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Error analysis of large-time approximations for decay chains

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ABSTRACT

Consider a radioactive decay chain $X_1 \to \cdots \to X_n \to$ and let $N_n(t)$ be the amount of X_n at time t. This paper establishes error bounds for large-time approximations to $N_n(t)$ that include and generalize the transient equilibrium approximations and other known approximations. The error bounds allow one to find the range of t for which these approximations can be used with a given degree of precision.

1. Introduction

Consider a radioactive decay chain $X_1 \to X_2 \to \cdots \to X_n \to$ where a fraction b_j of the nuclide X_j decays into X_{j+1} with decay constant λ_j and half-life $T = ln(2)/\lambda_j$. The remaining fraction $1 - b_j$ of X_j decays with the same decay constant λ_j into other nuclides outside the chain. If X_n is stable then $\lambda_n = 0$. Let $N_j(t)$ be that portion of the amount of X_j present at time t that has been produced by decays following the chain. The $N_j(t)$ satisfy the radioactive decay equations:

$$\begin{aligned} dN_1/dt &= -\lambda_1 N_1, \\ dN_j/dt &= b_{j-1}\lambda_{j-1}N_{j-1} - \lambda_j N_j \quad \text{for } j \geqslant 2, \end{aligned} \tag{1}$$

see Segrè [1, p. 172]. If $N_j(0) = 0$ for $j \geqslant 2$ then Bateman's formula [2] for $N_n(t)$ is

$$N_n(t) = N_1(0) b_{1,n}\lambda_{1,n} E_n(t; \lambda_1, \ldots, \lambda_n), \tag{2}$$

where

$$E_n(t; \lambda_1, \ldots, \lambda_n) = \sum_{j=1}^{n} C_j e^{-\lambda_j t}, \tag{3}$$

$$b_{i,j} = b_i b_{i+1} \cdots b_{j-1} \quad \text{and} \quad \lambda_{i,j} = \lambda_i \lambda_{i+1} \cdots \lambda_{j-1} \quad \text{and} \quad C_j = \prod_{\substack{i=1 \\ i \neq j}}^{n} (\lambda_i - \lambda_j)^{-1}$$

and $b_{i,i} = \lambda_{i,i} = 1$ and $E_1(t;\lambda) = e^{-\lambda t}$. This formula is only defined if all the λ_j are distinct. Formulas for $E_n(t;\lambda_1,\ldots,\lambda_n)$ when some of the λ_j are equal are more complicated; see Mathai [3, Section 2] and Cetnar [4, Section 3].

There are several useful large-time approximations for $N_n(t)$. These involve the smallest of $\lambda_1,\ldots,\lambda_n$ which we denote by λ_p. Note that in (3) the $e^{-\lambda_j t}$ for $j \neq p$ go to zero faster than $e^{-\lambda_p t}$ as $t \to \infty$ which implies

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$$N_n(t) \approx N_1(0)ae^{-\lambda_p t} \tag{4}$$

for large t with $a = b_{1,n}\lambda_{1,n}C_p$. This approximation holds in the sense that $N_n(t)/[N_1(0)ae^{-\lambda_p t}] \to 1$ as $t \to \infty$. The approximation (4) is widely used, see e.g. Rutherford et al. [5, p. 3] and Segrè [1, p. 174]. However, we could find no mention of how accurate it is. The following theorem gives an error bound for this approximation.

Theorem 1. *Let λ_p and λ_q be the smallest and 2nd smallest of $\lambda_1,\ldots,\lambda_n$ respectively and $a = b_{1,n}\lambda_{1,n}C_p$. Assume $\lambda_p < \lambda_q$ and λ_q is strictly less than the 3rd smallest of $\lambda_1,\ldots,\lambda_n$. Then*

$$|N_n(t) - N_1(0)ae^{-\lambda_p t}| \leqslant ce^{-(\lambda_q-\lambda_p)t}N_1(0)ae^{-\lambda_p t} \tag{5}$$

where $c = \prod_{\substack{j=1\\ j\neq p,q}}^{n}(\lambda_j - \lambda_p)/(\lambda_j - \lambda_q)$.

Theorem 1 is the special case of Theorem 3 below when $m = 1$ in that theorem. Note that it follows from (5) that the relative error in (4) will be less than ε if $t > \ln(c/\varepsilon)/(\lambda_q - \lambda_p)$, which tells how large t must be for (4) to hold with a given degree ε of precision. In particular, the relative error in (4) will be small if t is large compared to the second largest half-life unless the second largest half-life is close to the largest. See Example 1 in Section 3 for a typical application of Theorem 1.

Products like the one defining c in Theorem 1 appear in a number of the inequalities below, so it is useful to make the following definition.

