Isotopic Dating Maximum Datable Age

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Abstract

Isotopic dating using long-lived radionuclides is used to estimate ages of rocks pulled from strata. Closed system behavior is assumed unless discordant results are noted. Concordant or discordant results do not guarantee the system is closed or open. The calculated age without a test of openness/closeness of the system has no merit. A systematic method of determining the age of a multilayered rock structure having open system behavior is proposed. When rock atomic loss constants for different dating methods are similar and much greater than the decay constants, the apparent closed system ages will be similar. A discussion is given as to how much movement of radioisotopes and their progeny from one layer to the next is allowed in order for closed system results to be reliable. The maximum datable age is defined.

Introduction

If isotopes used for isotopic dating were fixed in the geological formations, then determining the age would be relatively simple (as is commonly supposed). Movement of the isotopes is an effect that is understood to have taken place in some formations. When results do not agree with the accepted age, or different isotopic dating methods conflict, open system behavior is often assumed to have taken place. It will be shown that it is difficult to exclude open system behavior as having occurred. A test for open system behavior is proposed and the maximum datable age is defined.

The test, based upon uniformitarian-like assumptions, defines the maximum datable age. The doctrine of uniformitarianism states that past geological processes operate in the same manner and at the same average rate as modern geological processes, assuming all catastrophic events are small and of a localized nature. The maximum datable age uses current open system behavior to place a limit on the calculated age based upon current rates of isotope addition/depletion from a geological layer.

Two mechanisms of loss of parent/progeny can be considered when open system behavior is active, namely first order loss and volume diffusion. Commonly set-up differential equations are of the form

First order loss

$$\frac{dc}{dt} = -k(t)c + p \tag{1}$$

Volume diffusion

$$\frac{\partial c}{\partial t} = D(t)\nabla^2 c + p \tag{2}$$

where c is the concentration of parent/progeny, p is the production rate of the parent/progeny, k is a first order rate coefficient for parent/progeny, and D is a diffusion coefficient (Dodson, 1979). Diffusion is the process of movement of molecules from one part of a system to another as

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a consequence of random molecular motion. Diffusion is strongly temperature dependent. First-order loss is the process where molecules may move from one part of a system to another, but not necessarily as a result of purely random molecular motion. For instance, if water infiltrates the cracks in a geological strata and the net flow is downward, the molecular motion would be more accurately described by first-order loss.

It may seem reasonable that below the closure temperature a mineral should behave as a closed system. Closure temperatures can be experimentally determined or can be predicted based upon the temperature-dependence of the volume diffusion process. However, if first-order loss is the principal mode of molecular movement, then closure temperatures do not represent true closure in the rock. They are calculated assuming diffusion is the principal cause of molecular loss.

Tilton considered the mathematics of volume diffusion for various one-dimensional mineral crystals in whole rock geometries. He noted that the volume diffusion could possibly explain certain discordant lead ages (Tilton, 1960). Nicolaysen demonstrated graphically the apparent age of a mineral versus the true age as a function of the parameter D/a^2 for volume diffusion of the progeny out of the mineral crystal (Nicolaysen, 1957). Here D is the diffusion coefficient and "a" is a diffusion size characteristic of the crystal.

The diffusion effect would be expected to be more pronounced for smaller diameter crystals in geological strata. Large 30 cm diameter Alpine biotites have the same parent to daughter (potassium40 to argon-40) ratio as nearby biotites (< 1 mm in diameter). Dodson considered three possible causes for this anomaly, one of which is that transport is dominated by first-order mechanisms (Dodson, 1979). In this paper, we will consider one-dimensional transport by first-order loss, accounting for radioactive decay for a system consisting of layers of arbitrary thickness. We will demonstrate graphically for first-order transport the apparent versus the true age for depletion of the parent/daughter near the earth's surface, much like Nicolaysen demonstrated the same for depletion of the daughter for diffusion.

Applying first-order loss to open system behavior is a natural extension of how it is applied to closed systems. In a closed system, atoms of the parent/progeny can only decrease/increase by radioactive decay. A *nuclide* is a specific nucleus with a given number of neutrons and a given number of protons. Open system behavior in rock strata can cause nuclides used to date the strata to be exchanged between the layers. In an open system, we also have atoms of the parent and progeny either entering or leaving the volume unit. Consider a rock formation on the earth surface consisting of nuclides A and B in a mineral media, where A decays to B a fraction α of the time, and B is a stable nuclide. If transport of species A and B did not occur between layers within the rock strata, then the first-order differential equation relating the rate of loss/gain of A and B atoms in any rock sample would be

$$\frac{dN_A}{dt} = -\lambda_A N_A \tag{3a}$$

$$\frac{dN_B}{dt} = \alpha\lambda_A N_A \tag{3b}$$

and the solution to the closed system equations when solved for the time since formation is

$$t = \frac{t_{1/2}}{\ln 2}\ln\left(1 + \frac{N_B(t) - N_B(0)}{\alpha N_A(t)}\right) \tag{4}$$

where

$t_{1/2}$= half-life of nuclide A

λ_A= decay constant of nuclide A

α= branching fraction—fraction of A that decay to B

$N_A(t)$ = number of atoms of A today

$N_B(t)$ = number of atoms of B today

$N_B(0)$ = number of atoms of B when the rock formed or became closed

t= age of rock

The branching fraction α is necessary for cases such as K/Ar decay, where beta-minus decay occurs 89.3% of the time leading to Ca-40 and electron capture occurs 10.7% of the time leading to Ar-40.

We now address the movement of atoms of nuclide A and B in the rock strata. Consider atoms of one species moving from one layer into an adjacent layer. Let the rock segment have an area A and thickness L as shown in Figure 1. There are initially N atoms in the volume. We allow fluid to infiltrate the rock with a velocity v_w so that the time for the water to exit the rock layer is L/v_w. During this time, a fraction f of the N atoms dissolve into solution and pass with the water out of the volume, so that the rate of atomic removal out of this volume is

$$\frac{fv_w}{L}N$$

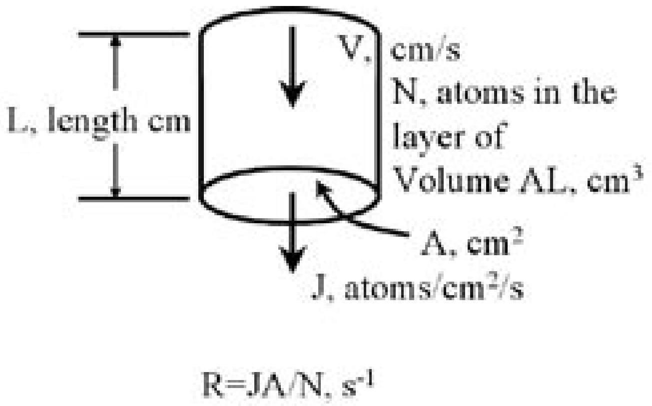

Figure 1. The rock layer atomic loss constant equals the rate of the atoms in volume AL passing through the area A into the adjoining volume divided by the total number of atoms in the layer.

