Transit-time and age distributions for nonlinear time-dependent compartmental systems

Holger Metzler[a,1], Markus Müller[a], and Carlos A. Sierra[a]

[a]Theoretical Ecosystem Ecology, Department of Biogeochemical Processes, Max Planck Institute for Biogeochemistry, 07745 Jena, Germany

Edited by Steven C. Wofsy, Harvard University, Cambridge, MA, and approved December 15, 2017 (received for review May 4, 2017)

Many processes in nature are modeled using compartmental systems (reservoir/pool/box systems). Usually, they are expressed as a set of first-order differential equations describing the transfer of matter across a network of compartments. The concepts of age of matter in compartments and the time required for particles to transit the system are important diagnostics of these models with applications to a wide range of scientific questions. Until now, explicit formulas for transit-time and age distributions of nonlinear time-dependent compartmental systems were not available. We compute densities for these types of systems under the assumption of well-mixed compartments. Assuming that a solution of the nonlinear system is available at least numerically, we show how to construct a linear time-dependent system with the same solution trajectory. We demonstrate how to exploit this solution to compute transit-time and age distributions in dependence on given start values and initial age distributions. Furthermore, we derive equations for the time evolution of quantiles and moments of the age distributions. Our results generalize available density formulas for the linear time-independent case and mean-age formulas for the linear time-dependent case. As an example, we apply our formulas to a nonlinear and a linear version of a simple global carbon cycle model driven by a time-dependent input signal which represents fossil fuel additions. We derive time-dependent age distributions for all compartments and calculate the time it takes to remove fossil carbon in a business-as-usual scenario.

transit time | age distribution | compartmental system | nonlinear | nonautonomous systems

Compartmental systems are used in a large variety of scientific fields such as systems biology, toxicology, ecology, hydrology, and biogeochemistry (1–6). The dynamics of such systems can be better understood if it is known how long particles need to travel through the system (transit time), how old particles in a specific compartment are (compartment age), and how old the particles in the system are (system age) (7, 8).

A first attempt to compute age structures of compartmental systems was to establish density formulas in dependence on a not explicitly known system response function (9). A similar response function approach was further used to derive explicit formulas for models with very simple structure (10) and to obtain age densities by long-term numerical simulations (11). Only recently, explicit density formulas have been derived by a probabilistic approach (12). Most importantly, all these approaches were concerned with linear time-independent models in steady state. This restriction is often very unrealistic since most systems in nature are intrinsically nonlinear and influenced by time-depending factors (e.g., a fluctuating external environment). For such systems out of steady state, formulas have been developed for one-compartment hydrological systems, without expanding the theory to networks of multiple interconnected compartments (13–15). A first milestone in this direction was the mean age system (16), a system of linear ordinary differential equations (linear ODEs) describing the time evolution of mean compartment ages of linear systems with time-dependent coefficients.

In this article, we derive formulas not only for means, but also for entire densities of transit time, compartment ages, and system age of time-dependent models. Moreover, our approach works even for nonlinear models. We further extend the mean age system to higher-order moments. This allows a simple computation also of the variance and the SD. Additionally, we provide ODEs to describe the time evolution of quantiles such as the median of age distributions. This framework results in much faster computations of entire age distributions than what was possible before. These results generalize many earlier results from different scientific fields such as atmospheric sciences, ecology, and hydrology. In *S6. The Relations to Earlier Results and Possible Applications*, we explain in detail how our framework relates to these fields.

As an example application of our theoretical results, we apply them to a simple global carbon cycle model and address the following questions: How old is atmospheric carbon? How long will a significant fraction of a pulse of fossil fuel carbon, emitted to the atmosphere today, remain in the system? We compare transit times and ages of a nonlinear and a linear version of the model and highlight significant differences in their age structure, which were not possible to characterize by the mean ages alone.

1. Well-Mixed Compartmental Systems

Following ref. 6, a compartment of a well-mixed system is an amount of kinetically homogeneous material. Kinetically homogeneous means that any material entering the system is

Significance

What is the distribution of the time required for particles to transit a compartmental system? What is the age distribution of particles in the system? These questions are important to understand the dynamics of modeled processes in many scientific fields. Until now, they could be answered only for systems in steady state, but the steady-state assumption is very restrictive and almost always unreasonable. Nonlinear relations between compartments and changing environmental factors keep systems permanently changing. A theory is presented here to describe the time evolution of age densities under changing environmental influences in nonlinear systems. An application is provided, addressing questions of high scientific and societal interest such as the lifetime of fossil fuel-derived atmospheric carbon.

Author contributions: C.A.S. designed research; H.M. and M.M. performed research; H.M. and M.M. programed the Python package; C.A.S. designed example application; and H.M. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

Data deposition: Jupyter (Python) notebook was compiled to HTML to reproduce the example from this paper, which is available at https://github.com/MPIBGC-TEE/CompartmentalSystems/blob/master/notebooks/PNAS. A python package that implements the proposed mathematical framework is available at https://github.com/MPIBGC-TEE/CompartmentalSystems.

