

Analytical solution and simplified analysis of coupled parent-daughter steady-state transport with multirate mass transfer

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[1] In this technical note, a steady-state analytical solution of concentrations of a parent solute reacting to a daughter solute, both of which are undergoing transport and multirate mass transfer, is presented. Although the governing equations are complicated, the resulting solution can be expressed in simple terms. A function of the ratio of concentrations, $\ln(\text{daughter/parent} + P)$, can be used as a metric of mass transfer and reaction if the reactions are mostly confined to the immobile domain, where P is the ratio of production of daughter to decay of parent. This metric is applied with the resazurin-resorufin (Raz-Rru) tracer system in a stream to obtain an integrated measure of respiration that occurs on or in a stream bed. The slope of the graph of $\ln(\text{Rru/Raz} + P)$ versus advective travel time is a function of the strength of surface–bed interaction and respiration. This graph can be used for rapid comparison of different experiments and streams.

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1. Introduction

[2] Reactive or decaying solutes in the environment, e.g., nutrients, tracers, radionuclides, and contaminants, are commonly exchanged between mobile and immobile domains and may approach steady-state concentrations at sufficiently long time with constant boundary conditions. In this technical note, an analytical solution for a parent compound undergoing a first-order reaction to a daughter compound is presented, which can also undergo first-order reaction. Both compounds experience multirate mass transfer with an immobile domain [e.g., Haggerty and Gorelick, 1995; Haggerty et al., 2000; Dentz and Berkowitz, 2003; Luo et al., 2008]. Reactions can occur in both the mobile and immobile domains; however, the analytical solution may be of most use when reactions are limited to the immobile domain. The ratio of the daughter to parent concentration is a simple function that is sensitive only to advective travel time, mass transfer, and reaction.

[3] Some related solutions are as follows: (1) steady-state transport of a single solute with first-order decay and immobile-domain diffusion [Cunningham and Mendoza-Sanchez, 2006]; (2) steady-state transport of two solutes with first-order decay [McLaren, 1969; Burnell et al., 2012]; (3) transient transport of a decay chain [Sun et al., 1999] extended to diffusion into a slab [Sun and Buscheck, 2003] and generalized to compounds with differing retardation factors [Srinivasan and Clement, 2008].

2. Model and Solution

[4] A model for transient solute transport and first-order reaction along a one-dimensional flow path where water is exchanged between mobile and a distribution of immobile zones is described by equations (1)–(4). These equations modified from Argerich et al. [2011] for a metabolically active transient storage fraction of 1 and a distribution of exchange rates as described by Haggerty et al. [2000] or Luo et al. [2008]. The integrodifferential terms result from distributions of exchange rates.

$$\frac{\partial C_1}{\partial t} + \beta \int_0^\infty \left[R_1 \frac{\partial S_1(\alpha)}{\partial t} + \lambda_1^a S_1(\alpha) \right] p(\alpha) d\alpha + \lambda_1^i C_1 = -\frac{Q}{A} \frac{\partial C_1}{\partial x} + \frac{1}{A} \frac{\partial}{\partial x} \left(AD \frac{\partial C_1}{\partial x} \right), \quad (1)$$

$$R_1 \frac{\partial S_1(\alpha)}{\partial t} = \alpha(C_1 - S_1(\alpha)) - \lambda_1^a S_1(\alpha), \quad 0 < \alpha < \infty, \quad (2)$$

$$\frac{\partial C_2}{\partial t} + \beta \int_0^\infty \left[R_2 \frac{\partial S_2(\alpha)}{\partial t} + \lambda_2^a S_2(\alpha) - \lambda_1^a y^a S_1(\alpha) \right] p(\alpha) d\alpha + \lambda_2^i C_2 - \lambda_1^i y^i C_1 = -\frac{Q}{A} \frac{\partial C_2}{\partial x} + \frac{1}{A} \frac{\partial}{\partial x} \left(AD \frac{\partial C_2}{\partial x} \right), \quad (3)$$

$$R_2 \frac{\partial S_2(\alpha)}{\partial t} = \alpha(C_2 - S_2(\alpha)) - \lambda_2^a S_2(\alpha) + \lambda_1^a y^a S_1(\alpha), \quad 0 < \alpha < \infty, \quad (4)$$

where subscripts 1 and 2 indicate parent and modeled daughter, C_1 is stream concentration of the parent (mol L^{-1}), C_2 is stream concentration of the daughter (mol L^{-1}), $S_1(\alpha)$ is storage domain concentration (mol L^{-1}) of the parent, $S_2(\alpha)$ is storage domain concentration (mol L^{-1}) of the

