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SOME CORRECTION FACTORS IN RADIOACTIVITY METHODS OF GEOLOGIC TIME MEASUREMENT

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ABSTRACT

The equations employed for the measurement of geologic time are considered for a general radioactive process. Modified expressions are derived to take account of the continued escape, or sudden removal of the parent or daughter elements. The case of the Pb^{207}/Pb^{206} method is also considered.

1. INTRODUCTION

It is well known that methods based on radioactive decay are applicable to problems of geochronology whenever element-separating processes are involved. In particular, when such processes are capable of altering the relative concentration of parent and daughter elements in the phases into which the larger system is differentiated, the time interval between the present and the element-separating event can be computed from a knowledge of the appropriate decay constants and any one of the following sets of data[1]:

(*a*) the initial and present concentrations of the parent;

(*b*) the initial and final concentration of a daughter and the present concentration of the parent or an "elder" daughter when radioactive equilibrium prevails.

FUNDAMENTAL EQUATIONS

Considering a parent element P_0 decaying through P_1, $P_2 \ldots$, P_n in a radioactive chain with decay constants $\lambda_0, \lambda_1, \ldots \lambda_n$ to form a stable daughter S, we have the equations,

$$[D + \lambda_0] P_0 = 0$$
$$[D + \lambda_1] P_1 = \lambda_0 P_0$$
$$\cdots\cdots\cdots\cdots$$
$$[D + \lambda_n] P_n = \lambda_{n-1} P_{n-1}$$

DS $\lambda_n P_n$

where D represents (d/dt).

Now, Bateman's solution[2] can be readily obtained by the Laplace transform method: denoting the transforms by barred symbols, — thus $\bar{P}_r \quad \bar{P}_r(p)$

$$[p + \lambda_0] \bar{P}_0 \quad P_0(i)$$
$$\cdots\cdots\cdots\cdots\cdots\cdots$$
$$[p + \lambda_n] \bar{P}_n = \lambda_{n-1} \bar{P}_{n-1} + P_{n-1}(i)$$

where $P_r(i)$ denotes the initial $(t = 0)$ concentration of P_r,
so that,

$$\bar{P}_n = \frac{\lambda_0 \lambda_1 \ldots \lambda_{n-1}}{(p + \lambda_0)\ldots(p + \lambda_n)} \cdot P_0(i) + \frac{\lambda_1 \ldots \lambda_{n-1}}{(p + \lambda_1)\ldots(p + \lambda_n)} \cdot P_1(i)$$
$$+ \ldots + \frac{1}{p + \lambda_n} \cdot P_{n-1}(i)$$

From this the solution follows:—

$$P_n = (\lambda_0 \lambda_1 \ldots \lambda_{n-1}) P_0(i) \sum_0^n c_{0r} \cdot e^{-\lambda_r t}$$
$$+ (\lambda_1 \ldots \lambda_{n-1}) P_1(i) \sum_1^n c_{1r} \cdot e^{-\lambda_r t}$$
$$\cdots\cdots\cdots\cdots\cdots$$
$$+ P_{n-1}(i) \cdot e^{-\lambda_n t}$$

where c_{0r} for instance, is obtained from the corresponding denominator by substituting $p = -\lambda_r$ in all terms except $(p + \lambda_r)$ which is suppressed.

(1) Considering the special case

$$P_r(i) = 0 \text{ for } r = 1 \ldots n,$$

the expression reduces to the first summation on the R.H.S.

Again, in all the natural radioactive series,

$$\lambda_0 \ll \lambda_r \text{ for } r = 1 \ldots n$$

In this case, only the first term of the summation is significant, thus

$$P_n[t] = P_0(i) \frac{\lambda_0 \ldots \lambda_{n-1}}{(\lambda_1 - \lambda_0)\ldots(\lambda_n - \lambda_0)} \cdot e^{-\lambda_n t}$$

$$\triangleq P_0(i)\frac{\lambda_0}{\lambda_n}\cdot e^{-\lambda_0 t}$$

while $P_0(t) = P_0(i)\cdot e^{-\lambda_0 t}$, so that $\lambda_0 P_0(t) = \lambda_n P_n(t)$.