We define the quantity f v_w/L to be the rock layer atomic loss constant (R). It has the same units as the decay constant λ. This is a heuristic explanation of what this rate constants represent and is not meant to be exhaustive. The dynamics driving the process are not considered here, we are only concerned with the kinematics. The rock layer atomic loss constant can also be written as

$$R = \frac{JA}{N} = \frac{J}{\rho L}$$

where J is the atomic current density passing *out* of the volume AL through the surface area A and ρ is the atomic density.

Transport Between Rock Layers and the Atmosphere

We now consider a cylindrical volume of the rock and the air above it as shown in Figure 2. Layer 0 is the atmosphere above the rock that will receive gaseous nuclides from the rock, if the B nuclide is a gas. Layers 1 to p are sequential layers in the rock strata. The differential equations relating the rate of change of nuclides A and B in layer n is

$$\frac{dN_{A,n}}{dt} = -(\lambda_A + R_{A,n,n-1} + R_{A,n,n+1})N_{A,n} + R_{A,n-1,n}N_{A,n-1} + R_{A,n+1,n}N_{A,n+1} \quad (5a)$$

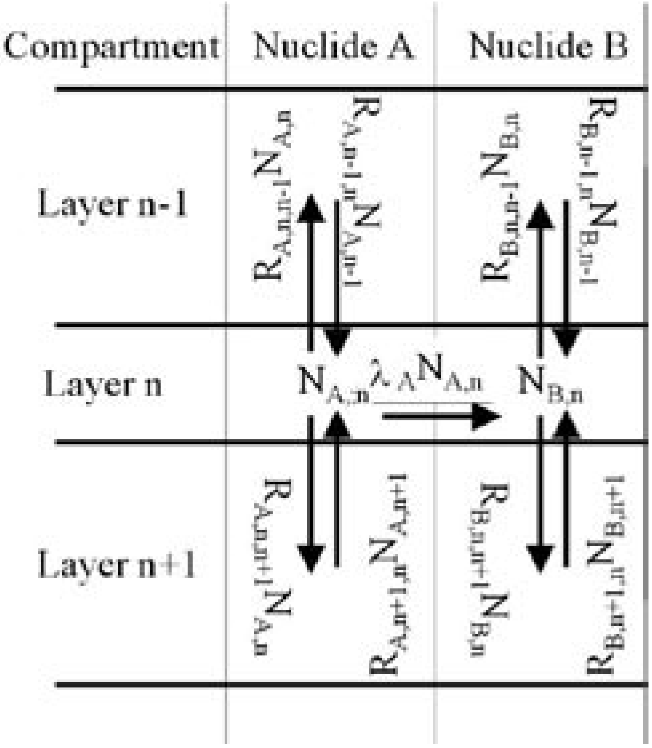

Figure 2. The transport processes in a one-dimensional layered system bounded above by the atmosphere and below by additional layers of rock. Nuclide A can leave layer n by three routes (top, bottom and decay of A) and come into the system by two routes (top and bottom). Nuclide B can leave by two routes (top and bottom) and enter by three routes (top, bottom & decay of A).

$$\frac{dN_{B,n}}{dt} = \alpha\lambda_A N_{A,n} - (R_{B,n,n-1} + R_{B,n,n+1})N_{B,n} + R_{B,n-1,n}N_{B,n-1} + R_{B,n+1,n}N_{B,n+1} \quad (5b)$$

Equation (5a.) represents the rate of change of A atoms in layer n due to radioactive decay and movement of A atoms into/out of the layer. Equation (5b) represents the rate of change of B atoms in layer n due to gain from radioactive decay of A atoms and movement of A atoms into/out of the layer. The notation $R_{A,n,n-1}$ represents the rock layer atomic loss constant of species A moving from layer n to n-1. To express the above differential equations in terms of concentrations, substitute $N_{A,n}(t)=(AL_n)C_{A,n}(t)$, and $N_{B,n}(t)=(AL_n)C_{B,n}(t)$ where L_n is the thickness of layer n and C is the concentration of nuclide A or B.

Some decay chains have several progeny and in that case, equation 5b can have a decay term ($-\lambda_B N_{B,n}$) added and similar equations set up for progeny C, D, … Z. However, if the half lives of the progeny are short in comparison to the parent, and the progeny rock atomic loss constants are much less than their decay constants, then equations 5 is a suitable approximation where B is the stable end product. This approximation would apply to decay chains commonly used for isotopic dating.

The net atomic current density passing through the interface of layers n, n+1, where down is considered positive is

$$J_{A,net,n,n+1} = \frac{1}{A}\left(R_{A,n,n+1} N_{A,n} - R_{A,n+1,n} N_{A,n+1}\right) \tag{6a}$$

$$J_{B,net,n,n+1} = \frac{1}{A}\left(R_{B,n,n+1} N_{B,n} - R_{B,n+1,n} N_{B,n+1}\right) \tag{6b}$$

and with no net flow, the above two equations would give net current densities of zero.

The rock layer atomic loss constants (R) may increase or decrease, since environmental factors will affect movement of nuclides in the rock. For this analysis we will consider the limitations imposed upon radiometric dating assuming R is nearly constant, which may be a reasonable approximation if the average R over one year intervals does not vary significantly with time. Under this approximation, we desire to understand the limitations of the closed system assumptions where the rock atomic loss constant (R) and their difference from layer to layer may significantly exceed the radioactive decay constant. The variation of atomic loss rate from layer to layer may cause accretion and depletion layers.

There are a number of idealistic boundary conditions that are related as follows:

A. Neither A or B are a gas under normal conditions and the system is closed above and below, so $R_{A,0,-1}$, $R_{A,0,1}$, $R_{A,-1,0}$, $R_{A,1,0}$, $R_{B,0,-1}$, $R_{B,0,1}$, $R_{B,-1,0}$, $R_{B,1,0}$, $R_{A,p,p+1}$, $R_{A,p+1,p}$, $R_{B,p,p+1}$, and $R_{B,p+1,p}$ equal zero.

B. A is not a gas under normal conditions and the system is closed above and below, but B is a gas so that $R_{A,0,-1}$, $R_{A,0,1}$, $R_{A,-1,0}$, $R_{A,1,0}$, $R_{B,0,-1}$, $R_{B,-1,0}$, $R_{A,p,p+1}$, $R_{A,p+1,p}$, $R_{B,p,p+1}$, and $R_{B,p+1,p}$ equal zero.