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daughter, superscript “a” refers to a reaction rate associated with the storage domain (relatively “active”), superscript “i” refers to a reaction rate associated with the stream (relatively “inactive”), λ_1^a is the rate coefficient for the decay of parent to all daughters (h^{-1}), λ_2^a is the modeled daughter reaction rate coefficient (h^{-1}), γ is the conversion factor between parent and modeled daughter (–), p is the probability density function of mass transfer exchange rates (h), β is the ratio of storage to stream mass at equilibrium (–), R is retardation due to sorption (–) with subscripts indicating parent and daughter, D is the dispersion coefficient ($\text{m}^2 \text{h}^{-1}$), Q is discharge ($\text{m}^3 \text{h}^{-1}$), and A is cross-sectional area of the stream (m^2). If γ^a or $\gamma^i = 1$, then parent is converted only to the modeled daughter, whereas if γ^a or $\gamma^i < 1$, some parent is simultaneously converted to another daughter that is not modeled. The values p , S_1 , and S_2 are functions of α ; however, we subsequently drop the functional notation. Here, it is assumed that the reaction only happens in the aqueous phase, a valid assumption for biological reaction [McMurry, 2010, p. 210] but not for radioactive decay. Radioactive decay would require retardation factors in the decay terms above.

[5] During a constant injection to plateau concentrations, the equations eventually simplify to steady state after plateau is reached:

$$C_1 \lambda_1^i + \beta \int_0^\infty p \lambda_1^a S_1 d\alpha = -\frac{Q}{A} \frac{\partial C_1}{\partial x} + \frac{1}{A} \frac{\partial}{\partial x} \left(AD \frac{\partial C_1}{\partial x} \right), \quad (5)$$

$$S_1 = \frac{\alpha}{\alpha + \lambda_1^a} C_1, \quad (6)$$

$$C_2 \lambda_2^i + \beta \int_0^\infty p (\lambda_2^a S_2 - \lambda_1^a \gamma^a S_1) d\alpha - \lambda_1^i \gamma^i C_1 = -\frac{Q}{A} \frac{\partial C_2}{\partial x} + \frac{1}{A} \frac{\partial}{\partial x} \left(AD \frac{\partial C_2}{\partial x} \right), \quad (7)$$

$$S_2 = \frac{\alpha}{\alpha + \lambda_2^a} C_2 + \frac{\lambda_1^a \gamma^a}{\alpha + \lambda_2^a} S_1. \quad (8)$$

Using a first-type upstream boundary condition ($C_1(x=0) = C_{1,b}$, $C_2(x=0) = C_{2,b}$) and concentration of 0 at infinite distance, the after appropriate substitutions are as follows:

$$C_2 \lambda_2^i + \beta \int_0^\infty p \lambda_2^a \left[\frac{\alpha}{\alpha + \lambda_2^a} C_2 + \frac{\lambda_1^a \gamma^a \alpha}{(\alpha + \lambda_2^a)(\alpha + \lambda_1^a)} C_1 \right] d\alpha + \left[\beta \int_0^\infty \frac{p \lambda_1^a \gamma^a}{\alpha + \lambda_1^a} d\alpha - \lambda_1^i \gamma^i \right] C_1 = -\frac{Q}{A} \frac{\partial C_2}{\partial x} + \frac{1}{A} \frac{\partial}{\partial x} \left(AD \frac{\partial C_2}{\partial x} \right), \quad (10)$$

[6] Defining three effective reaction rate functions,

$$f_1 = \lambda_1^i + \lambda_1^a \beta \int_0^\infty \frac{p \alpha}{\alpha + \lambda_1^a} d\alpha, \quad (11)$$

(the effective rate of decay of C_1),

$$f_2 = \lambda_2^i + \lambda_2^a \beta \int_0^\infty \frac{p \alpha}{\alpha + \lambda_2^a} d\alpha, \quad (12)$$

(the effective rate of decay of C_2), and

$$f_3 = \beta \lambda_2^a \lambda_1^a \gamma^a \int_0^\infty \frac{p \alpha}{(\alpha + \lambda_2^a)(\alpha + \lambda_1^a)} d\alpha - \lambda_1^i \gamma^i - \beta \lambda_1^a \gamma^a \int_0^\infty \frac{p \alpha}{\alpha + \lambda_1^a} d\alpha \quad (13)$$

(the effective rate of production of C_2 from C_1), equation (10) can be rewritten as follows:

$$f_2 C_2 + f_3 C_1 = -\frac{Q}{A} \frac{\partial C_2}{\partial x} + \frac{1}{A} \frac{\partial}{\partial x} \left(AD \frac{\partial C_2}{\partial x} \right). \quad (14)$$

[7] The velocity is $v = Q/A$, the Peclet number is $Pe = vL/D$, L is the downstream distance to the end of the reach. All parameters are assumed to be uniform, although solution could be accomplished with parameters that vary in space using separation of variables. Results would be similar to ours except that a number of parameters would be defined as integrals over space and that the validity of $Pe \gg 1$ would need careful evaluation.