This is the case of radioactive equilibrium, and in all cases except the post-Pleistocene formations, this condition is fulfilled for the uranium, Actino-uranium, and Thorium series.

Further, in this case

$$\frac{ds}{dt} = \lambda_n P_n(t) = \lambda_0 \cdot P_0(i)\cdot e^{-\lambda_0 t}$$

so that we have

$$S(t) - S(i) = P_0(i)\cdot [e^{\lambda_0 t} - 1]$$

Knowledge of $S(t)$, $S(i)$, and $P(t)$ enables the calculation of the age value t.

(2) If we now consider the presence of one of the daughter elements, say $P_m(i)$ at $t = 0$, this contributes a term $(\lambda_m \ldots \lambda_{n-1})\ P_m(i)\cdot \sum_m^n C_{mr}\cdot e^{-\lambda_r t}$. Considering again the case, $\lambda_0 \ll \lambda_r$, so that $\lambda_r t \gg 1$, for the values of t, physically important, and integrating, we find the contribution to $S(t)$,

$$[\lambda_m \ldots \lambda_{n-1}) P_m(i)\cdot \Sigma \frac{C_{mr}}{\lambda_r} = P_m(i)$$

as can be directly worked out.

This is physically evident, as the whole of the initial daughter has gone over to the stable product.

3. Escape of Parent, Daughter and Intermediates

More interesting is the case where one of the daughter products is lost continuously (say) by diffusion. A practical case is the leakage of radon from uranium minerals—a factor that vitiates age determinations on such minerals by the lead method.[3]

One can assume to a first approximation that the loss by diffusion is proportional to the concentration so that, we have for the build-up of the daughter element P_m,

$$DP_m = \lambda_{m-1}\cdot P_{m-1} - \lambda_m P_m - \alpha\cdot P_m$$

where α represents a leakage factor.

Hence the Laplace transform

$$\bar{P}_m(p) = \frac{\lambda_{m-1}}{(p + \lambda_m + \alpha)}\cdot \bar{P}_{m-1}(p)$$

Again for the case $\lambda_0 \leqslant \lambda_r$ for $r = 1 \ldots n$

$$\overline{P}_n = \frac{\lambda_0 \ldots \lambda_{n-1}}{(\lambda_0 + p)\ldots(\lambda_m + \alpha + p)\ldots(\lambda_n + p)} \cdot P_0(i)$$

Hence as before

$$P_n = P_0(i) \cdot \frac{\lambda_m}{\lambda_m + \alpha} \cdot e^{-\lambda_0 t}$$

and

$$S(t) - S(i) = \frac{\lambda_m}{\lambda_m + \alpha} \cdot P_0(t)\,[e^{-\lambda_0 t} - 1]$$

The end-product is thus reduced by a fraction, $(\lambda_m/\lambda_m + \alpha)$, where α represents the leakage factor.

A special case arises when the parent itself is bled away at a constant rate. In this case evidently,

$$P_n = \frac{\lambda_0 \ldots \lambda_{n-1}}{(\lambda_0 + \alpha + p)\ldots(\lambda_n + p)} \cdot P_0(i)$$

so that again

$$P_n = P_0(i) \cdot \frac{\lambda_0}{\lambda_n} \cdot e^{-(\lambda_0 + \alpha)t}$$

$$S(t) - S(i) = \frac{\lambda_0}{\lambda_0 + \alpha} \cdot P_0(t)\,[e^{(\lambda_0 + \alpha)t} - 1]$$