Under these boundary conditions, the layer 0 to p system is closed to external influx/outflux of species A and B so that there is no net loss or gain of atoms by the system except by radioactive decay. This is the same as saying $N_A(t)$ and $N_B(t)$ satisfy equations (3a.) and (3b.) where these are the solution for the entire system. In general, the lower boundary will tend to be open, so we desire conditions that will let us know whether a given sampled strata will be sufficiently closed over the calculated age for the strata to be confident in the age result. Picking boundaries above and below for a one-dimensional system where the whole system can be assumed to be approximately closed to nuclide transport should improve as the length of the layer increases and the average atomic loss rate of both species (R_A, R_B) decreases and there are no large atomic current densities into/out of the upper or lower boundaries. These conditions can be stated quite simply for the upper and lower boundaries that the amount of nuclide A or B atoms that leak into or out of the boundary in time T must be much less than the inventory in layers 0 to p. Stated mathematically,

$$\begin{aligned} A\left|J_{A,net,0,1}\right|T &<< \sum_{n=0}^{p} N_{A,n}(t) \\ A\left|J_{B,net,0,1}\right|T &<< \sum_{n=0}^{p} N_{B,n}(t) \\ A\left|J_{A,net,p,p+1}\right|T &<< \sum_{n=0}^{p} N_{A,n}(t) \\ A\left|J_{B,net,p,p+1}\right|T &<< \sum_{n=0}^{p} N_{B,n}(t) \end{aligned} \tag{7}$$

where from t=0 to T the above conditions must hold for the upper and lower boundaries for the one-dimensional open system results to be reliable.

For a specific layer n the maximum datable age (MDAge) is operationally defined for nuclide A or B as the current inventory in layer n divided by the magnitude of the net transport rate in/out of that layer. The thicker the layer, the greater the inventory of atoms. The primary limiting factor on MDAge will be how many atoms can you detect going in/out of the layer. If the experimental method can only detect $\pm 10^9$ (billion) atoms/year and the inventory is 10^{18} (billion-billion) atoms, then the maximum datable age would be a billion years.

Systematic Method of Determining the Age of a Multilayered Rock Strata with Open System One-Dimensional Transport

Once the $N_{A,n}(0)$, $N_{A,n}(t)$, $N_{B,n}(0)$, $N_{B,n}(t)$, and the rock layer atomic loss constants are either experimentally determined or estimated with their corresponding uncertainties, each layer will have an estimated age that we can calculate from the solution of equation (5a.) and (5b.) given in the Appendix. We call the ages t_n. Each of the results will have a calculated uncertainty so that for p layers there will be p estimates of the age of the multilayered strata given by $t_n \pm \delta t_n$. The weighted estimate of the age of the rock strata is T and is given by

$$T = \frac{\sum_{n=1}^{p}\left[\frac{1}{\delta t_n^{\,2}} t_n\right]}{\sum_{n=1}^{p}\left[\frac{1}{\delta t_n^{\,2}}\right]} \tag{8}$$

$$\delta T = \frac{1}{\sum_{n=1}^{p}\left[\frac{1}{\delta t_n^{\,2}}\right]} \tag{9}$$

Although the estimates $t_n \pm \delta t_n$ may be very uncertain, the fractional uncertainty of the multilayered result would be less than any individual layer result and for many layers would be expected to be much better. We will consider a simple one/two layer system near the earth surface in the next section and calculate the maximum datable age for the layer, the apparent closed system age and the actual open system age when transport is dominated by nuclide A or B depletion. A one/three layer system can be used to an initial attempt to date deep layers.

General Experimental Method to Determine the Rock Layer Atomic Loss Constants

The diffusion equation's diffusion coefficient is relatively easy to experimentally determine, but the first order loss' rock atomic loss constant is more difficult to resolve. We are describing the method because if first order loss dominates, then it is necessary to experimentally determine the constants. To determine rock layer atomic loss constants (R), a core of the rock should be taken and cut into layers as shown in Figure 3. The layers should be separated and smoothed. An artificial rock or natural rock containing the same mineral content, but smaller concentrations of nuclides A and B should be prepared and cut into corresponding layers. Alternating rock and artificial rock layers will form two columns, one estimating the odd/even rock atomic loss constant, and the other estimating the even/odd atomic loss rates. Blanks of the artificial or natural rocks will be kept for background concentration subtraction.

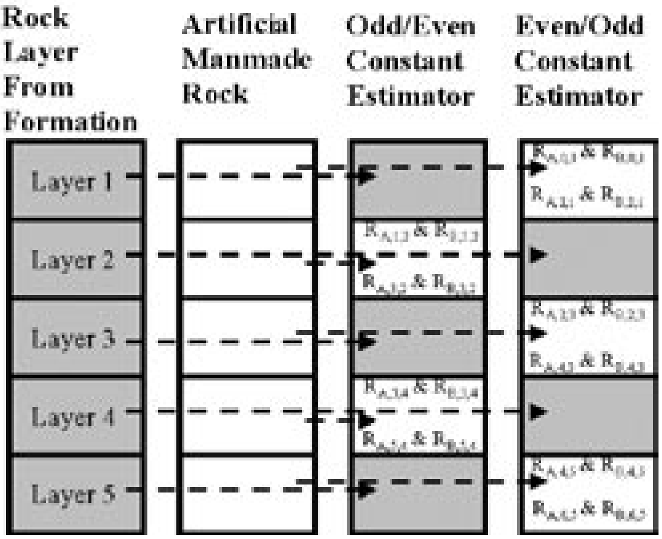

Figure 3. The rock layer atomic loss constant (R) can be measured for a formation where one-dimensional transport is believed to have occurred by separating the layers and stacking native and artificial rock layers under heat, fluid, pressure so nuclide transport can occur for an extended period of time and R can be estimated for the surfaces.

To eliminate uncertainties in natural environmental conditions, the two cores can be taken from the rock in its natural environment and the above process of alternating rock and artificial rock layers repeated in situ. The resulting data would more closely approximate natural conditions without having to know what type of environmental factors may influence the movement of the nuclides.

When the rock and artificial rock layers are interfaced for a period of time (ΔT) under the same or at least similar conditions as the native rock strata, nuclides A and B will move from the rock layers to the artificial rock layers depending on the mobility of the nuclides under those conditions. Over the period ΔT, the amount of movement may be estimated by separating the layers, dissolving the surface of the artificial rocks and analyzing them for trace amounts of nuclides A and B. Dissolving, concentrating and analyzing for nuclides A and B by an integrated light spectrum from the heated sample may be one method of determining the amount of A and B which can be used to calculate the rock atomic loss constants. A similar analysis should be done on blank artificial rocks to see if they produce any spurious counts over a similar counting period for nuclide A and B. If ΔN is the number of atoms that move into the artificial rock layer in a period ΔT and N is the number of atoms in the layer from which the atoms are coming from, then

$$R = \frac{\frac{\Delta N}{\Delta T}}{N} \tag{10}$$

If we treat the N atoms of species A or B in the volume AL as a uniformly distributed fluid that is leaking out the bottom area A (with replenishment from the top) at a rate RN=JA, then the time for all the original atoms in the volume to leak out is 1/R. In that time, an atom at the top has moved a length L, so the average velocity of the particles exiting the bottom of the area A is

$$\begin{aligned} V_{A,n,n+1} &= LR_{A,n,n+1} \\ V_{B,n,n+1} &= LR_{B,n,n+1} \end{aligned} \tag{11}$$