Definition 1. Let α_p and α_q be the smallest and second smallest of $\alpha_1,\ldots,\alpha_k$. If β is different from $\alpha_1,\ldots,\alpha_k$ and α_q is strictly smaller than the third smallest of the α_j let

$$\phi(\alpha_1,\ldots,\alpha_k|\beta) = \prod_{j=1}^{k}\alpha_j/(\alpha_j - \beta),$$
$$\chi(\alpha_1,\ldots,\alpha_k) = \prod_{\substack{j=1\\ j\neq p,q}}^{k}(\alpha_j - \alpha_p)/(\alpha_j - \alpha_q).$$

In the case $k = 2$ let $\chi(\alpha_1,\alpha_2) = 1$.

Note that $\chi(\alpha_1,\ldots,\alpha_k) \approx 1$ if α_q is substantially smaller than the third smallest of the α_j which occurs frequently in decay chains. Also note that $c = \chi(\lambda_1,\ldots,\lambda_n)$ in Theorem 1.

Another useful approximation is

$$N_n(t) \approx aN_k(t) \tag{6}$$

for large t; see Segrè [1, p. 174]. Here $p \leqslant k < n$ and $a = (b_{k,n}\lambda_k/\lambda_n)\phi(\lambda_{k+1},\lambda_{k+2},\ldots,\lambda_n|\lambda_p)$ and (6) holds in the sense that $N_n(t)/[aN_k(t)] \to 1$ as $t \to \infty$. One way to see that this is valid is to apply (4) to both $N_n(t)$ and $N_k(t)$ giving $N_n(t)/[N_1(0)b_{1,n}\lambda_{1,n}C_p e^{-\lambda_p t}] \to 1$ and $N_k(t)/[N_1(0)b_{1,k}\lambda_{1,k}C_{p,k}e^{-\lambda_p t}] \to 1$ as $t \to \infty$ where $C_{p,k} = \prod_{\substack{i=1\\ i\neq p}}^{k}(\lambda_i - \lambda_p)^{-1}$. Dividing gives $N_n(t)/[aN_k(t)] \to 1$ as $t \to \infty$. The following theorem gives two error bounds for (6). The first, (7), is similar to (5) while (8) shows that the approximation (6) is good for a larger range of values of t than (7). However (8) is not always applicable.

Theorem 2. *Let λ_p and λ_q be the smallest and 2nd smallest of $\lambda_1,\ldots,\lambda_n$ respectively, $p \leqslant k < n$ and $a = (b_{k,n}\lambda_k/\lambda_n)\phi(\lambda_{k+1},\lambda_{k+2},\ldots,\lambda_n|\lambda_p)$. For (7) assume $\lambda_p < \lambda_q$ and λ_q is strictly less than the 3rd smallest of $\lambda_1,\ldots,\lambda_n$ and let $c = \chi(\lambda_1,\ldots,\lambda_n)$. For (8) assume λ_q is strictly less than all of $\lambda_{k+1},\ldots,\lambda_n$ and let $W = (k-1)\chi(\lambda_p,\lambda_q,\lambda_{k+1},\ldots,\lambda_n)\sum_{j=k+1}^{n}1/(\lambda_j - \lambda_p)$. Then*

$$|N_n(t) - aN_k(t)| \leqslant ce^{-(\lambda_q-\lambda_p)t}aN_k(t), \tag{7}$$
$$|N_n(t) - aN_k(t)| \leqslant aN_k(t)W/t. \tag{8}$$

Theorem 2 is the special case of Theorem 3 when $m = k$ and $\sigma(j) = j$ for $j = 1,\ldots,n$ in that theorem. Note that it follows from (8) that if $q < p$ then the relative error in (6) will be less than ε if $t > W/\varepsilon$. Again, this tells how large t must be for (6) to hold with a given degree of precision. If fact, if $\lambda_{k+1},\ldots,\lambda_n$ are all substantially larger than λ_q then W will be of the same order of magnitude as the largest of the half-lives of $X_{k+1},\ldots,X_n$. In this case the relative error in (6) will be small if t is large compared to the half-lives of $X_{k+1},\ldots,X_n$. This will be a larger range of values of t than the range implied by (7). For a typical application, see Example 1 in Section 3.

The *activity*, $A_j(t)$ of X_j is the rate of decay of X_j, i.e. $A_j(t) = \lambda_j N_j(t)$. It follows from (6) that $A_n(t)/A_p(t) \approx b_{p,n}[\lambda_{p+1}/(\lambda_{p+1} - \lambda_p)]\cdots[\lambda_n/(\lambda_n - \lambda_p)]$ for large t. In other words the activity of X_n is approximately proportional to that of X_p for large t, a situation that is termed *transient equilibrium*; see Segrè [1, p. 174].

A generalization of (4) and (6) is obtained by approximating the original chain by a reduced chain that is obtained by deleting some nuclides with small half-lives, usually the nuclides whose half-lives are less than a certain threshold. This type of approximation is also widely used; see e.g. Benedict et al. [6, p. 39], Ball and Adams [7], Bell [8] and Thomas and Barber [9]. To be precise, assume the following.