Finally, we have to consider the case of the escape of the daughter element. In this case,

$$\frac{dS}{dt} = \lambda_0 P_0(i)\, e^{-\lambda_0 t} - \alpha S$$

where α is the escape factor. In this case,

$$\bar{S}(p) = \frac{\lambda_0 \cdot P_0(i)}{(p + \lambda_0)(p + \alpha)} + \frac{S(i)}{p + \alpha}$$

and since $P_0(t) = P_0(i) \cdot e^{-\lambda t}$, we have for $S(i) = 0$,

$$S[t] = \frac{\lambda_0}{\lambda_0 - \alpha} \cdot P_0[t] \cdot [e^{(\lambda_0 - \alpha)t} - 1] \quad \text{if } \lambda > \alpha$$

while for the case, $\alpha > \lambda$,

$$S[t] = \frac{\lambda_0}{\alpha - \lambda_0} \cdot P_0(t)\,[1 - e^{-(\alpha - \lambda_0)t}]$$

The application of this case to the diffusion of helium from rocks and minerals has been discussed by the author.[4]

4. Sudden Removal of Parent or Decay Element

Equally important is the loss of parent or daughter elements due to processes that can be regarded as "catastrophic". Thus, consider the case where a fraction f of the parent is lost at time $t = t_1$;

$$S[t] - S[i] = \lambda_0 \cdot P_0(i) \cdot \left[\int_0^t e^{-\lambda_0 t}\, dt + f \int_{t_1}^t e^{-\lambda_0 t}\, dt \right]$$

$$P_0(t) = (1-f) \cdot P_0(i) \cdot e^{-\lambda t}$$

so that,

$$S(t) - S(i) = \frac{P_0(t)}{1-f} \cdot [e^{\lambda_0 t} - 1 + f \cdot e^{\lambda_0 t}(1 - e^{-\lambda_0 t_1})]$$

Actually, the chemical lead-uranium and lead-thorium methods are vitiated by selective leaching of the uranium and thorium by acid waters.[5] It is interesting to note that the (Pb^{206}/Pb^{207}) method, based on a comparison of the lead isotopes resulting from the decay of the uranium isotopes, suffers less from this error as both the isotopes are removed to the same extent by leaching. In fact, it happens that compensation is exact for $\lambda_0 t \ll 1$, for, in this case, retaining only the first term in the exponentials of the above equation,

$$S(t) - S(i) = (Pb^{206}) = \frac{(U^{238})}{(1-f)} \cdot \lambda_0 (t - f t_1)$$

$$S(t) - S(i) = (Pb^{207}) = \frac{(U^{235})}{(1-f)} \cdot \lambda_0^* (t - f t_1)$$

so that,

$$\frac{(Pb^{206})}{(Pb^{207})} = \frac{(U^{238})}{(U^{235})} \cdot \frac{\lambda_0}{\lambda_0^*}$$

where λ_0 and λ_0^* are the decay constants of U^{238} and U^{235} respectively.

Again, the same relation holds when we consider the case of the continuous escape of the parent, treated in the last paragraph this gives

$$S(t) - S(i) = \frac{\lambda_0}{\lambda_0 + a} \cdot P_0(t) \cdot [(\lambda_0 + a)\, t + (\lambda_0 + a)^2\, t^2/2 + \ldots]$$

For the case where $(\lambda_0 + a)\, t \ll 1$, retaining only the first terms in the exponential, the ratio Pb^{207}/Pb^{206} remains unchanged.

Finally, we can consider the case where a fraction f of one of the intermediate daughters is removed at time $t = t_1$. It is evident that for the case $\lambda_0 \ll \lambda_r$, the decrease in the final decay product, $S(t) - S(i)$ is,

$$\Delta S(t) = P_0(i) \cdot e^{-\lambda_0 t_1} \cdot \frac{\lambda_0}{\lambda_r}$$

$$P_0(t) \cdot \frac{\lambda_0}{\lambda_1} \cdot e^{-\lambda_0(t-t_1)}$$

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