So in a time ΔT, the number of atoms (ΔN) transferred to the artificial layer n+1 from layer n and the average depth to which they penetrate (Δz) is given by

$$\begin{aligned} \Delta N_{A,n,n+1} &= R_{A,n,n+1} N_{A,n} \Delta T \\ \Delta N_{B,n,n+1} &= R_{B,n,n+1} N_{B,n} \Delta T \\ \Delta z_{A,n,n+1} &= LR_{A,n,n+1} N_{A,n} \Delta T \\ \Delta z_{B,n,n+1} &= LR_{B,n,n+1} N_{B,n} \Delta T \end{aligned} \tag{12}$$

The atomic radioactive decay constant λ_A and the rock layer atomic loss constant R are dimensionally the same but represent different physical processes, namely λ_A represents loss/gain due to radioactive decay and R represents loss/gain due to atoms moving from one layer to the next. Without performing experiments to determine the rock atomic loss constants, assuming a closed system is paramount to neglecting the possibility that transport of nuclides in a rock layer that may and most likely will dominate loss/gain of nuclides in that layer. For long-lived radionuclides used for dating rocks, λ_A typically varies between 5 x 10^{-19} to 3 x 10^{-17} s^{-1} and even using experimental periods ΔT=1 year, choosing layers dimensions so the number of atoms is about Avogadro's number (N_0) in the volume AL, and hoping that R is no more than 3 x 10^{-17} s^{-1}, we determine ΔN on the order of 10^{15} atoms and a penetration depths on the order of 100 angstroms (for rock length = 10 cm) into the artificial rock layer.

The original distribution $N_{A,n}$ (0) and $N_{B,n}$ (0) may be inferred by the experimentally determined rate constants and today's distributions $N_{B,n}$ (t) and $N_{A,n}$ (t), assuming this model. The present day distributions are assumed to be due to the original distribution and to accretion/depletion brought about by differing rates of nuclide transport between layers and radioactive decay. If the rate constants R are about the same for each layer and small compared to λ_A, then the present day distributions $N_{B,n}$ (t) and $N_{A,n}$ (t) are simply related to the original $N_{A,n}$ (0) and $N_{B,n}$ (0) by radioactive decay, and the closed system approximation would be satisfactory. If the rate constants vary widely and locations where rate constants are high corresponding to accretion/depletion layers, this would imply the closed system assumption would produce ages that differ significantly from the open system calculated ages. Table I compares how closed system assumptions would calculate differing ages than the open system constant rate model under different atomic loss rate assumptions.

Isochron dating methods assume a closed system. If the data plot on a straight line, then the y-intercept gives information about the initial B nuclide concentration. If the data do not plot on a straight line, the isochron method is invalid. In an open system dating method, calculating the original distribution of A and B is based upon the current distribution $N_{A,n}$ (t) and $N_{B,n}$ (t) and the experimentally determined rock atomic loss constants. This is desirable because although the dynamic processes driving the isotope movement may be known, there could be significant variation in the magnitude of the effect from one position to the next.

Limitation of Closed System Assumptions Near the Earth's Surface

If the only nonzero rock atomic loss constants from layer 1 are $R_{A,1,2}$ and $R_{B,1,2}$, then the solution to the layer 1 differential equations for this open system is

$$\begin{aligned} N_{A,1}(t) &= N_{A,1}(0) e^{-(\lambda_A + R_{A,1,2})t} \\ N_{B,1}(t) &= N_{B,1}(0) e^{-(R_{B,1,2})t} \\ &+ \frac{\alpha \lambda_A N_{A,1}(0)}{(\lambda_A + R_{A,1,2} - R_{B,1,2})} \left[e^{-R_{B,1,2}t} - e^{-(\lambda_A + R_{A,1,2})t} \right] \end{aligned} \tag{13}$$

Scenario A

If $R_{B,1,2} \gg R_{A,1,2}$ and λ_A, then

$$\frac{N_{B,1}(t)}{N_{A,1}(t)} \approx \frac{\alpha\lambda_A}{R_{B,1,2}} \tag{14}$$

We place this result into equation (4) to calculate the apparent age ($t_{\text{closed system apparent}}$) based upon the actual open system behavior.

$$t_{closed\_system\_apparent} = \frac{1}{\lambda_A}\ln\left[1+\frac{\lambda_A}{R_{A,1,2}}-\frac{N_{B,1}(0)}{\alpha N_{A,1}(t)}\right] \tag{15}$$

If we consider the system is actually open below, we have $t_{\text{open system actual}}$ as

$$t_{open\_system\_actual} \approx \frac{1}{\lambda_A}\ln\left[\frac{R_{B,1,2}}{\alpha\lambda_A}\frac{N_{A,1}(t)N_{B,1}(t)}{N_{A,1}(0)^2}\right] \tag{16}$$

So that under the closed system approximation and neglecting the actual open system behavior where the transport is dominated by nuclide B leaving the system, the rock sample appears younger than actuality. The age may even calculate to be a negative value.

Scenario B

If $R_{A,1,2} \gg R_{B,1,2}$ and λ_A, then

$$\frac{N_{B,1}(t)}{N_{A,1}(t)} \approx \left[\frac{N_{B,1}(0)}{N_{A,1}(0)}+\frac{\alpha\lambda_A}{R_{A,1,2}}\right]e^{R_{A,1,2}t}-\frac{\alpha\lambda_A}{R_{A,1,2}} \tag{17}$$

We place this result into equation (4) to calculate the apparent age ($t_{\text{closed system apparent}}$) based upon the actual open system behavior.

$$t_{closed\_system\_apparent} \approx \frac{R_{A,1,2}}{\lambda_A}t+\frac{1}{\lambda_A}\ln\left[\frac{N_{B,1}(0)}{N_{A,1}(0)}+\frac{\alpha\lambda_A}{R_{A,1,2}}\right] \tag{18}$$

Here t is the open system age and the open system age estimate for the layer is

$$t_{open\_system\_actual} \approx \frac{1}{R_{A,1,2}+\lambda_A-R_{B,1,2}} \times \ln\left[\frac{\dfrac{N_{B,1}(t)}{N_{A,1}(t)}+\dfrac{\alpha\lambda_A}{R_{A,1,2}}}{\dfrac{N_{B,1}(0)}{N_{A,1}(0)}+\dfrac{\alpha\lambda_A}{R_{A,1,2}}}\right] \tag{19}$$

So that under the closed system approximation and neglecting the actual open system behavior where the transport is dominated by nuclide A leaving the system, the rock sample appears much older than actuality.

By restricting nuclides A and B transport so that depletion of layer one dominates, the solution of the general differential Equations (5a.) and (5b.) reduces to a solution expressible as the simple solution of equation (13). In general, numerical solutions for real-life systems will need to be made. Equation (13) expresses how transport affects the number of atoms of A or B in layer one when the system is open. Figure 4 displays the apparent closed system ages near the earth's surface where depletion of nuclide A or B is governed by Equations (13) and this result is placed into Equation (4) to estimate the apparent closed system age. This figure demonstrates if the rock atomic loss constants are large in comparison to λ_A, the closed system results will differ significantly from the actual age.

Isotopic Dating Maximum Datable Age

As demonstrated in the previous section, when transport of nuclides A and B is significant, the closed system assumptions will give unreliable ages. Here we discuss the one-dimensional open system dating method and its reliability.