[8] The solution for C_1 (in equation (5)) is identical to solutions that are well known because the steady-state equation is the same as the Laplace-transformed transient equation [cf., Valocchi, 1985] and because the distribution of mass transfer rates is not a function of space:

$$C_1 = C_{1,b} \exp[m(f_1)], \quad (15)$$

where

$$m(f) = \frac{Pe \chi}{2} \left(1 - \sqrt{1 + \frac{4\tau f}{Pe \chi}} \right), \quad (16)$$

$$\tau = \frac{x}{v}, \quad (17)$$

$$\chi = \frac{x}{L}. \quad (18)$$

Observation at x is assumed, and $\chi = 1$ if $x = L$. The solution to C_2 is as follows:

$$C_2 = C_{2,b} \exp[m(f_2)] + C_{1,b} f_3 \left[\frac{\tau Pe \chi (\exp[m(f_2)] - \exp[m(f_1)])}{\tau Pe \chi f_2 + [Pe \chi - m(f_1)]m(f_1)} \right]. \quad (19)$$

[9] The ratio of C_2 to C_1 is given by the following equation:

$$\frac{C_2}{C_1} = \frac{C_{2,b}}{C_{1,b}} \exp[m(f_2) - m(f_1)] + \frac{\tau Pe \chi f_3 \{\exp[m(f_2) - m(f_1)] - 1\}}{m(f_1)(Pe \chi - m(f_1)) + \tau Pe \chi f_2}. \quad (20)$$

[10] The structure of (20) can be seen more clearly if we assume $Pe \gg 1$:

$$\frac{C_2}{C_1} = \frac{C_{2,b}}{C_{1,b}} \exp[\tau(f_1 - f_2)] + \frac{f_3}{f_2 - f_1} \{\exp[\tau(f_1 - f_2)] - 1\}, \quad (21a)$$

or

$$\ln\left(\frac{C_2}{C_1} + \frac{f_3}{f_2 - f_1}\right) = (f_1 - f_2)\tau + \ln\left(\frac{C_{2,b}}{C_{1,b}} + \frac{f_3}{f_2 - f_1}\right) \quad (21b)$$

3. Application to Resazurin

[11] These relationships are now used for the resazurin-resorufin (Raz-Rru) tracer system, which is sensitive to water column-bed exchange and respiration [Argerich *et al.*, 2011; González-Pinzón *et al.*, 2012]. Raz transforms to Rru almost exclusively at the bed or in the sediment [Haggerty *et al.*, 2008], which means that λ_1^i and λ_2^i can be neglected. The ratio of effective rate functions simplifies tremendously and becomes a function only of reaction parameters:

$$\frac{f_3}{f_2 - f_1} = \frac{y^a \lambda_1^a}{\lambda_1^a - \lambda_2^a} \equiv P. \quad (22)$$

This ratio can be thought of as the rate of production of C_2 relative to the rate of decay of C_1 , which we will call the production-decay ratio, P . If reaction is isolated to the storage zones, P is independent of the mass transfer rate and relative quantity of storage zone. The effects of mass transfer are isolated within two effective decay coefficients as follows:

$$\lambda_{1e} = \int_0^\infty \frac{\alpha \lambda_1^a p}{\alpha + \lambda_1^a} d\alpha \quad \text{and} \quad \lambda_{2e} = \int_0^\infty \frac{\alpha \lambda_2^a p}{\alpha + \lambda_2^a} d\alpha, \quad (23)$$

Furthermore, for $Pe/\beta\tau\lambda_{e1} \gg 1$ and $Pe/\beta\tau\lambda_{e2} \gg 1$ (see Cho [1971] and Burnell *et al.* [2012]—greater than about 6 is sufficient), the equations can be simplified. Equation (21b) therefore becomes:

$$\ln\left(\frac{C_2}{C_1} + P\right) = \beta(\lambda_{1e} - \lambda_{2e})\tau + \ln\left(\frac{C_{2,b}}{C_{1,b}} + P\right). \quad (24)$$

