Summary of Solubility Calculations
Conversion from moles/L to mg/L

Solubility values are often presented in units of moles/volume, but GoldSim requires that solubility be entered in units of mass/volume. The conversion from moles/volume to mass/volume is simple for non-radioactive elements, but becomes complicated for elements with multiple isotopes that are included in the model. Below is a detailed description of how these calculations were performed for the NTS GCD GoldSim model.

Solubility of a chemical species in water enters the GoldSim model in the definition of the reference fluid. Although GoldSim allows an input for each species (and thus an apparently different solubility for each isotope of an element), it really assigns only one solubility value to each element. Thus, for each element, the model must choose a value for the elemental solubility, if there is more than one isotope in the model. GoldSim uses the first isotope that is encountered in the species list for the elemental solubility. For example, if U-233 were the first uranium isotope in the species list, GoldSim would use the solubility entered for U-233 as the elemental solubility of U. If GoldSim solubilities were expressed in moles/L, there would be no concern from which isotope the elemental solubility is chosen, since that value would be constant across all isotopes.

As mentioned above, GoldSim expects solubility to be in units of **mass** per volume, but the total solubility used in traditional chemistry literature is in units of **moles** per volume. Solubility is dictated by reactions (precipitation/dissolution or adsorption/desorption) that are in terms of moles, not mass, since a mole is an amount of substance (e.g., number of molecules or atoms).

Converting moles to mass is not straightforward when dealing with multiple isotopes. For elements with one isotope defined in GoldSim, the number of moles is multiplied by the atomic weight, in g/mol. For elements with more than one isotope, mass-based ratios of each isotope present must be considered since each isotope has its own atomic weight. As different isotopic ratios change with time, the total **mass** per liter can change, even though the **moles** per liter is held constant. For example, suppose the solubility limit were 1 mmol/L for U. If all U were U-233, the solubility limit would be 233 mg/L, but if all U were U-238, there would be 238 mg/L. These variations result in about a 2% difference in the solubility of U total, which would allow either more or less U in solution. Thus, in order to provide the most accurate solubility values, it is important to determine the isotopic distribution of an element and the resultant atomic weight.

In order to determine the isotope-weighted atomic weight, an mass-based average of all isotope atomic weights should be calculated. For uranium the equation used would be

$$C_{totUsol}(g/L) = C_{totUsol}(mol/L)\sum \frac{m_i}{m_{TotU}} \cdot atomicwt_i \ , \qquad [1]$$

where

m_i	=	mass of isotope *i* in the cell of interest;
$atomicwt_i$	=	atomic weight of isotope *i*;
m_{TotU}	=	total mass of uranium in cell of interest;
$C_{totUsol}(g/L)$	=	solubility of U in units of g/L; and
$C_{totUsol}(mol/L)$	=	solubility of U in units of mol/L.

In GoldSim, this isotope-weighted atomic weight should ideally be calculated for each cell and for each time step, since different isotopic ratios could occur in different cells and at different times. This level of detail is not possible in GoldSim, since only one value of solubility exists at each time step — there can't be different solubility constants in different cells at the same time. Fortunately, for our model the only cell that reaches the solubility limit is the waste cell. Hence we can use the isotopic ratios in the waste cell to determine the isotope-weighted atomic weight.

When Eq. 1 is implemented in GoldSim, a feedback loop is created between that equation and the mass in the waste cell. The equation is used to calculate solubility, which affects the mass of each element allowed to leave the waste cell and thus the mass of each isotope in the waste cell. Feedback loops are created by using a tilde (~) in front of the necessary variables in the equation. For example, in Eq. 1 a tilde would be placed in front of all the mass output values from the waste cell.

Since these calculations include a feedback loop, the model runs much more slowly than it would without the feedback loop. It appears that these calculations do not make much difference (less than 1%) in the final mass in cells for this model. Therefore, it is recommended that estimates of the Pu, Th, and U atomic masses be calculated with an initial deterministic run of the model using the mean input distribution values for the inventory, and averaging the isotopic rations over time. This average atomic mass can be used for subsequent simulations to convert solubility from mol/L to mg/L.