For a rock strata divided into layers of length L, the weighted age is given by Equation (8). The maximum datable age(s) are based on current measured transport rates. When the weighted age is much less than the maximum datable age(s), the system may be dated to an age less than MDAge and the result may be reliable.

Figure 4 illustrates that a multitude of different nuclide transport scenarios could mimic any apparent age. Consider a rock with an apparent age of 1 billion years. The rock could be 1 billion years old if the rock atomic loss constants of the parent and daughter are less than 1 x 10^{-10} yr^{-1}. But it could be 100 million years old if the parent has a rock

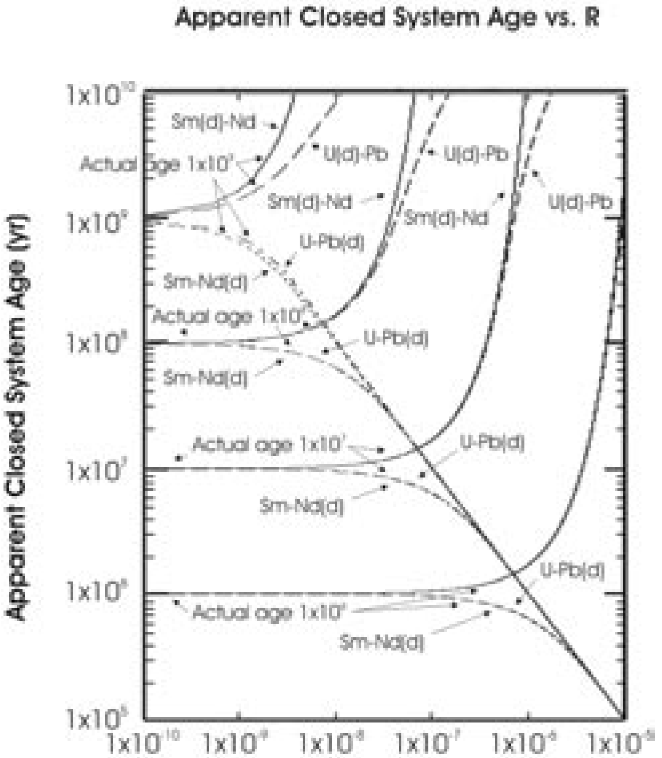

Figure 4. The apparent closed system age near the earth's surface where depletion (d) of either the parent or progeny dominates. Sm-Nd (half life of 106 billion years) and U-235 (half life of 0.704 billion years) curves illustrate that the age distortion increases as the half-life of the dating nuclide increases or as the rock atomic loss constant increases. Here we consider four assumed actual ages 1 million, 10 million 100 million and 1 billion year-old rock strata.

For example, a sample is determined by Sm-Nd dating to have an age of 100 million years based upon closed system assumptions. If only the parent is depleting at the rock atomic loss constant of 1x10^{-7} yr^{-1}, then the actual age is 20 billion years. If only the daughter is depleting at the rock atomic loss constant of 1x10^{-6} yr^{-1}, then the actual age is 1 million years.

atomic loss constant of 2 x 10^{-10} yr^{-1}. However, it could be 10 billion year old if the daughter isotope has a rock atomic loss constant of 1 x 10^{-10} yr^{-1}. The actual age is strongly dependent on the transport of the parent or daughter isotopes used for the dating method.

Figure 5. To use this chart, take the experimentally determined value of the transport rate of isotope A or B in atoms-yr^{-1} and divide each by the Current layer Inventory (CLI) atoms of A or B, respectively. Determine the Maximum Datable Age for each isotope A and B and use the lesser MDAge for determining whether an individual layers result are reliable, or whether the scope of the investigation needs to be expanded.

Figure 5 illustrates effect of transport on the maximum datable age for layers that are not necessarily near the earth's surface. If the rate of gain/loss of nuclide A or B is great enough from a given layer, any conclusion about the age of that layer alone without additional measured transport results from adjacent layers would be unreliable. The maximum datable age for isotope A is defined as

$$MDAge - A = \frac{N_{A,n}}{\left| -(R_{A,n,n-1} + R_{A,n,n+1})N_{A,n} + R_{A,n-1,n}N_{A,n-1} + R_{A,n+1,n}N_{A,n+1} \right|} \tag{20}$$

but a more practical estimator for those wanting to know whether the experimental method will be sufficient to achieve the desired age range is

$$MDAge - A = \frac{N_{A,n}}{2\delta N_A} \cdot \Delta T \tag{21}$$

where δN_A is the minimum detectable atoms that can be detected as having crossed any boundary. There are a few different scenarios that can be explored for MDAge. One is where nuclides transport is dominated by nuclides leaving layer n. In that case, the MDAge is on the order of $(R_{A,n,n+1} + R_{A,n,n-1})^{-1}$ and where nuclide transport is dominated by nuclides entering from the layer above or below (with $N_{A,n} \approx N_{A,n-1} \approx N_{A,n+1}$) the MDAge is $(R_{A,n-1,n} + R_{A,n+1,n})^{-1}$. When nuclides are entering and/or leaving layer n, the MDAge may be large since the denominator in the MDAge definition may be close to zero.

A simple MDAge equation based on water current density J_w (g/cm^2-yr) flowing through the rock of length L (cm) near the earth's surface is

$$MDAge - A = \frac{100\rho_A L}{J_W S_{A-W}} \tag{22}$$

Where ρ_A is the density of A (g/cm^3) and $S_{A\text{-}W}$ is the solubility. The solubility is a unitless number equal to the number of grams of solute of A that can dissolve in 100 cm^3 of water. Even for insoluble compounds of A or B dating isotopes and low current densities, it is difficult for MDAge to achieve a billion year time frame without thick layers.

Solute transport, unlike diffusion transport, has no closure temperature. Below the diffusion closure temperature, solute transport will be the principal cause of dating isotope loss. Dodson (1979) considered first-order loss but assumed the first-order rate coefficient had the same temperature dependence as diffusion. Jenkin et al. (1995) considered Rb-Sr dating systems for bi-mineralic rock. They noted that fluids within the rock can affect closure. No discussion was made as to how much fluid infiltration was allowed in order to have confidence the system was closed and for diffusion loss to dominate. Solute transport is likely to be the primary limitation for isotopic dating of any geological formation on the earth's surface as displayed in Figure 8.

Miller et al. (1991) noted that fluid movement through the rock below the conventional closure temperature might render the system open. The history of the formation could render a given rock sample susceptible to one or more fluids. In particular, the Dalradian rocks of Connemara, Western Ireland had in filled small cracks indicating significant fluid flow after the rock had passed through the closure temperature. The variable nature of the age estimates for different samples indicates the fluid flow may not have been pervasive, but channeled.

