

# General solution of Bateman equations for nuclear transmutations

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## Abstract

The paper concerns the linear chain method of solving Bateman equations for nuclear transmutation in derivation of the general solution for linear chain with repeated transitions and thus elimination of existing numerical problems. In addition, applications of derived equations for transmutation trajectory analysis method is presented.

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## 1. Introduction

The time evolution of nuclide concentrations undergoing serial or linear decay chain is governed by a set of first-order differential equation, called Bateman equations. For complex radioactive decay schemes or transmutations in particle flux the equations differ from linear chain in that a higher number of coefficients and equations appears leading to complication of the solution. The analytical solution of Bateman equations for the case of radioactive decay in a linear chain given early by Bateman (1910), is used then also for a non-linear chain after breaking it down into a set of linear chains. This is called the linear chain method. Examples of this method application in numerical codes are BISON (Furuta et al., 1987) and CINDER (Wilson et al., 1998). The matrix calculus is applied in other methods, among which the most often used is ORIGEN (Croff, 1980). Generally, both methods assume that all coefficients appearing in the equations are different, which in a real case of transmutation or compound decay with branching and chain loops is not always satisfied. In such cases numerical algorithms face complication due to not satisfied assumptions lying behind analytical solutions or a lack of numerical convergence (Thomas and Barber, 1994). Improvement of algorithms based on linear chain method

can be obtained when the Bateman equation solution for a linear chain in general case is derived and applied.

## 2. Bateman equations for serial decay chain

The Bateman equations for radioactive decay case of  $n$ -nuclide series in linear chain describing nuclide concentrations are as follows:

$$\begin{aligned} \frac{dN_1}{dt} &= -\lambda_1 N_1 \\ \frac{dN_i}{dt} &= \lambda_{i-1} N_{i-1} - \lambda_i N_i \quad (i = 2, n) \end{aligned} \quad (1)$$

where  $\lambda_i$  is the decay constant of  $i$ th nuclide.

Assuming zero concentrations of all daughters at time zero

$$N_1(0) \neq 0 \quad \text{and} \quad N_i(0) = 0 \quad \text{when } i > 1 \quad (2)$$

the concentration of  $n$ th nuclide after time  $t$  was given by Bateman.

$$N_n(t) = \frac{N_1(0)}{\lambda_n} \sum_{i=1}^n \lambda_i \alpha_i \exp[-\lambda_i t] \quad (3)$$

where

$$\alpha_i = \prod_{\substack{j=1 \\ j \neq i}}^n \frac{\lambda_j}{(\lambda_j - \lambda_i)} \quad (4)$$

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Although in most cases of radioactive decay the above expression is sufficient, it is apparent that the coefficients  $\alpha$  can be calculated only if all decay constants are different. Otherwise infinity in Eq. (4) occurs. The presence of infinities does not mean in our case that concentrations are infinite. Simply, there are infinite terms of opposite side in the sum present in the equation, thus the sum does not converge. Also in cases when the constants are different but very close the digital errors in calculating Eq. (3) can strongly bias the numerical results. Nevertheless, this simple formulas are often used also in burnup codes. The Bateman equations for nuclear transmutation system are more complicated than for cases of radioactive decay, which implies that the infinities occur, thus the above solutions cannot be readily applied. Similar case occurs concerning the matrix where the decay constant matrix, which is triangular, must be replaced by the matrix of transmutation constants, which in general can become singular.

### 3. Removing infinities

There are two ways of removing infinities. The first is to artificially shift the constants that are equal and obtain an approximate solution. The second is to derive the solutions of the general case that allows for equalities among the constants. In case of transmutation system we will deal with transmutation constants instead of the decay constant. A transmutation chain is generally not linear and path branching will have to be considered but the solution will differ from the case without branching only by a multiplicative constant. For simplifying the consideration let us put aside for a moment the possible branching in transmutation chain.