[12] Equation (24) is very useful. In a stream, the mass transfer effects (e.g., bed exchange, resulting in respiration) are confined to the slope of the line, $\beta(\lambda_{1e} - \lambda_{2e})$. When used with Raz-Rru, the slope of the line is therefore an integrated measure of bed exchange and ecosystem respiration. Although the details of bed exchange may contain many

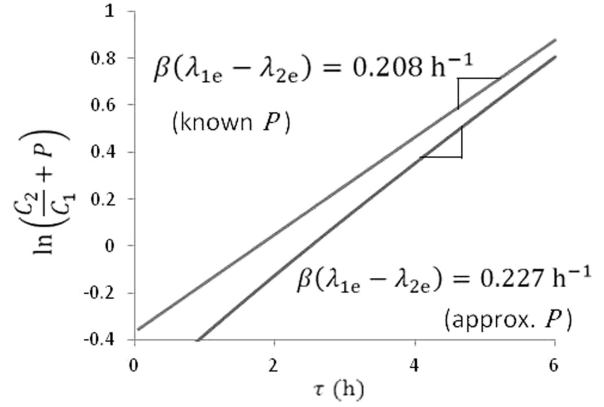


Figure 1. Plots of equation (24) using a known and an approximate value of P . The known (true) value of P is 0.667, and the approximated value of P is 0.5. The approximation error is 33% but the $\beta(\lambda_{1e} - \lambda_{2e})$ slope error is about 9%. The lines were generated from a full simulation of the advection-dispersion-mass transfer reaction equations with parameters $D = 3600 \text{ m}^2 \text{ h}^{-1}$; $v = 250 \text{ m h}^{-1}$; $\beta = 0.3$; $\lambda_1^i = 9.89 \times 10^{-4} \text{ h}^{-1}$; $\lambda_2^i = 1.75 \times 10^{-3} \text{ h}^{-1}$; $\lambda_1^a = 1 \text{ h}^{-1}$; $\lambda_2^a = 0.1 \text{ h}^{-1}$; $y^i = 1$; $y^a = 0.6$; and constant A . The distribution of rate coefficients was single rate, i.e., $p(\alpha) = \delta(\alpha)$ with $\alpha = 4 \text{ h}^{-1}$. Injection was constant for 20 h and $C_{2,b}/C_{1,b} = 0.025$. The reaction and mass transfer parameters yield effective reaction rates $\lambda_{1e} = 0.8 \text{ h}^{-1}$ and $\lambda_{2e} = 0.098 \text{ h}^{-1}$. The correct value of $\beta(\lambda_{1e} - \lambda_{2e})$ from the simulation is 0.211 h^{-1} .

different rates and volumes which may be nonunique, the slope of the line can be compared from one location to another or from one time to another.

[13] The production-decay ratio P is often known or can be approximated with little consequence. For example, the parent may decay only to the daughter, and the daughter may be stable, in which case $P = 1$. If the parent decays to other daughters at the same rate as the daughter decays, $P = 1$. For the Raz-Rru system, our work to date suggests that P is of order 0.3 – 0.8 [Haggerty *et al.*, 2008, 2009; Argerich *et al.*, 2011; and data in preparation for publication]. If the estimate of P is incorrect, (24) is nonlinear. For this reason, large errors in P will generate a nonlinear graph and large errors will therefore be obvious. However, small errors in P can result in nearly linear graphs and therefore generate errors in the slope of the line that are not obvious.

[14] To check if the slope of the line (24) recovers the correct value of $\beta(\lambda_{1e} - \lambda_{2e})$, the full transport equations (1)–(4) were solved using STAMMT-L [Haggerty and Reeves, 2002]. Figure 1 shows the full results, using the known value $P = 0.667$ (blue) and using an incorrect value $P = 0.5$ (red). When P was known, the slope of the line $\beta(\lambda_{1e} - \lambda_{2e})$ was almost exactly correct. The correct value of the slope was 0.211, and the small error was due to the approximations of zero dispersion and no reaction in the water column in the graphed function.