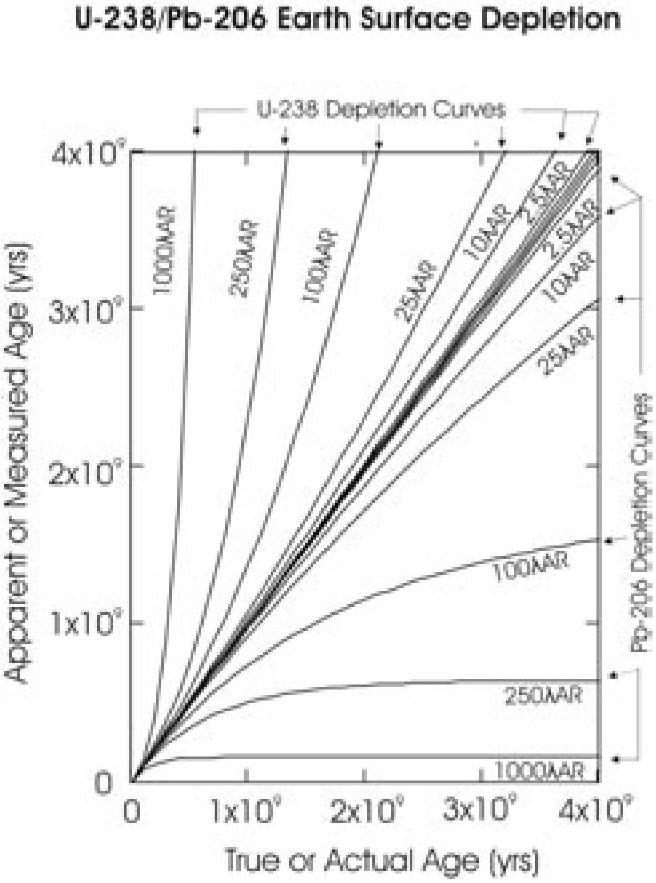

Figure 6. Measured age versus the actual age near the earth's surface where depletion of the parent or daughter dominates. The curves above and below the diagonal represent depletion of U-238 and Pb-206, respectively. The value λAR is selected as the rock atomic loss constant equal to the decay constant of Sm-147 (half life = 106 billion years). Just a little transport of dating isotopes can distort the actual age. Plots for other dating isotopes look similar to this.

Figure 6 illustrates the problem of isotopic depletion of the parent or daughter and the effect on the apparent age versus the actual age. We choose the rock atomic loss constants of the parent or progeny to be multiples of $\lambda AR \equiv 6.5 \times 10^{-12}\ yr^{-1}$. If both the parent and progeny are being lost, then it is best to go back to equation (13) and solve it from there.

If there is a dynamic process that causes the rock atomic loss constants for different dating nuclides to be similar and larger than the decay constants, then the apparent ages based upon closed system assumptions may be similar. Elementally U-235 and U-238 dating would be expected

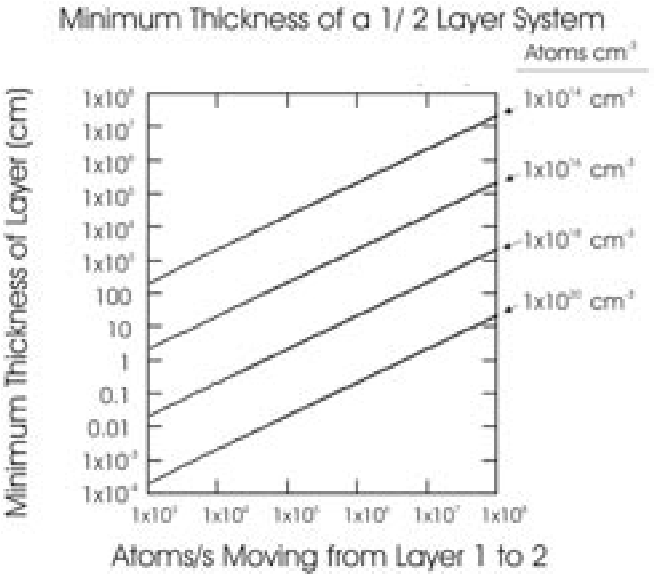

Figure 7. Estimate of the minimum thickness of a one/two layer system near the earth's surface based on the atoms/sec passing from layer 1 to 2 of the parent or daughter. Area of the layer is 10,000 cm^2 and the critical rock atomic loss rate is taken as the radioactive decay rate of U238. A concentration of 1x10^{20} atoms/cm^3 corresponds roughly to a 1% by mass concentration of the parent (U238) or daughter. As the nuclide concentration and area of the layer decrease, the minimum thickness of the layer increases.

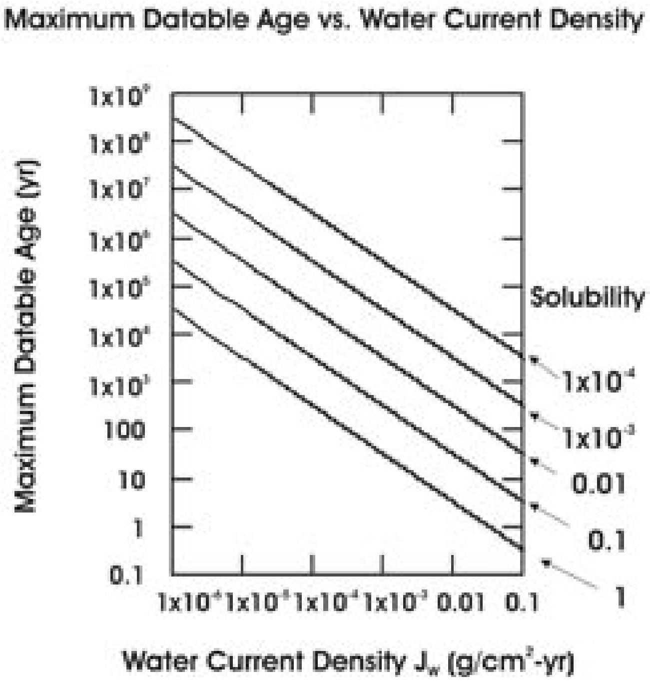

Figure 8. The maximum datable age for a rock sample is strongly dependent on the solubility of the dating isotopes and the water flow through the rock. The graph is for a 100 cm long rock core where the dating isotopes are insoluble (10^{-4} g per 100 ml of water) to soluble (1 g per 100 ml of water). Concentration is 1 PPM and density of rock is 3 g/cm^3. A combination of high concentrations of dating isotopes, long rock cores, low water flow rates, and insoluble dating isotopes are required to achieve maximum datable ages of a billion years. For comparison with the water flow through rock, rainfall in arid regions may be 10 g/cm^2-yr. A small fraction of this rain penetrating the rock may render the system open.

to result in similar rock atomic loss constants. In this case, similar age results could either mean the system is closed and the age is the actual, or more likely the system is open and the age is apparent. This is an explanation why different isotopic dating techniques may often give similar results.