Let us consider a case of linear chain that involves  $n$  different direct nuclide-to-nuclide transitions described by a set of  $n$  transmutation constants  $\lambda_i$ , ( $i = 1, n$ ) but with possible repetitions. Let the number of  $k$ th transition occurrences in the chain equals  $m_k$ . The shift procedure changes in the chain set of  $m_k$  equal constants:  $\lambda_k$  into set of  $m_k$  constants separated by a small number  $\Delta$ :  $\lambda_k, \lambda_k + \Delta, \dots, \lambda_k + \mu_k \Delta$ , where for notation simplicity we use  $\mu_k = m_k - 1$ . If a numerical algorithm based on Eq. (3) applies an appropriate value of  $\Delta$ , it will be able to produce an accurate result of the numerical calculation (within the limits of accepted numerical errors of order as low as  $10^{-10}$ ). This approach was successfully applied in the initial version of TTA code (Cetnar, 1999) and then MCB (Cetnar et al., 1998), where a numerically stable algorithm was established.

However, in some extreme cases of very high radiation flux resulting in complication of the transmutation chain this approach can also face a digital limitation. For such cases the derivation of an exact general solution is a necessity. For simplicity we assume zero concentrations of all daughters at time zero and unit concentration of the first nuclide

$$N_1(0) = 1 \quad \text{and} \quad N_i(0) = 0 \quad \text{when } i > 1 \quad (5)$$

Now, we will use an approximate solution in the shift procedure for disintegration rate of the last chain nuclide. In cases when the last chain nuclide occurs more than once in the chain this expression is a partial disintegration rate since it does not include disintegrations taking place earlier in the chain. For finding the general solution we will find the limit of approximate value as  $\Delta$  approaches zero

$$\begin{aligned} A(t) &= \frac{N_n(t)}{N_1(0)} \lambda_n \\ &= \sum_{i=1}^n \lim_{\Delta_i \rightarrow 0} \sum_{m=0}^{\mu_i} \exp[-(\lambda_i + m\Delta_i)t] \\ &\quad \times \left( \prod_{\substack{l=0 \\ l \neq m}}^{\mu_i} \frac{\lambda_i + l\Delta_i}{(l-m)\Delta_i} \right) \cdot \prod_{\substack{j=1 \\ j \neq i}}^n \prod_{k=0}^{\mu_j} \lim_{\Delta_j \rightarrow 0} \frac{\lambda_j + k\Delta_j}{\lambda_j + k\Delta_j - \lambda_i - m\Delta_i} \\ &= \sum_{i=1}^n \lambda_i^{m_i} \exp[-\lambda_i t] \cdot \lim_{\Delta_i \rightarrow 0} \Delta_i^{-\mu_i} \sum_{m=0}^{\mu_i} \exp[-m\Delta_i t] \\ &\quad \times \left( \prod_{\substack{l=0 \\ l \neq m}}^{\mu_i} \frac{1}{(l-m)} \right) \prod_{\substack{j=1 \\ j \neq i}}^n \prod_{k=0}^{\mu_j} \lim_{\Delta_j \rightarrow 0} \\ &\quad \times \frac{\lambda_j + k\Delta_j}{\lambda_j - \lambda_i + k\Delta_j - m\Delta_i} \end{aligned} \quad (6)$$

In the above equation we calculate the limit of  $\Delta_j$ , by a direct substitution, expand the exponent function which contains  $\Delta_i$  into Taylor series as well as apply the relation

$$\prod_{\substack{l=0 \\ l \neq m}}^{\mu_i} \frac{1}{(l-m)} = \frac{(-1)^m}{m!(\mu_i - m)!} = \frac{(-1)^m}{\mu_i!} \binom{\mu_i}{m} \quad (7)$$

to obtain

$$\begin{aligned} A(t) &= \sum_{i=1}^n \frac{\lambda_i^{m_i} \exp[-\lambda_i t]}{\mu_i!} \cdot \lim_{\Delta \rightarrow 0} \Delta^{-\mu_i} \sum_{m=0}^{\mu_i} (-1)^m \binom{\mu_i}{m} \\ &\quad \cdot \sum_{k=0}^{\infty} \frac{(-m\Delta \cdot t)^k}{k!} \prod_{\substack{j=1 \\ j \neq i}}^n \frac{\lambda_j^{m_j}}{(\lambda_j - \lambda_i - m\Delta)^{m_j}} \end{aligned} \quad (8)$$

