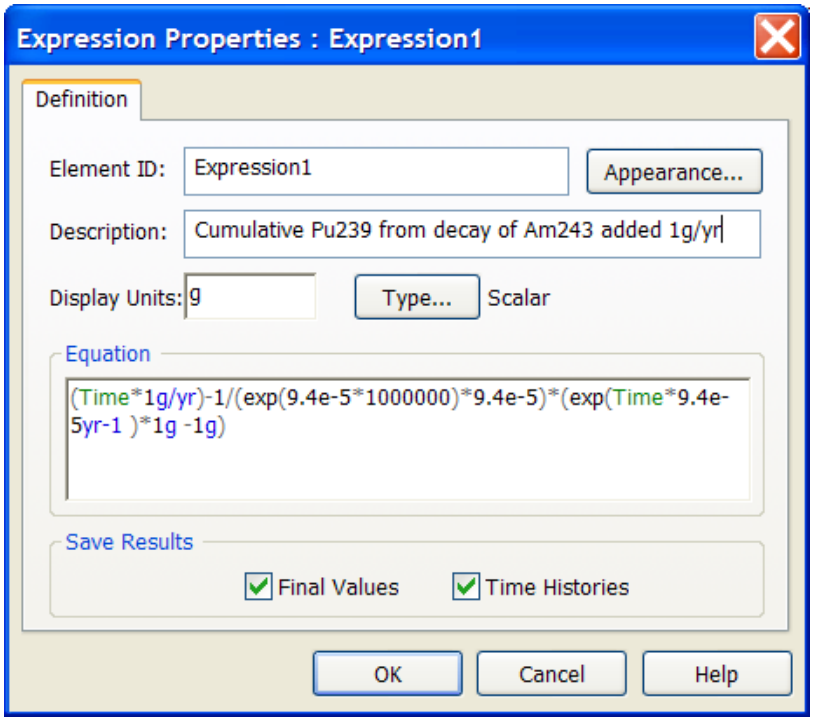
The Solid Properties of Sz_Alluvium_1 and Sz_Alluvium is the same as used in the uranium series model.



	Value
Pu239	0.1 m3/kg
U235	0.2 m3/kg
Pa231	5.5 m3/kg
Ac227	.1 m3/kg
Ra223	0.50 m3/kg
Am243	1.0 m3/kg
Np239	0.006 m3/kg
Th227	5.5 m3/kg
Pu239_Boost	0.10 m3/kg

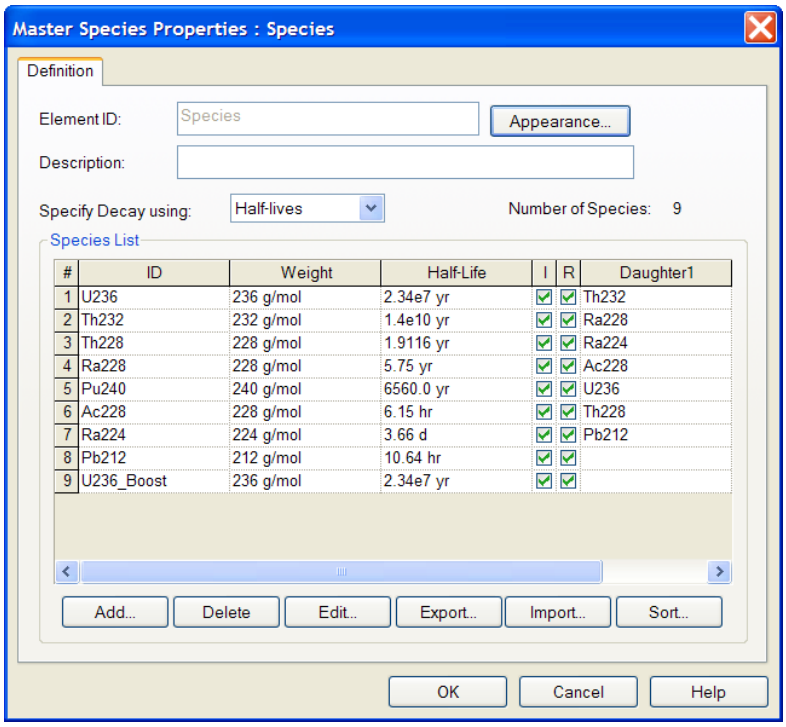
The Partition coefficients for Sz_Alluvium are the same as for Sz_Alluvium_1 except for U-235, which is listed as 0.0087 m^3/kg. Since only the parent and daughter are plotted in the results, the partition coefficient of U-235 is irrelevant.

The Pipe elements for this model are the same as used in the uranium series model.