Figure 7 shows the minimum thickness of a one/two layer system as a function of the rate that atoms are passing from layer 1 to 2 (K_a) by selecting the critical rock atomic loss constant ($R_{A,1,2}$) and calculating

$$Minimum\_thickness \approx \frac{K_A}{R_{A,1,2}\,\rho_{atomic}\,A} \quad (23)$$

There are a number of conditions that could cause this open system dating method to produce unreliable results, although current conditions may appear to produce reliable results. Some of these conditions are as follows:

1. The rock atomic loss constants have not been constant for the time 0 to T. If the values varied significantly in the past from their current values, this could significantly increase or decrease the calculated age. Unusual environmental conditions over a short period could result in transport of nuclide A and/or B that could mimic ages either less than or greater than the actual time.
2. If the system does not behave as one dimensional, but significant transport occurs perpendicular to the assumed axis of transport, then nuclides A and B can enter/leave in a way not accounted for by one-dimensional transport. Three-dimensional transport may be required for these types of rock strata. This paper does not consider three-dimensional transport.

3. When erosional processes have taken away layers above or below the present day surface layer. Thus, nuclides could have leached into/out of the present layers from a layer that no longer exists and that we cannot reasonably account for.

Just as the ages calculated for rocks or strata assuming closed system behavior may not represent the actual time since formation, so the calculated age assuming one-dimensional transport may be more reliable but the age may still differ from the actual time since formation.

Summary

The whole-rock isochron method based upon a cogenetic suite of rocks will likely produce meaningless results if isotopic transport rates vary significantly with horizontal location. The isochron diagram generated for a cogenetic suite where the rock sample results produce concordant results could be the result of random chance coincidence driven by differing isotopic transport at different locations if the number of sampled rocks is small enough. Isotopic transport needs to be accounted for to have confidence in any calculated age.

It is operationally necessary for the closed system or open system results to be less than the maximum datable age. If it can be reasonably argued that isotopic transport has not been significantly greater than the current transport rate, then this could be a sufficient condition to have confidence in the calculated open system age. To verify that isotopic transport has not been significantly greater in the past, subjecting samples to laboratory conditions similar to those believed to have existed in the past, could verify transport would not have significantly impacted the maximum datable age restriction on the calculated open system age.

When various types of igneous/metamorphic strata are determined to typically have high isotopic transport rates, this may require the use of thick layers. The thickness would be the prime limiting factor for thin igneous layers. The isotopic transport into/out of the layer by sedimentary layers above/below this would be a confounding factor that could limit the maximum datable age due to isotopic transport into/out of the layer based upon a source of isotopes that cannot be dated.

Near the earth's current surface or near interfaces between geological strata of different transport properties, closed system approximations may over/under estimate the actual age depending on the movement of the isotopes between layers over the ages. The age estimated using the open system one-dimensional transport model could result in a more reliable age estimate if the number of layers and their total length is great enough that insignificant amounts of the dating nuclides leak out of the upper and lower boundaries during the estimated age. A surprising result is that different isotopic dating methods can produce similar apparent closed system ages if the isotopic transport rates of the dating nuclides are similar. **However, for cases where both mineral isochrons and whole rock isochrons give the same erroneous age, this paper has not proven that a fluid transport explanation is sufficient.** When rock atomic loss constants of different isotopic dating methods are the same and much greater than the decay constant, the calculated "erroneous" ages will be the same for the different isotopic dating methods. However, the values of the atomic loss constants are not readily available in the relevant papers of the literature.

Diffusion is an insignificant mode of dating nuclide loss. Water flow through rock at unmeasurably low-levels can cause different dating methods to give coincidentally the same age. Movement of dating nuclides in rock fundamentally limits the isotopic dating method.

Acknowledgements

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Appendix on Mathematical Considerations for a One Two Layer System

We now consider a rock stratum near the earth's surface and consider the problem of determining its age with an open system kinematic back calculation by experimentally determining the rock atomic loss constants for layer 1 and upper part of layer 2. This one/two layer approximation may be sufficient for some strata. For simplicity, we number the steps in the process.

Step 1: A representative composite sample of the strata is taken from the strata. The following data is experimentally determined:

ρ_s = density of strata, g/cm^3
PPM$_A$ = parts per million of nuclide A in the composite of strata
PPM$_B$ = parts per million of nuclide B in the composite of strata

Based upon this information, we can calculate the "first approximation" closed system age, assuming all the atoms of nuclide B are radiogenic from the atoms of nuclide A in layer 1. That is, $N_{B,1}(0) = 0$ and $N_{A,1}(0) = N_{A,1}(t) + N_{B,1}(t)/\alpha$ where t = today. This may not be the case, since A and B can leak into or out of layer 1.

Step 2: We need to estimate the thickness L and area A of layer 1, based upon the understanding that the closed system results will become increasingly unreliable if $R_{A,1,2}$ or $R_{B,1,2}$ are significantly greater than λ_A and the minimum detectable number of atoms (δN_A or δN_B) that the experimental method can determine in the artificial rock as limiting factors. The number of atoms crossing the boundary must be greater than the minimum detectable atoms (δN) in ΔT. The limiting condition for the A and B nuclides for layer 1 can be written as

$$\frac{\delta N_A}{\Delta T} \le R_{A,1,2} N^0_{A,1}(t)$$
$$\frac{\delta N_B}{\Delta T} \le R_{B,1,2} N^0_{B,1}(t) \qquad (24)$$

where we seek to determine the minimum number of atoms of A or B that must be in the layer, which will be used to estimate the required volume AL of the strata. Now the rock atomic loss constants can produce unreliable closed system results if they are on the order of λ_A or greater and the zero order approximation of $N_{A,1}(t)$ and $N_{B,1}(t)$ is given by step 1 derived parameters in the following relations:

$$N^0_{A,1}(t) = \frac{N_0 \rho_s AL \dfrac{PPM_A}{1x10^6}}{W_{M-A}}$$
$$N^0_{B,1}(t) = \frac{N_0 \rho_s AL \dfrac{PPM_B}{1x10^6}}{W_{M-B}} \qquad (25)$$

Solving for the volume of rock that must be sampled, it is the greater volume of the following two conditions:

$$(LA)_A = \frac{\dfrac{\delta N_A}{\Delta T} W_{M-A}}{\lambda_A N_0 \rho_s \dfrac{PPM_A}{1x10^6}}$$
$$(LA)_B = \frac{\dfrac{\delta N_B}{\Delta T} W_{M-B}}{\lambda_A N_0 \rho_s \dfrac{PPM_B}{1x10^6}} \qquad (26)$$

The greater the area (A) of the rock/artificial rock interface, the greater the number of atoms that will be available to pass over the interface. If we take the volume to be a cube of height L and area $A=L^2$, then we can solve for the height of the volume element.

Step 3: We build three artificial rocks of dimensions L x L x L. Chemically they are the same as the rock strata, except they do not have the two isotopic dating nuclides A or B, but could be doped with chemically similar nuclides A' and B' in hope that there will be no preference for the A and B nuclides to move either to the artificial rock or to the natural rock. The atomic concentrations in the artificial rock are adjusted so that $PPM_{A'}/W_{A'} \approx PPM_A/W_A$ and $PPM_{B'}/W_{B'} \approx PPM_{B}./W_B$ where W are the molar weights. This is done to assure that it is unlikely that the lack of nuclides of A or B in the artificial rock strata will not cause accelerated movement across the interface. For core 1 of the sampling strata, one artificial rock will fit between layer 1 and 3, and for core 2 we have an artificial rock at layer 1 and natural rock beneath it. The last artificial rock strata will remain in the laboratory as a blank to have whatever tests performed on it that the in situ artificial rock layers received, to see if any spurious counts may be determined on the blank, implying a small trace concentration of A or B and for calculating the minimum detectable atoms in the artificial rock surface. A thin A & B isotope permeable membrane may be used between the layers to aid in the separation of the layers.