[1]To whom correspondence should be addressed. Email: hmetzler@bgc-jena.mpg.de.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1705296115/-/DCSupplemental.

immediately mixed with the material of the compartment. A well-mixed compartmental system is then a collection of d compartments and is usually described by a d-dimensional system of first-order differential equations. We fix an initial time $t_0 \in \mathbb{R}$ and a finite time horizon $T > t_0$ and consider the d-dimensional initial value problem

$$\begin{aligned} \frac{d}{dt}\mathbf{x}(t) &= \mathrm{B}(\mathbf{x}(t), t)\,\mathbf{x}(t) + \mathbf{u}(t), \quad t > t_0, \\ \mathbf{x}(t_0) &= \mathbf{x^0}. \end{aligned} \qquad \textbf{[1]}$$

Here, $\mathbf{x}(t) \in \mathbb{R}^d$ is the vector of compartment contents at time t, $\mathrm{B} = (B_{ij})_{i,j=1,2,\ldots,d}$ is a matrix-valued function depending on the current system content and time, $\mathbf{u}$ is a vector-valued function depending on time, and $\mathbf{x^0} \in \mathbb{R}^d$ is the initial system content at time t_0. Throughout this paper, we assume all involved functions to be sufficiently smooth. In particular, Eq. **1** is supposed to admit a unique solution on $[t_0, T]$.

Eq. **1** describes how mass flows into the system through the vector-valued input function $\mathbf{u}$ and is then cycled by the matrix-valued function B until it eventually leaves the system. Consequently, $\mathbf{u}$ and $\mathbf{x^0}$ are nonnegative and the system must obey the law of mass conservation. This sets the additional restriction on B to be a compartmental matrix for all $\mathbf{x}$ and t.

A quadratic matrix is called compartmental if all its diagonal entries are nonpositive, all its off-diagonal entries are nonnegative, and all its column sums are nonpositive (6).

A. Classification of Compartmental Systems. It is important to note that, in a general compartmental system, B may depend on the system state $\mathbf{x}(t)$ as well as on time t. This makes the system in general nonlinear and time dependent. If $\mathrm{B}(\mathbf{x}(t), t) = \mathrm{B}(t)$, i.e., B is independent of the system state, then the system is called linear. If $\mathrm{B}(\mathbf{x}(t), t) = \mathrm{B}(\mathbf{x}(t))$ and $\mathbf{u}(t) = \mathbf{u}$, i.e., B and $\mathbf{u}$ do not explicitly depend on time t, then the system is called time independent.

B. Linear Interpretation of the Nonlinear Solution. Only in special cases can we find an analytical solution to Eq. **1**. Nevertheless, we assume to know the unique solution at least numerically and denote it by $\mathbf{x}$. We define a time-dependent and matrix-valued function $\tilde{\mathrm{B}}$ by plugging the solution into B; i.e., $\tilde{\mathrm{B}}(t) := \mathrm{B}(\mathbf{x}(t), t)$. The linear time-dependent compartmental system

$$\begin{aligned} \frac{d}{dt}\mathbf{y}(t) &= \tilde{\mathrm{B}}(t)\,\mathbf{y}(t) + \mathbf{u}(t), \quad t > t_0, \\ \mathbf{y}(t_0) &= \mathbf{x^0}, \end{aligned} \qquad \textbf{[2]}$$

has a unique solution $\mathbf{y}$. Since $\mathbf{x}$ is the unique solution to system **1** and both systems are equivalent, $\mathbf{y} = \mathbf{x}$. Below we consider linear systems only, because we can always think of the solution of the nonlinear system **1** as being the solution of the equivalent linear system **2**. This linear interpretation of $\mathbf{x}$ allows us to derive semianalytical formulas for many properties of nonlinear systems. The prefix "semi" reflects here the fact that all of the theory works under the assumption that $\mathbf{x}$ is already known.

C. General Solution of the Linear System. We consider the linear time-dependent compartmental system

$$\begin{aligned} \frac{d}{dt}\mathbf{x}(t) &= \mathrm{B}(t)\,\mathbf{x}(t) + \mathbf{u}(t), \quad t > t_0, \\ \mathbf{x}(t_0) &= \mathbf{x^0}. \end{aligned} \qquad \textbf{[3]}$$

The unique solution $\mathbf{x}$ to this system on $[0, T]$ is given by

$$\mathbf{x}(t) = \Phi(t, t_0)\,\mathbf{x^0} + \int_{t_0}^{t} \Phi(t, \tau)\,\mathbf{u}(\tau)\,d\tau, \qquad \textbf{[4]}$$

where Φ denotes the state transition matrix (*S1. The State Transition Matrix* Φ). This state transition matrix describes the transport of particles through the system and is a generalized Green's function. Since the system is time dependent, Φ depends on two time variables, and since Φ is matrix valued, it maps an input vector to an output vector. In particular, if $\mathbf{v} := \Phi(t, \tau)\,\mathbf{u}$, then $\mathbf{v}$ is the vector that describes the time-t positions of the particles that had positions according to $\mathbf{u}$ at time τ.