Taking advantage of the following relations:

$$\begin{aligned} \frac{1}{(a-x)} &= \frac{1}{a} \sum_{n=0}^{\infty} \left(\frac{x}{a}\right)^n \quad \text{and} \quad \left(\sum_{n=0}^{\infty} x^n\right)^{k+1} \\ &= \sum_{n=0}^{\infty} \binom{n+k}{k} x^n \end{aligned} \quad (9)$$

the last term in Eq. (8) can be written as

$$\begin{aligned} \frac{\lambda_j^{m_j}}{(\lambda_j - \lambda_i - m\Delta)^{m_j}} &= \left(\frac{\lambda_j}{\lambda_j - \lambda_i}\right)^{m_j} \frac{1}{\left(1 - \frac{m\Delta}{\lambda_j - \lambda_i}\right)^{m_j}} \\ &= \left(\frac{\lambda_j}{\lambda_j - \lambda_i}\right)^{m_j} \sum_{h=0}^{\infty} \binom{h + m_j}{m_j} \left(\frac{m\Delta}{\lambda_j - \lambda_i}\right)^h \end{aligned} \quad (10)$$

After substituting in Eq. (8) we have

$$\begin{aligned}
 A(t) &= \left( \prod_{i=1}^n \lambda_i^{m_i} \right) \sum_{i=1}^n \frac{\exp[-\lambda_i t]}{\mu_i!} \cdot \sum_{m=0}^{\mu_i} (-1)^m \binom{\mu_i}{m} \\
 &\cdot \lim_{\Delta \rightarrow 0} \frac{1}{\Delta^{\mu_i}} \sum_{k=0}^{\infty} \frac{(-m\Delta \cdot t)^k}{k!} \\
 &\times \prod_{\substack{j=1 \\ j \neq i}}^n \left[ \frac{1}{(\lambda_j - \lambda_i)^{m_j}} \sum_{h=0}^{\infty} \binom{h + \mu_j}{\mu_j} \left( \frac{m\Delta}{\lambda_j - \lambda_i} \right)^h \right] \quad (11)
 \end{aligned}$$

In Eq. (11) the expression under the limit is made of products of  $n$  functions of  $m\Delta$  expanded into Taylor series and divided by  $\Delta^{\mu}$ . With  $\Delta$  approaching zero only the terms of the sum products that contain  $\Delta^{\mu}$  will not vanish. The terms with smaller order will cancel out

$$\begin{aligned}
 A(t) &= \sum_{i=1}^n \left( \prod_{\substack{j=1 \\ j \neq i}}^n \left( \frac{\lambda_j}{\lambda_j - \lambda_i} \right)^{m_j} \right) \lambda_i^{m_i} \frac{\exp[-\lambda_i t]}{\mu_i!} \\
 &\cdot \sum_{m=0}^{\mu_i} m^{\mu_i} (-1)^m \binom{\mu_i}{m} \\
 &\cdot (-1)^{\mu_i} \sum_{k=0}^{\mu_i} \sum_{h_1=0}^{\mu_i} \sum_{h_2=0}^{\mu_i} \dots \sum_{h_n=0}^{\mu_i} \frac{t^k}{k!} \prod_{\substack{j=1 \\ j \neq i}}^n \binom{h_j + \mu_j}{\mu_j} \\
 &\times \left( \frac{1}{\lambda_i - \lambda_j} \right)^{h_j} \delta \left( \mu_i, k + \sum_{\substack{l=0 \\ l \neq i}}^n h_l \right) \quad (12)
 \end{aligned}$$

where the Kronecker delta was used

$$\begin{aligned}
 \delta(i, j) &= 1 \iff i = j \\
 \delta(i, j) &= 0 \iff i \neq j
 \end{aligned} \quad (13)$$

We can find out that

$$\sum_{m=0}^{\mu_i} \frac{m^{\mu_i} (-1)^m}{\mu_i!} \binom{\mu_i}{m} = (-1)^{\mu_i} \quad (14)$$

which will be canceled with the similar term in Eq. (12) which appeared there from the sign changes in the sum product terms. Finally, we can write the solution in the following form:

$$A(t) = \sum_{i=1}^n \lambda_i \alpha_i \exp[-\lambda_i t] \cdot \sum_{m=0}^{\mu_i} \frac{(\lambda_i t)^m}{m!} \cdot \Omega_{i, \mu_i - m} \quad (15)$$

where

$$\alpha_i = \prod_{\substack{j=1, n \\ j \neq i}} \left( \frac{\lambda_j}{\lambda_j - \lambda_i} \right)^{m_j} \quad (16)$$