Step 4: The surface of the *in situ* artificial rock layers are separated from the natural rock and the three artificial rock layers are tested to determine the amount of A and B nuclides that leaked in time ΔT, and then $R_{A,1,2}$, $R_{B,1,2}$, $R_{A,2,1}$, and $R_{B,2,1}$ are calculated. Also, layer 1 and 2 of the natural rock is analyzed to determine $N_{A,1}(t)$, $N_{B,1}(t)$, $N_{A,2}(t)$ and $N_{B,2}(t)$. Then $J_{A,1,2,net}$ and $J_{B,1,2,net}$ are calculated. The maximum datable age (MDAge) is

$$MDAge - A \equiv \frac{N_{A,1}(t)}{J_{A,1,2,net} A}$$
$$MDAge - B \equiv \frac{N_{B,1}(t)}{J_{B,1,2,net} A} \qquad (27)$$

If the time calculated with the closed system assumption is significantly greater than this, the closed system result is called unreliable.

Step 5: If the system appears quite open and depletion is occurring for this layer, and if the following conditions hold:

$$R_{A,1,2}N_{A,1}(t) > R_{A,2,1}N_{A,2}(t)$$
$$R_{B,1,2}N_{B,1}(t) > R_{B,2,1}N_{B,2}(t) \quad (28)$$

then it may be possible to date the layer using equations (13). If the assumption $N_{B,1}(0)=0$ is valid, then the open system time is calculated to be

$$T_{open\_system\_depl\_dominates\_layer1} \approx$$
$$\frac{1}{\lambda_A + R_{A,1,2} - R_{B,1,2}} \ln\left[1 + \frac{N_{B,1}(t)}{N_{A,1}(t)}\left(1 + \frac{R_{A,1,2} - R_{B,1,2}}{\lambda_A}\right)\right] \quad (29)$$

If this calculated open system age is significantly greater than MDAge, the result is termed unreliable and the strata cannot be dated with just this one layer. If the result is less than MDAge, then the result may be reliable if it is much less than MDAge.

Step 6: When the closed system and open system results are determined to be unreliable because nuclide A and B are leaking out or into the strata at a high rate, then either select additional layers or do an additional experiment where the new length of the layer L is selected for a one/two layer system in anticipation of what the maximum datable age may be for that height of a layer. Here we have used a one/two layer system near the earth's surface with two cores of natural/artificial and artificial/natural. A one/three layer system could be used to determine the maximum datable age deeper in the earth with two cores of natural/artificial/natural and artificial/natural/artificial layers at the same depth.

Appendix on General Solution to the First Order Loss Equation

A general solution of equations (5a.) and (5b.) can be written as:

$$N_{A,n}(t) = N_{A,n}(0) + \sum_{i=1}^{\infty} M_{A,n,i}t^i \quad (30)$$

$$N_{B,n}(t) = N_{B,n}(0) + \sum_{i=1}^{\infty} M_{B,n,i}t^i \quad (31)$$

where $M_{A,n,i}$, and $M_{B,n,i}$ are coefficients of the Maclaurin series expansion given by the following recurrence relations:

$$M_{A,n,i+1} = \frac{1}{i+1}\Big[-\left(\lambda_A + R_{A,n,n-1} + R_{A,n,n+1}\right)M_{A,n,i}$$
$$+ R_{A,n-1,n}M_{A,n-1,i} + R_{A,n+1,n}M_{A,n+1,i}\Big] \quad (32)$$

$$M_{B,n,i+1} = \frac{1}{i+1}\Big[\alpha\lambda_A M_{A,n,i} - \left(R_{B,n,n-1} + R_{B,n,n+1}\right)$$
$$\times \; M_{B,n,i} + R_{B,n-1,n}M_{B,n-1,i} + R_{B,n+1,n}M_{B,n+1,i}\Big] \quad (33)$$

where $M_{A,n,0} = N_{A,n}(0)$ and $M_{B,n,0} = N_{B,n}(0)$. Using the recurrence relationship of equations (32) and (33), and the following definitions:

$$\begin{aligned}
\varepsilon_{n,-1} &\equiv R_{A,n-1,n}\\
\varepsilon_{n,0} &\equiv -\left(\lambda_A + R_{A,n,n-1} + R_{A,n,n+1}\right)\\
\varepsilon_{n,1} &\equiv R_{A,n+1,n}\\
\delta_{n,-1} &\equiv R_{B,n-1,n}\\
\delta_{n,0} &\equiv -\left(R_{B,n,n-1} + R_{B,n,n+1}\right)\\
\delta_{n,1} &\equiv R_{B,n+1,n}
\end{aligned} \quad (34)$$

we can then write the general term of the Maclaurin series expansion of equations (30) and (31) for the open system multilayered solution as:

$$M_{A,n,k} = \frac{1}{k!}\sum_{i_1=-1}^{1}\sum_{i_2=-1}^{1}\cdots\sum_{i_k=-1}^{1}$$
$$\times \; \left[\varepsilon_{n,i_1}\varepsilon_{n+i_1,i_2}\cdots\varepsilon_{n+i_1+\cdots+i_{k-1},i_k}N_{A,n+i_1+i_2+\cdots+i_k}(0)\right] \quad (35)$$

$$M_{B,n,k} = \frac{1}{k!}\sum_{i_1=-1}^{1}\sum_{i_2=-1}^{1}\cdots\sum_{i_{k-1}=-1}^{1}\left\{\begin{array}{l}\alpha\lambda_A\left[\begin{array}{l}\varepsilon_{n,i_1}\varepsilon_{n+i_1,i_2}\cdots\varepsilon_{n+i_1+\cdots+i_{k-2},i_{k-1}} \\ +\sum_{p=1}^{k-1}\delta_{n,i_1}\delta_{n+i_1,i_2}\cdots\delta_{n+i_1+\cdots+i_{p-1},i_p}\varepsilon_{n+i_1+\cdots+i_p,i_{p+1}}\cdots\varepsilon_{n+i_1+\cdots+i_{k-2},i_{k-1}}\end{array}\right]N_{A,n+i_1+\cdots+i_{k-1}}(0) \\ +\sum_{i_k=-1}^{1}\delta_{n,i_1}\delta_{n+i_1,i_2}\cdots\delta_{n+i_1+\cdots+i_{p-1},i_p}N_{B,n+i_1+\cdots+i_k}(0)\end{array}\right\} \tag{36}$$

It is readily verifiable that the above solution reduces to simple closed system exponential if the rock layer atomic loss rate are set equal to zero, then the solution can be manipulated and expressed as the solution of equation (4).

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