From Eq. **4** we see that the vector of compartment contents at time t is given as the sum of two terms. The first term, $\Phi(t, t_0)\,\mathbf{x^0}$, describes how much mass has remained from the initial contents, whereas the second term, $\int_{t_0}^{t} \Phi(t, \tau)\,\mathbf{u}(\tau)\,d\tau$, describes how much has remained until time t of inputs that came later than t_0. In particular, $\Phi(t, \tau)\,\mathbf{u}(\tau)\,d\tau$ describes the mass that entered the system in the infinitesimal time interval $d\tau$ and is still in the system at time t. Consequently, at time t the mass $\Phi(t, \tau)\,\mathbf{u}(\tau)\,d\tau$ has age $t - \tau$.

2. Age Distributions

A. Compartment Age Densities. We now assume that the initial content $\mathbf{x^0}$ has a given age density $\mathbf{p^0}$ such that $\mathbf{x^0} = \int_0^\infty \mathbf{p^0}(a)\,da$, where $\mathbf{p^0}(a)\,da$ is the vector with nonnegative components of mass with age infinitesimally close to a at time t_0. The previous observation on the age of $\Phi(t, \tau)\,\mathbf{u}(\tau)\,d\tau$ motivates the conjecture that the age density of the compartment contents at age $a \geq 0$ and time $t \geq t_0$ is given by

$$\mathbf{p}(a, t) = \mathbf{g}(a, t) + \mathbf{h}(a, t), \qquad \textbf{[5]}$$

where

$$\mathbf{g}(a, t) = \mathbb{1}_{[t-t_0,\infty)}(a)\,\Phi(t, t_0)\,\mathbf{p^0}(a - (t - t_0)) \qquad \textbf{[6]}$$

is the age density of the mass that has been in the system from the beginning, and

$$\mathbf{h}(a, t) = \mathbb{1}_{[0,t-t_0)}(a)\,\Phi(t, t - a)\,\mathbf{u}(t - a) \qquad \textbf{[7]}$$

is the age density of the mass that has entered the system after t_0. The indicator function $\mathbb{1}_S(a)$ of a set S equals 1 if $a \in S$; otherwise it equals 0.

It turns out that $\mathbf{p}$ satisfies indeed a multidimensional version of the well-known McKendrick–von Foerster equation (17, 18) that describes the evolution of the age structure of system **1**, since (*S2. The McKendrick–von Foerster Equation*), for $a > 0$ and $t > t_0$,

$$\left(\frac{\partial}{\partial a} + \frac{\partial}{\partial t}\right)\mathbf{p}(a, t) = \mathrm{B}(t)\,\mathbf{p}(a, t), \qquad \textbf{[8]}$$

with boundary condition

$$\mathbf{p}(0, t) = \mathbf{u}(t), \quad t > t_0, \qquad \textbf{[9]}$$

and initial condition

$$\mathbf{p}(a, t_0) = \mathbf{p^0}(a), \quad a \geq 0. \qquad \textbf{[10]}$$

B. System Age Density. The age density of the entire system is just the sum of the compartment age densities, and we denote it by

$$\|\mathbf{p}(a, t)\| := \sum_{i=1}^{d} p_i(a, t), \quad a \geq 0, \quad t \geq t_0. \qquad \textbf{[11]}$$

We can interpret $\|\cdot\|$ as a norm here, since $\mathbf{p}(a, t)$ is a vector with nonnegative entries $p_i(a, t)$ only.

C. Cumulative Compartment Age Distribution. We denote by uppercase letters the cumulative age distributions corresponding to age densities. This means for the initial age density $\mathbf{p^0}$ and

$\xi \geq 0$ that $\mathbf{P}^{\mathbf{0}}(\xi) = \int_0^{\xi} \mathbf{p}^{\mathbf{0}}(a)\, da$ is the vector of initial compartment contents with age $a \leq \xi$. Then, by Eq. **5**,

$$\mathbf{P}(\xi, t) = \mathbf{G}(\xi, t) + \mathbf{H}(\xi, t), \qquad \textbf{[12]}$$

where

$$\mathbf{G}(\xi, t) = \Phi(t, t_0)\, \mathbf{P}^{\mathbf{0}}(\xi - (t - t_0)), \quad \xi \geq t - t_0, \qquad \textbf{[13]}$$

is the vector of compartment contents with age $a \leq \xi$ at time t that have been in the system from the beginning, and

$$\mathbf{H}(\xi, t) = \int\limits_{\max\{t-\xi, t_0\}}^{t} \Phi(t, \tau)\, \mathbf{u}(\tau)\, d\tau = \mathbf{x}(t) - \Phi(t, t-\xi)\, \mathbf{x}(t-\xi) \qquad \textbf{[14]}$$

is the vector of compartment contents that came into the system after t_0 and have age $a \leq \xi$ at time t. The latter can also be expressed as the compartment contents at time t minus all of the mass that was already in the system at time $t - \xi$ and survived until time t.

D. Cumulative System Age Distribution. The mass in the system with age $a \leq \xi$ at time $t \geq t_0$ is given by $\|\mathbf{P}(\xi, t)\|$.