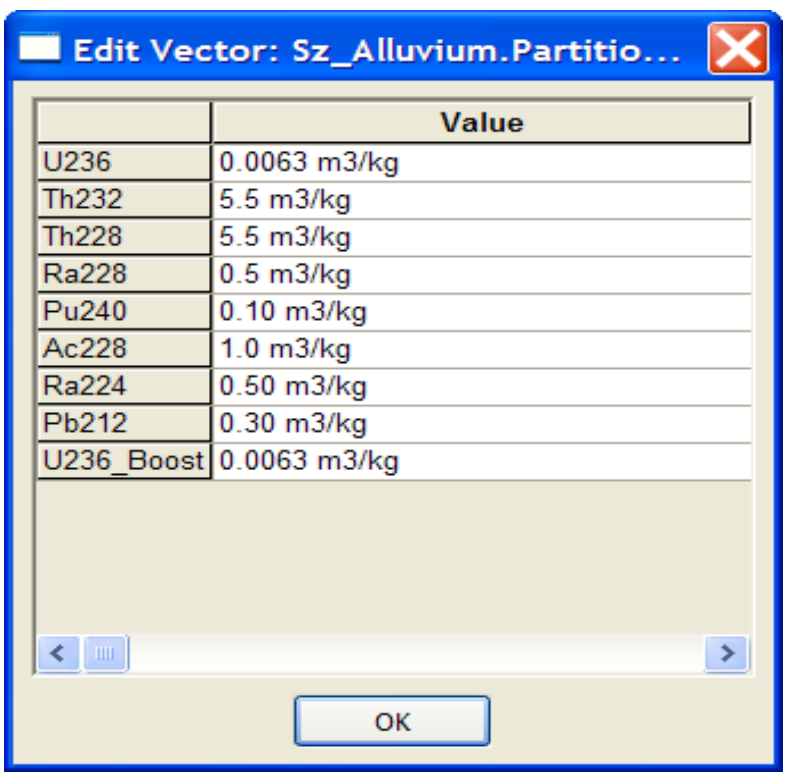
Note that the equation does not include the ratio of the atomic weight of the daughter to that of the parent. This factor decreases the daughter mass by only 2%. Consequently, it was decided not to rerun the simulations with the corrected equation because the results would not have changed significantly.

Thorium Series GoldSim Elements

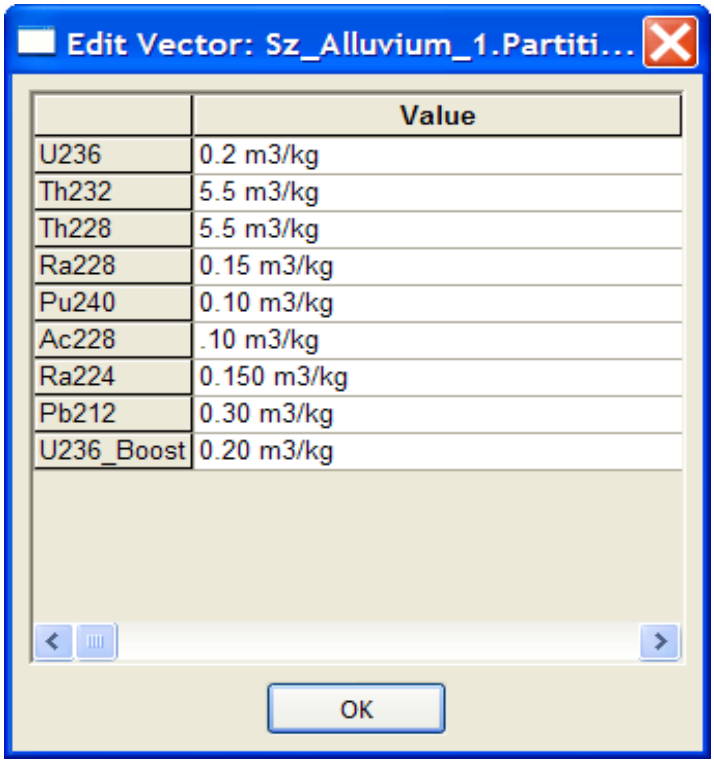


#	ID	Weight	Half-Life	I	R	Daughter1
1	U236	236 g/mol	2.34e7 yr	✓	✓	Th232
2	Th232	232 g/mol	1.4e10 yr	✓	✓	Ra228
3	Th228	228 g/mol	1.9116 yr	✓	✓	Ra224
4	Ra228	228 g/mol	5.75 yr	✓	✓	Ac228
5	Pu240	240 g/mol	6560.0 yr	✓	✓	U236
6	Ac228	228 g/mol	6.15 hr	✓	✓	Th228
7	Ra224	224 g/mol	3.66 d	✓	✓	Pb212
8	Pb212	212 g/mol	10.64 hr	✓	✓	
9	U236_Boost	236 g/mol	2.34e7 yr	✓	✓	