and the omega terms for  $i \in [1, n]$  and  $j \in [0, \mu_i]$  take the following forms:

$$\begin{aligned}
 \Omega_{i,j} &= \sum_{h_1=0}^j \sum_{h_2=0}^j \dots \sum_{h_n=0}^j \\
 &\times \prod_{\substack{k=1 \\ k \neq i}}^n \binom{h_k + \mu_k}{\mu_k} \left( \frac{\lambda_i}{\lambda_i - \lambda_k} \right)^{h_k} \delta \left( j, \sum_{\substack{l=0 \\ l \neq i}}^n h_l \right) \quad (17)
 \end{aligned}$$

thus particularly

$$\Omega_{i,0} = 1$$

Although the formulas in obtained solution look rather complex they can be effectively calculated digitally. The binomial coefficients for all required omega functions can be calculated altogether in a single digital loop as they are presented in a recurrent formula. For this purpose, the binomial coefficients can be obtained recurrently from constructed Pascal's triangle.

#### 4. Nuclear transmutation systems

In a transmutation system nuclides are being transmuted also due to interaction with particle flux, mainly neutrons, but in the presence of a spallation target also high energy protons or pions. The decay constants that govern Bateman equations for a decay case are then substituted by transmutation constants. At this point, by the transmutation constant  $\lambda_{i,j}$  we understand probability of the  $i$ th nuclide production per time unit from the  $j$ th nuclide destruction, as a result of nuclear interaction with the whole spectrum of interacting particles or due to the natural nuclear decay. It can be expressed as follows:

$$\lambda_{i,j} = b_{i,j}^d \cdot \lambda_j^d + \sum_{x=n,p,\pi} \int \Phi^x(E) \sigma_{i,j}^x(E) dE \quad (18)$$

where  $\lambda_j^d$  is the decay constant of  $j$ th nuclide;  $b_{i,j}^d$  is the branching ratio of  $j$ th nuclide decay into  $i$ th nuclide;  $\Phi^x$  is the particle flux ( $x = \text{neutrons, protons, pions } +, 0, -$ );  $\sigma_{i,j}^x$  is the cross-section for production of  $i$ th nuclide by particles  $x$  during interaction with  $j$ th nuclides.

The transmutation constants appear as the coefficients of the Bateman equations describing the general, non-linear transmutation chain for  $w$  nuclides as follows:

$$\frac{dN_i}{dt} = \sum_{j=1, w} \lambda_{i,j} \cdot N_j \quad (i = 1, w) \quad (19)$$

where  $N_i$  denotes the concentration of  $i$ th nuclide. The constants  $\lambda_{i,i}$ , which are not covered in Eq. (18) are defined using the mass flow balance as follows:

$$\lambda_{i,i} = - \sum_{\substack{j=1 \\ j \neq i}}^w \lambda_{i,j} \quad (20)$$

and they physically represent nuclide disintegration rate. The mass flow balance condition guarantees that the overall nuclide mass remains constant. Since the set of Eq. (18) is a set of first-order linear differential equations, the

solution of a complex case can be obtained as a linear superposition of the solutions in simpler cases.

### 5. Transmutation trajectory analysis

Usually, in the applied numerical methods the set of linear chains is prepared arbitrarily, which is sufficient for well-defined cases. However, for more general case application of the procedure that resolves the non-linear chain into a set of linear chains is necessary to assure the mass flow balance and the numerical solution stability.

For the purpose of deriving the solution for a general case basing on the known solution for a linear decay case, it is convenient to focus on the transmutation transition from one nuclide to the other one after elapsing time  $t$ . The transmutation transitions can lead through many paths, which possibly branch, forming a non-linear chain. Now, let us define a transmutation trajectory as a sequence of direct nuclide to nuclide transitions, starting from the first nuclide and ending at the last,  $n$ th nuclide. The transmutation trajectory is almost equivalent to a decay chain, but due to an occurrence of branching in the non-linear chain the mass flow is not preserved on a single trajectory level. It is preserved, however, all over the trajectories that can be extracted from the non-linear chain. The Bateman equations for transmutation trajectory will have the following form:

$$\begin{aligned} \frac{dN_1}{dt} &= -\lambda_1 N_1 \\ \frac{dN_i}{dt} &= b_i \lambda_{i-1} N_{i-1} - \lambda_i N_i \quad (i = 2, n) \end{aligned} \quad (21)$$

where

$$\begin{aligned} \lambda_i &= \sum_{\substack{j=1 \\ j \neq i}}^w \lambda_{i,j} \\ b_i &= \frac{\lambda_{i,i+1}}{\lambda_i} \quad (i = 1, n) \end{aligned} \quad (22)$$