3. Moments of Age Distributions

For any nonnegative integer k and any age density $\mathbf{p}$ of a nonnegative vector $\mathbf{x} \in \mathbb{R}^d$, we define

$$\bar{\mathbf{a}}^{\mathbf{x},k} := \mathrm{X}^{-1} \int\limits_0^{\infty} a^k\, \mathbf{p}(a)\, da \qquad \textbf{[15]}$$

to be the kth moment of the density $\mathbf{p}$, where $\mathrm{X} = \mathrm{diag}(x_1, x_2, \ldots, x_d)$ is a diagonal matrix. Note that $\bar{\mathbf{a}}^{\mathbf{x},0} = \mathbf{1}$, the vector comprising ones. For $k = 1$ we obtain the mean age vector. The unboundedness of the upper limit of the integral causes issues in the numerical computation of an age moment directly from Eq. **15**.

A. Moments of Compartment Ages.

A.1. Semiexplicit formula for compartment age moments. To circumvent this problem, we can use the McKendrick–von Foerster Eq. **8** to compute (*S3. The Semiexplicit Formula for Compartment Age Moments*) the nth moment $\bar{\mathbf{a}}^n(t) := \bar{\mathbf{a}}^{\mathbf{x}(t),n}$ of the age distribution of the compartmental system at time t by

$$\begin{aligned}\bar{\mathbf{a}}^n(t) &= \mathrm{X}(t)^{-1} \\ &\times \left[\sum_{k=0}^{n} \binom{n}{k} (t - t_0)^{n-k}\, \Phi(t, t_0)\, \mathrm{X}^0\, \bar{\mathbf{a}}^{0,k} \right. \\ &\left. + \int\limits_0^{t-t_0} a^n\, \Phi(t, t-a)\, \mathbf{u}(t-a)\, da \right].\end{aligned} \qquad \textbf{[16]}$$

Here, $\mathrm{X}(t) = \mathrm{diag}(x_1, x_2, \ldots, x_d)(t)$ is the diagonal matrix containing the compartment contents at time t, $\mathrm{X}^0 = \mathrm{X}(t_0)$, and $\bar{\mathbf{a}}^{0,k}$ for $k = 1, 2, \ldots, n$ denote the moments of the initial age distribution. Note that the integral involved is now over the half-open but finite interval $[0, t - t_0)$.

A.2. Compartment age moment system. Another way to compute the age moments is to set up and solve an appropriate system of first-order differential equations which we call the compartment age moment system. This system is a straightforward $d \cdot (n+1)$-dimensional generalization of the mean age system derived in ref. 16. It is given by (*S4. The Compartment Age Moment System*)

$$\frac{d}{dt} \begin{pmatrix} \mathbf{x} \\ \bar{\mathbf{a}}^1 \\ \vdots \\ \bar{\mathbf{a}}^n \end{pmatrix} (t) = \begin{pmatrix} \mathrm{B}(t)\, \mathbf{x} + \mathbf{u}(t) \\ \boldsymbol{\gamma}^1(t, \mathbf{x}, \mathbf{1}, \bar{\mathbf{a}}^1) \\ \vdots \\ \boldsymbol{\gamma}^n(t, \mathbf{x}, \bar{\mathbf{a}}^{n-1}, \bar{\mathbf{a}}^n) \end{pmatrix}, \quad t > t_0, \qquad \textbf{[17]}$$
$$(\mathbf{x}, \bar{\mathbf{a}}^1, \ldots, \bar{\mathbf{a}}^n)(t_0) = (\mathbf{x}^{\mathbf{0}}, \bar{\mathbf{a}}^{0,1}, \bar{\mathbf{a}}^{0,2}, \ldots, \bar{\mathbf{a}}^{0,n}),$$

where, for $k = 1, 2, \ldots, n$, $\boldsymbol{\gamma}^k = \left(\gamma_1^k, \gamma_2^k, \ldots, \gamma_d^k\right)^T$ and for $i = 1, 2, \ldots, d$,

$$\begin{aligned}\gamma_i^k(t, \mathbf{x}, \bar{\mathbf{a}}^{k-1}, \bar{\mathbf{a}}^k) &= k\, \bar{a}_i^{k-1} \\ &+ \frac{1}{x_i} \left[\sum_{j=1}^{d} B_{ij}\, x_j\, \left(\bar{a}_j^k - \bar{a}_i^k\right) - \bar{a}_i^k\, u_i \right].\end{aligned} \qquad \textbf{[18]}$$

Note that we occasionally omitted the time dependencies to simplify notation and that $\mathbf{v}^T$ denotes the transpose of the vector $\mathbf{v}$. Because of its particular structure, the compartment age moment system **17** has the advantage of solving the compartments' age moments through time alongside the compartments' contents. This procedure is both fast and numerically robust.

B. System Age Moment. The nth moment of the system age at time $t \geq t_0$ is defined by

$$\bar{A}^n(t) := \frac{1}{\|\mathbf{x}(t)\|} \int\limits_0^{\infty} a^n\, \|\mathbf{p}(a, t)\|\, da. \qquad \textbf{[19]}$$

A straightforward calculation shows $\bar{A}^n(t) = \frac{\mathbf{x}^T(t)\, \bar{\mathbf{a}}^n(t)}{\|\mathbf{x}(t)\|}$.

4. Age Quantiles

In addition to moments, quantiles of the age distributions are important statistics. As a special case, the unique age 2 quantile is the median of the age distribution. Let k and n be positive integers such that $k < n$ and define $q := k/n$.