[15] Using (24) with field data from an experiment reported by Argerich *et al.* [2011] gives results shown in Figure 2. Briefly, Raz was injected on 17–18 September 2007 to steady-state concentration in the bedrock reach of watershed 3 at the H. J. Andrews Experimental Forest,

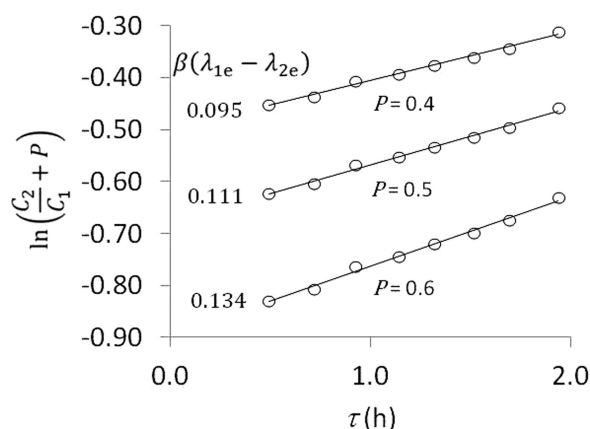


Figure 2. Plot of equation (24) for field results given in *Argerich et al.* [2011]. The three sets of markers result from using different assumed values of the production decay ratio P . The resulting line slopes are shown in the figure. From a full simulation of transport [see *Argerich et al.*, 2011], the slope of the line should be 0.105 h^{-1} .

Oregon. Concentrations were measured at eight locations in the bedrock reach over a distance of 256.5 m. The average velocity of water in the reach was 132.5 m h^{-1} . As perfect knowledge of P did not exist, the results were plotted for three different values: $P = 0.4, 0.5$, and 0.6 . This generates a slope of $\beta(\lambda_{1e} - \lambda_{2e}) = 0.095$ to 0.134 h^{-1} *Argerich et al.* used a temporal moments method to calculate the value of 0.105 h^{-1} .

[16] The results in Figures 1 and 2 suggest that the slope of $\ln(C_2/C_1 + P)$ indicates or at least qualifies the amount respiration and the amount of interaction between the water column and the bed. Furthermore, $\ln(C_2/C_1 + P)$ is easy to graph and thus different sites or conditions can be compared without numerical modeling. Depending on the assumptions that can be made and the purpose of the experiment, the results could be sufficiently accurate that no other modeling is necessary. Of course, the estimate could always be improved with more complete use of the data and numerical modeling.

[17] The results of this note are applicable to any tracer or compound during steady-state transport that is undergoing first-order decay in the aqueous phase to a second compound. An example of a stream tracer, resazurin, was given. The results are also applicable to radionuclides, provided that they are weakly sorbing so that most of the decay happens in the aqueous phase. The results are also applicable to contaminants in groundwater.

Notation

- A cross-sectional area of the stream (m^2).
- C_1 stream concentration of the parent solute (mol L^{-1}).
- $C_{1,b}$ upstream boundary condition for parent solute (mol L^{-1}).
- C_2 stream concentration of the daughter solute (mol L^{-1}).
- $C_{2,b}$ upstream boundary condition for daughter solute (mol L^{-1}).
- D dispersion coefficient ($\text{m}^2 \text{ h}^{-1}$).

- f_1 effective decay rate function for parent solute (h^{-1}).
- f_2 effective decay rate function for daughter solute (h^{-1}).
- f_3 effective production rate function for daughter solute (h^{-1}).
- L reach length (L).
- $m(f)$ function (see (16) for definition) (–).
- $p(\alpha)$ probability density function of mass transfer exchange rates (h).
- P production-decay ratio (see equation (22)) (–).
- Pe Peclet number (–).
- Q discharge in the stream ($\text{m}^3 \text{ s}^{-1}$).
- R_1 retardation factor for parent solute due to sorption (–).
- R_2 retardation factor for daughter solute due to sorption (–).
- $S_1(\alpha)$ storage domain concentration of the parent solute as a function of α (mol L^{-1}).
- $S_2(\alpha)$ storage domain concentration of the daughter solute as a function of α (mol L^{-1}).
- t time (h).
- v velocity of the stream (m h^{-1}).
- x spatial coordinate (L).
- y^a conversion factor between parent and modeled daughter in the storage zone (–).
- y^i conversion factor between parent and modeled daughter in the water column (–).
- α first-order rate coefficient for exchange between the stream and the storage zone, and equal to the inverse of the mean residence time in the storage zone (h^{-1}).
- β ratio of storage to stream mass at equilibrium, and equal to A_s/A as defined in most transient storage literature (–).
- λ_1^a rate coefficient for the decay of parent to all daughters in the storage zone (h^{-1}).
- λ_1^i rate coefficient for the decay of parent to all daughters in the water column (h^{-1}).
- λ_{1e} effective decay coefficient for the parent solute (h^{-1}).
- λ_2^a rate coefficient for the decay of daughter solute in the storage zone (h^{-1}).
- λ_2^i rate coefficient for the decay of daughter solute in the water column (h^{-1}).
- λ_{2e} effective decay coefficient for the daughter solute (h^{-1}).
- τ advective travel time (h).
- χ dimensionless distance defined in (18) (–).

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