The numbering of nuclides in each trajectory ( $i = 1, n$ ) is independent from actual nuclide numbering occurring in the whole non-linear chain ( $j = 1, w$ ), first for the sake of simplicity, but also due to the fact that a particular trajectory can lead through one nuclide several times, following the already passed direct nuclide transitions (reaction channel or decay). The solution of Eq. (21) can be obtained from those of the decay case by simple introduction of a branching factor. The concentration of the last trajectory nuclide due to the trajectory transition can be written using Eq. (15) as:

$$N_n(t) = N_n(0) \frac{B}{\lambda_n} A(t) \quad (23)$$

where the branching factor

$$B = \prod_{k=1, n-1} b_k \quad (24)$$

The disintegration or removal rate of the last trajectory nuclide can be expressed as follows:

$$A_n(t) = N_n(0) \cdot B \cdot A(t) \quad (25)$$

It can be thought of as generalized activity. The physical meaning of the concentration (23) and the disintegration rate (25) concerns, respectively, only partial contributions to actual nuclide concentration and disintegration, due to transitions along the considered trajectory. The time integral of the removal rate (25) leads to the following function:

$$I(t) = \int_0^t A_n(t) dt = N_1(0) B \sum_{m=0}^{\mu_i} \Omega_{i, \mu_i - m} \cdot f_{i,m}(t) \quad (26)$$

where

$$f_{i,m}(t) = 1 - \exp[-\lambda_i t] \cdot \sum_{k=0}^m \frac{(\lambda_i t)^k}{k!} \quad (27)$$

The above stands for the sum of the concentrations of nuclides formed as a result of the  $n$ th nuclide disintegration, or of their daughters after being produced from the transition along considered trajectory. For given time  $t$ , every transmutation trajectory can be characterized by two quantities: the trajectory transition:

$$T(t) = N_n(t) / N_1(0) \quad (28)$$

and the trajectory passage:

$$P(t) = I(t) / N_1(0) \quad (29)$$

The transition and passage functions are important for the mass balance of transmutations, which is an ultimate parameter for checking correctness and convergence of any numerical algorithm for calculation of time evolution of concentrations in a transmutation system. They can be used to control the numerical algorithm of breaking down a non-linear transmutation chain into a series of transmutation trajectories for which the concentration can be calculated.

#### 5.1. Transmutation trajectory generator

Although the number of possible transmutation trajectories is infinite, the functions  $P(t)$  and  $T(t)$  allow us to make a trajectory generator that forms the trajectories reflecting their formation process in reality and to put them in a series while controlling the mass balance. Let us consider non-linear chain that starts from one nuclide, that is all other nuclide densities equal zero at the beginning of calculations. The trajectories form generations as they are formed in the following manner. The first generation consists of only one trajectory, which contains only one, initial nuclide. Physically, it represents the survival of the initial nuclide without decay or reaction. In this case the transition and passage are expressed as follows:

$$T_{1,0}(t) = 1 - \exp[-\lambda_i t] \quad (30)$$

$$P_{1,0}(t) = \exp[-\lambda_i t] \quad (31)$$

In the notation used for functions  $P$  and  $T$  the first index is the trajectory number while the second one is the parent trajectory number.