A. Compartment Age Quantiles. For $i \in \{1, 2, \ldots, d\}$ the kth n quantile of the age of compartment i at time $t \geq t_0$ is defined as $\xi_i(t)$ such that

$$P_i(\xi_i(t), t) = q\, x_i(t). \qquad \textbf{[20]}$$

In general, the computation of the quantile relies on the computationally expensive inverse of the cumulative age distribution. It

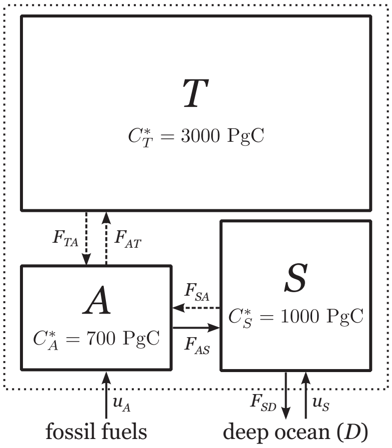

Fig. 1. Simple global carbon cycle model with three compartments (solid boxes within dashed square): atmosphere (*A*), terrestrial biosphere (*T*), and surface ocean (*S*). The indicated carbon contents are the respective steady-state values. External to the modeled system are fossil fuel sources and the deep ocean (*D*). The model compartments and the external sources are connected by linear (solid arrows) and possibly nonlinear (dashed arrows) fluxes of carbon.

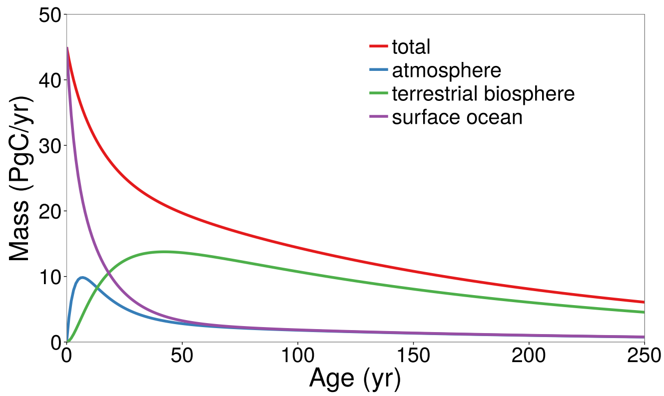

Fig. 2. Preindustrial age densities of the three compartments: atmosphere (blue), terrestrial biosphere (green), and surface ocean (purple). The red curve shows the age density of the entire system.

is numerically much faster and easier to instead use the fact (*S5. The Age Quantiles*) that ξ_i solves the initial value problem, for $t > t_0$,

$$\frac{d}{dt}\,\xi_i(t) = 1 + \frac{u_i(t)\,(q-1) + [\mathrm{B}(t)\ (q\,\mathbf{x}(t) - \mathbf{P}(\xi_i, t))]_i}{p_i(\xi_i, t)},$$
$$\xi_i(t_0) = \xi_i^0, \qquad [21]$$

where ξ_i^0 is given such that $P_i^0(\xi_i^0) = q\,x_i^0$. Now, only the inverse of the initial age distribution remains to be computed.

B. System Age Quantiles. Likewise, the kth n quantile ξ of the system age solves the initial value problem, for $t > t_0$,

$$\frac{d}{dt}\,\xi(t) = 1 + \frac{\|\boldsymbol{u}(t)\|\ \ (q-1) + \sum_{i=1}^{d} [\mathrm{B}(t)\ (q\,\mathbf{x}(t) - \mathbf{P}(\xi, t))]_i}{\|\mathbf{p}(\xi, t)\|},$$
$$\xi(t_0) = \xi^0, \qquad [22]$$

where ξ^0 is given such that $\|\mathbf{P}^0(\xi^0)\| = q\,\|\mathbf{x}^0\|$.

5. Transit-Time Distributions

A. Backward Transit Time. Following ref. 16, we define the backward transit time $\mathrm{BTT}(t_e)$ as the age of particles in the output from the system at exit time $t_e \geq t_0$. The vector $\boldsymbol{\rho}(t_e)$ of outflow rates (unit: time^{-1}) from the system at time t_e is given by

$$\rho_j(t_e) = -\sum_{i=1}^{d} B_{ij}(t_e), \quad j = 1, 2, \ldots, d. \qquad [23]$$

We can write the age density of the outflow at time t_e as

$$p_{\mathrm{BTT}}(a, t_e) = \rho^T(t_e)\,\mathbf{p}(a, t_e), \quad a \geq 0, \quad t_e \geq t_0. \qquad [24]$$