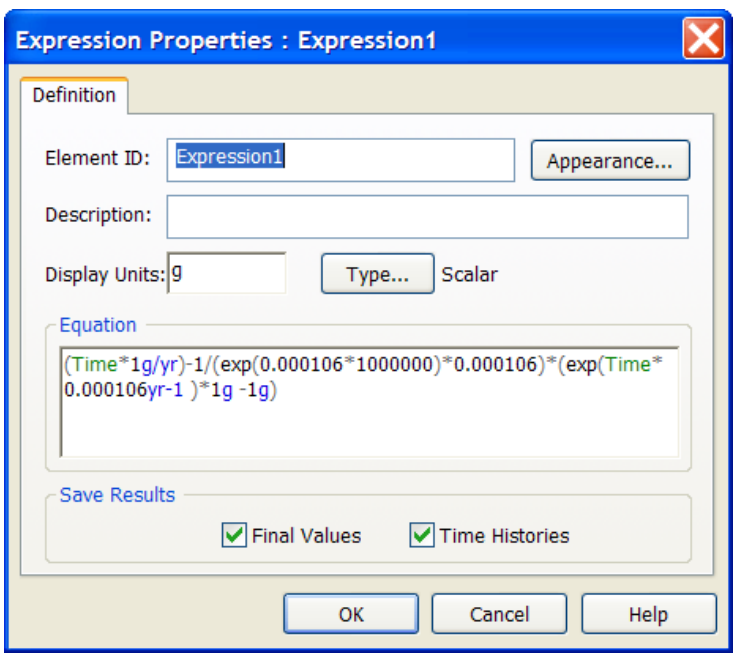
The Solid Properties of Sz_Alluvium_1 and Sz_Alluvium is the same as used in the uranium series model.



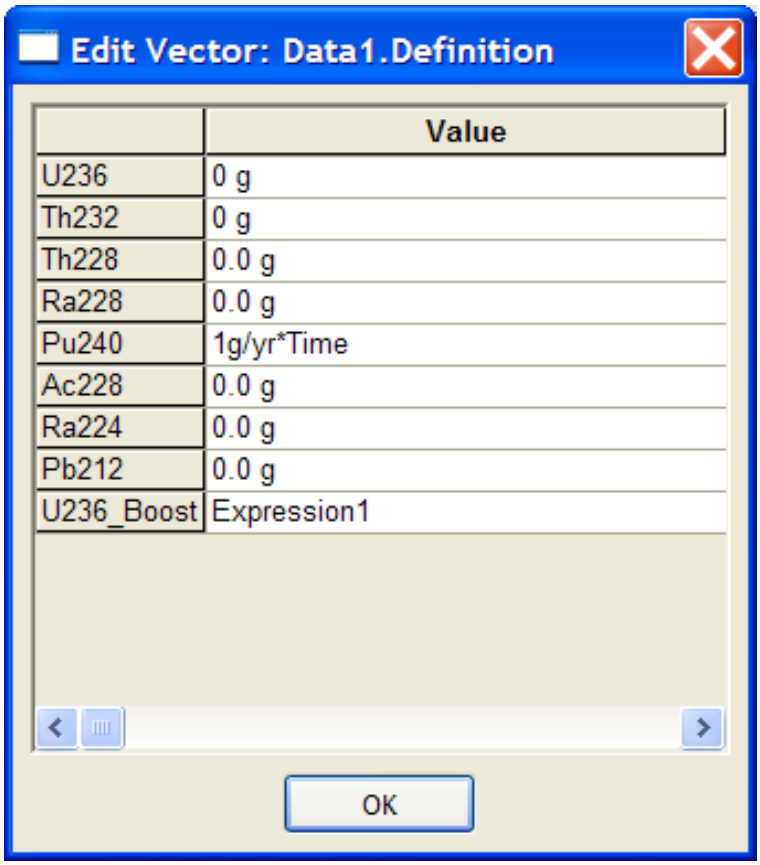
	Value
U236	0.0063 m3/kg
Th232	5.5 m3/kg
Th228	5.5 m3/kg
Ra228	0.5 m3/kg
Pu240	0.10 m3/kg
Ac228	1.0 m3/kg
Ra224	0.50 m3/kg
Pb212	0.30 m3/kg
U236_Boost	0.0063 m3/kg



The Pipe Elements for this model are the same as used in the uranium series model.



Note that the equation does not include the ratio of the atomic weight of the daughter to that of the parent. This factor decreases the daughter mass by only 2%. Consequently, it was decided not to rerun the simulations with the corrected equation because the results would not have changed significantly.



All models are run deterministically. There are 1,000 time steps of 1,000 years each.