The second generation of trajectories is formed by appending the last nuclide by one of the possible nuclides that can be formed from the last one through a possible reaction channel or decay path. All trajectories in the second generation can be considered daughter trajectories of the only trajectory in the first generation. The next trajectory generations are formed in similar way by appending an additional nuclide to a trajectory from the older generation. The generation number equals the trajectory length. Theoretically the trajectory generator can create infinite number of generations with growing number of trajectories, but, as one can expect, the trajectory extension process leads to a consecutive decrease of transmutation passage. After the complete extension of a trajectory, let us say  $k$ , to daughter trajectories, let us say from  $l$  to  $m$ , the concentration represented by the passage  $P_{k,x}(t)$  is distributed to transitions and passages of its daughter trajectories:

$$P_{k,x}(t) = \sum_{i=1}^m (T_{i,k}(t) + P_{i,k}(t)) \quad (32)$$

By the complete trajectory extension we understand generation of all daughter trajectories to a concerned parent, which can be physically formed. To keep the mass flow properly we can analyze mass balance after a particular complete trajectory extension. For generated series of  $m$  completely extended trajectories, the residual passage  $R_m(t)$  is defined as a sum of passages for trajectories that were not yet extended:

$$R_m(t) = \sum_{i \notin X} P_{i,x}(t) \quad (33)$$

where  $X$  is the set of parent trajectories indexes.

The total transmutation transition is

$$T_m(t) = \sum_{i=1}^m T_{i,x}(t) \quad (34)$$

It follows that:

$$R_m(t) + T_m(t) = 1 \quad (35)$$

which is the transmutation mass flow balance equation.

After breaking down of non-linear transmutation chain into the series of  $m$  trajectories the residual passage  $R_m(t)$  represents the fraction of initial concentration which is not yet assigned to any nuclide while the total transition  $T_m(t)$  represents the sum of assigned concentrations. Obviously the residual passage decreases with the generation number and it is greater than concentration of any nuclide that would emerge due to further series extension, so it can be used as a criterion for truncation of the trajectory series. To conclude trajectory generation process one needs to

control the trajectory extension process and do not extend trajectories that will not lead to transition contributions above the desired level. To control quantitatively this process the value of trajectory passage  $P_{i,j}(t)$  of the trajectory considered for extension is significant. If it falls below assumed truncation parameter,  $\varepsilon$ , the extension of this particular trajectory is blocked. This way the infinite series of trajectories is reduced to the limited series of meaningful trajectories that can contribute to nuclide density vector above the required threshold level. After complete transmutation trajectory formation process the nuclide concentrations of the general non-linear chain (19), are obtained by the summation of respective contributions:

$$N_k(t) = N_0(t) \sum_{i=1}^m T_{i,x}(t) \cdot \delta_{k,ix(m)} \quad (36)$$

where  $ix(m)$  is the index of the last nuclide in  $m$ th trajectory.

The transmutation trajectory analysis method was first applied for TTA code dedicated to accelerator driven systems (Cetnar, 1999).

## 6. Fission product transmutation modeling

Derived equations concerning the case of the boundary condition (2) can be also adapted for the case of continuously supplied nuclides. In this case, the transmutation trajectory starting nuclide concentration is described as follows:

$$\frac{dN_1}{dt} = -\lambda_1 N_1 + s_1 \quad (37)$$

where  $s_1$  is the nuclide production rate. The production rate can be represented by the decay of an artificial nuclide  $N_0$  with respective constants of:  $b_0$  and  $\lambda_0$ , which satisfy following conditions:

$$N_0(0) = s_1 t \quad \text{and} \quad N_i(0) = 0 \quad \text{when} \quad i > 0$$

and

$$\lambda_0 = \frac{1}{t} \cdot \ln \frac{b_0}{b_0 - 1} \quad \text{where} \quad b_0 \gg 1 \quad (38)$$

Introduction of the artificial nuclide  $N_0$  allows for using Eq. (2) with boundary condition (38) instead of Eq. (37). The continuous supply case occurs for fission products, which are produced with assumed constant rate from actinide fission during the irradiation period. This approach was initially applied in BISON-C (Cetnar and Gronek, 2000) for extending transmutation calculations also to fission products and then in MCB.

## 7. Conclusions

The general solution of Bateman equations for a linear chain with repeated transitions is presented. Derived formulas in combination with presented transmutation trajectory analysis method provide us with tools for developing

numerically stable algorithm for modeling of nuclear transmutation in a general case with an assurance of mass flow balance and generated numerical error. In the presented method the physical conditions of transmutation system such as neutron flux level, considered time scale or transmutation constant range can be much larger than in the case of simple formula or in the matrix approach due to elimination of a possible appearance of infinities in the derived solutions. The numerical algorithm currently undergoes testing of its numerical limitations, which will be the subject of a separate paper. In near future it will be implemented in the new version of MCB.

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