Owing to the well-mixed assumption, the outflow from compartment i at time t_e is given by $r_i(t_e) := \rho_i(t_e)\,x_i(t_e)$. Consequently, $\mathbf{r}(t_e)$ denotes the vector of outflows from the system at time t_e. By Eq. **15** we obtain for the nth moment of the backward transit time at time t_e the expression

$$\overline{\mathrm{BTT}}^n(t_e) = \frac{1}{\|\mathbf{r}(t_e)\|} \int_0^\infty a^n\, p_{\mathrm{BTT}}(a, t_e)\ da$$
$$= \frac{1}{\|\mathbf{r}(t_e)\|}\,\rho^T(t_e) \int_0^\infty a^n\,\mathbf{p}(a, t_e)\,da, \qquad [25]$$

which, based on $\mathbf{r}^T(t_e) = \rho^T(t_e)\,\mathrm{X}(t_e)$ and Eq. **15**, yields

$$\overline{\mathrm{BTT}}^n(t_e) = \frac{1}{\|\mathbf{r}(t_e)\|}\,\mathbf{r}^T(t_e)\,\bar{\mathbf{a}}^n(t_e). \qquad [26]$$

Note that to guarantee the existence of the nth moment of $\mathrm{BTT}(t_e)$, the nth moment of the initial age density must exist, too.

B. Forward Transit Time. For a particle entering the system at its arrival time $t_a > t_0$, we consider its forward transit time $\mathrm{FTT}(t_a)$ as the age $a \geq 0$ that the particle will have when it exits the system at time $t_e = t_a + a$. The density

$$p_{\mathrm{FTT}}(a, t_a) = \rho^T(t_a + a)\,\mathbf{p}(a, t_a + a) \qquad [27]$$

describes the part from the input at time t_a that leaves the system at time $t_a + a$. By the relation $t_e = t_a + a$, we obtain immediately a generalized version of Niemi's theorem (19),

$$p_{\mathrm{FTT}}(a, t_a) = p_{\mathrm{BTT}}(a, t_e), \qquad [28]$$

which shows the intrinsic connection between forward and backward transit times.

If we want to compute the moments of $\mathrm{FTT}(t_a)$, we must rely on Eq. **15** and deal with an integral from zero to infinity. Unfortunately, we cannot profit from the close link (Eq. **28**) between FTT and BTT, since the exit time $t_e = t_a + a$ depends on a.

6. Application to a Simple Global Carbon Cycle Model

We consider the simple global carbon cycle model introduced in ref. 3 and depicted in Fig. 1. It consists of three compartments: atmosphere (A), terrestrial biosphere (T), and surface ocean (S). Furthermore, it depends on two parameters α and β which control the fluxes from the atmosphere to the terrestrial biosphere and from the surface ocean to the atmosphere, respectively. We consider two different parameter sets: (*i*) $(\alpha, \beta) = (0.2, 10)$ and (*ii*) $(\alpha, \beta) = (1, 1)$. Parameter set *i* is from the original publication (3) and describes a nonlinear scenario. Parameter set *ii* makes the model become linear and we use this scenario as a reference measure for the nonlinear version (*i*). A detailed description and how the notation from the original publication (3) can be transformed to fit Eq. **1** is given in *S7. The Detailed Model Description*. In *S8. The Derivation of the Results from the Example Application* we explain in detail how our derived formulas can be used to produce the following results.

We consider the system in equilibrium in the year 1765 and observe different age densities in the different compartments (Fig. 2). After the year 1765, we perturb the system by an

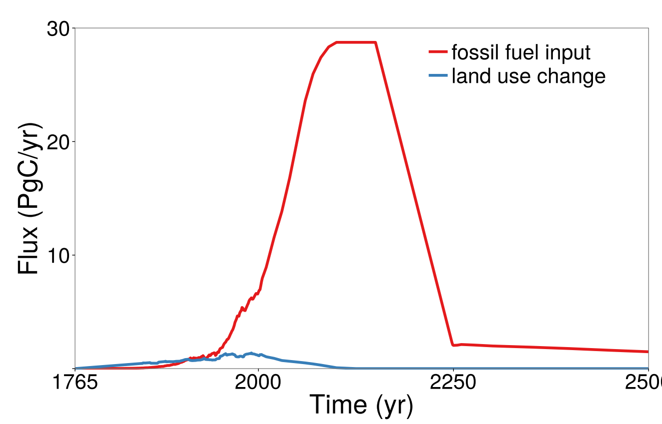

Fig. 3. Anthropogenic perturbations of the global carbon cycle by fossil fuel emissions (u_A, red) and land use change (f_{TA}, blue) according to RCP/ECP8.5.

APPLIED MATHEMATICS

ECOLOGY

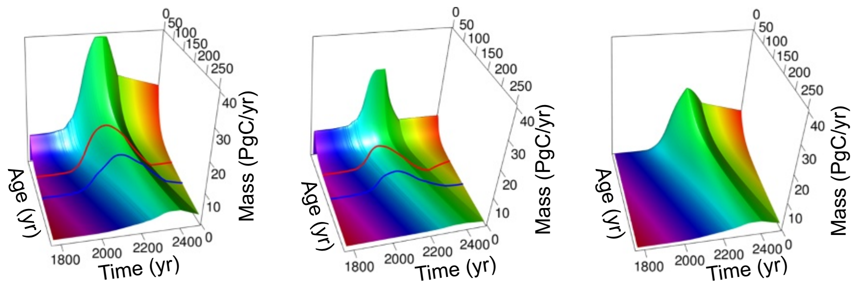

Fig. 4. Time evolution of the atmospheric carbon's age density. *Left*, the nonlinear version; *Center*, the linear version; *Right*, the difference between the two (*Left* minus *Center*). Red curves show the median age and blue curves the mean age. The surface color is constant along the time–age diagonal; it reflects the moment of entry into the system. At the very left edges of *Left* and *Center* panels (time = 1765 y) we can identify the equilibrium age density of the atmospheric carbon (compare Fig. 2), whereas the front edges (age = 250 y) show how much mass is in the system with age equal to 250 y from the year 1765 through the year 2500.

additional external input flux u_A of carbon to the atmosphere caused by fossil fuel combustion and an additional internal independent flux f_{TA} caused by land use change (Fig. 3). For the interval 1765–2100, the data correspond to the Representative Concentration Pathways Scenario 8.5 (RCP8.5) (20), whereas the data for the interval 2100–2500 stem from the Extended Concentration Pathways Scenario 8.5 (ECP8.5) (21). We assume constant emissions after 2100, followed by a smooth transition to stabilized atmospheric CO_2 concentrations after the year 2250 achieved by linear adjustment of emissions after the year 2150 (21). The perturbations make the age densities change with time such that they can be depicted by 2D surfaces in a 3D space (Fig. 4).

To obtain useful information from these density functions, we address two climate-relevant questions inspired by O'Neill et al. (22):

How Old Is Atmospheric Carbon? The entire time evolutions of the atmospheric carbon's age density derived from the two versions of the model are depicted in Fig. 4 (*Left*, nonlinear; *Center*, linear), and so we can answer the question of age of atmospheric carbon for all years between 1765 and 2500. In the year 2017, its mean age is $126.35\,\mathrm{y}$ (linear: $128.32\,\mathrm{y}$) and the median age is equal to $61.76\,\mathrm{y}$ ($62.69\,\mathrm{y}$). The SD equals $161.72\,\mathrm{y}$ ($162.92\,\mathrm{y}$), indicating that the age distribution has a long tail, a feature which cannot be derived from the mean only.

In these numbers we recognize only very little differences between the nonlinear and the linear model. Nevertheless, we can observe important differences in the entire evolution of the age distributions depicted in Fig. 4, *Left* and *Center*. The differences are twofold. First, the pure amount of atmospheric carbon is much higher in the nonlinear model. Second, the age distributions of atmospheric carbon show also different shapes for the two scenarios. This results in the nonflat surface shown in Fig. 4, *Right*, depicting the difference between the densities of atmospheric carbon in the nonlinear and the linear version of the model.

How Long Will a Significant Fraction of a Pulse of Fossil Fuel Carbon, Emitted to the Atmosphere Today, Remain in the System? We consider carbon entering directly into the atmosphere at specific times t_a and want to know how long it will take to remove it from the system. The forward transit time at time t_a describes how old mass entering the system at time t_a will be at the time of its exit. As indicated in Fig. 5, *Left* for the nonlinear model the forward transit-time distribution of mass injected between 1800 and 2170 constantly shifts to older ages, while it shifts back to younger ages after 2170. The medians of the forward transit time of mass injected in the years 1800, 1990, 2015 (Paris Agreement), 2170, and 2300 are given by $79.85\,\mathrm{y}$, $82.91\,\mathrm{y}$, $86.12\,\mathrm{y}$, $108.91\,\mathrm{y}$, and $102.61\,\mathrm{y}$, respectively. As Fig. 5, *Right* shows, the situation is very different in the linear scenario. Here, the forward transit-time distribution does not depend at all on the injection time and remains the same as in the steady state in the year 1765, because the coefficients of B remain constant over time. Obviously, taking into account nonlinear processes leads to a significant increase in the lifetime of fossil fuel-derived carbon according to this model.

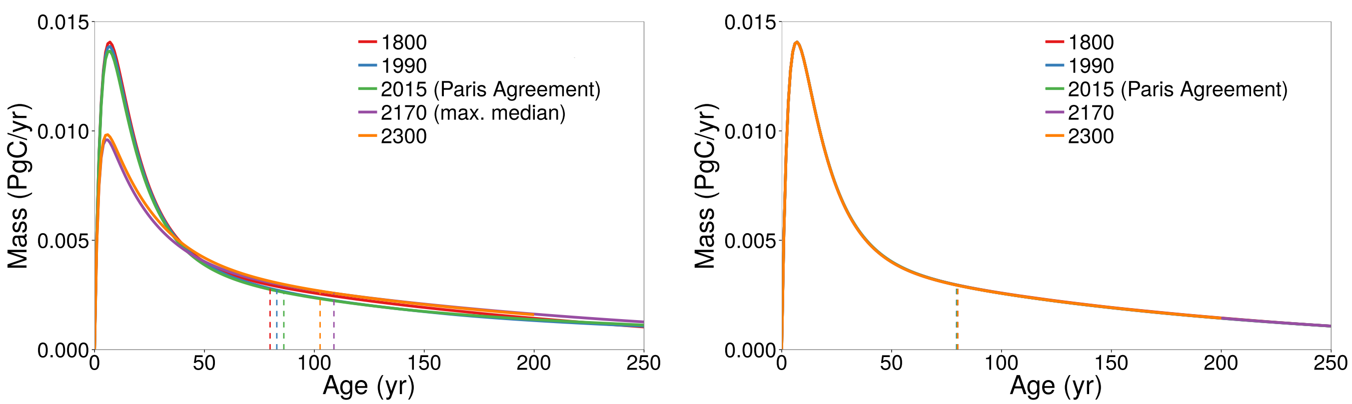

Fig. 5. Forward transit-time densities of fossil fuel carbon entering the atmosphere in the years 1800 (red), 1990 (blue), 2015 (green), 2170 (purple), and 2300 (orange). *Left* shows the nonlinear version and *Right* shows the linear one. Orange curves end at the age of 200 y, because our simulation lasts only until the year 2500. The medians (dashed vertical lines) in the nonlinear version increase until the year 2170 and then start decreasing; in the linear version the distributions and medians remain constant.

7. Summary and Conclusions

We obtained transit-time and age distributions for well-mixed compartmental models. Our results are not restricted to linear models or systems in steady state, but hold even for nonlinear time-dependent models. This fundamental advance allows us to drop the assumption that the system is in equilibrium, an assumption which is unreasonable for most natural systems.

The derivation of the formulas for the age densities relies only on the general solution formula for linear time-dependent systems (Eq. **4**). In nonlinear systems, known solution trajectories are interpreted linearly and then the systems can be treated as if they were linear. This approach also allows us to consider age densities of subsystems such as all mass that entered the system through a specific compartment (*S8. The Derivation of the Results from the Example Application*).

Additionally, we obtained ODEs to compute means, higher-order moments, and quantiles (e.g., the median) of ages. This leads, by very fast computations, to much more precise characterizations of age distributions than were possible before by looking only at the means.

The power of these results is shown in an application to a simple global carbon cycle model. We demonstrate how much transit-time and age distributions differ between a nonlinear and a linear version of the model. First, nonlinearities lead to a tremendously higher amount of carbon in the atmosphere. Second, these two versions of a simple model already suggest that the lifetime of fossil fuel-derived atmospheric carbon is substantially increased by nonlinear processes (23). Sizable differences in transit-time and age distributions of the two model versions might be a criterion to select one version or the other.

We stress that the model used here is very simple and used mainly to demonstrate the power and versatile applications of our mathematical framework in a comprehensible manner. It is important to emphasize that for any global carbon cycle model represented as a well-mixed compartmental system, no matter how many compartments it comprises, we could answer questions of high scientific and societal interest (e.g., the age of the current atmospheric carbon and the future exit age of carbon that now enters the system).

Our results are not restricted to carbon cycle models, of course, but can be readily applied to all possible well-mixed compartmental systems. To that end, we provide a Python package which implements all theoretical results and makes them usable by a few simple commands (https://github.com/MPIBGC-TEE/CompartmentalSystems). This package also includes a demonstration (Jupyter) notebook and an HTML file with code to reproduce the figures and to show more characteristics of the model.

ACKNOWLEDGMENTS. We thank S. E. Trumbore, D. Feist, C. Simon, and two anonymous reviewers for providing comments on previous versions of the manuscript. Funding was provided by the Max Planck Society and the German Research Foundation through its Emmy Noether Program (SI 1953/2–1).

1. Anderson DH (1983) *Compartmental Modeling and Tracer Kinetics* (Springer Science & Business Media, Berlin).
2. Eriksson E (1971) Compartment models and reservoir theory. *Annu Rev Ecol Syst* 2: 67–84.
3. Rodhe H, Björkström A (1979) Some consequences of non-proportionality between fluxes and reservoir contents in natural systems. *Tellus* 31:269–278.
4. Manzoni S, Porporato A (2009) Soil carbon and nitrogen mineralization: Theory and models across scales. *Soil Biol Biochem* 41:1355–1379.
5. Nash J (1957) The form of the instantaneous unit hydrograph. *Int Assoc Sci Hydrol* 3:114–121.
6. Jacquez JA, Simon CP (1993) Qualitative theory of compartmental systems. *SIAM Rev Soc Ind Appl Math* 35:43–79.
7. Bolin B, Rodhe H (1973) A note on the concepts of age distribution and transit time in natural reservoirs. *Tellus* 25:58–62.
8. Sierra CA, Müller M, Metzler H, Manzoni S, Trumbore SE (2016) The muddle of ages, turnover, transit, and residence times in the carbon cycle. *Glob Chang Biol* 23: 1763–1773.
9. Nir A, Lewis S (1975) On tracer theory in geophysical systems in the steady and non-steady state. Part I. *Tellus* 27:372–383.
10. Manzoni S, Katul GG, Porporato A (2009) Analysis of soil carbon transit times and age distributions using network theories. *J Geophys Res Biogeosci* 114:1–14.
11. Thompson MV, Randerson JT (1999) Impulse response functions of terrestrial carbon cycle models: Method and application. *Glob Chang Biol* 5:371–394.
12. Metzler H, Sierra CA (2017) Linear autonomous compartmental models as continuous-time Markov chains: Transit-time and age distributions. *Math Geosci* 50:1–34.
13. Botter G, Bertuzzo E, Rinaldo A (2011) Catchment residence and travel time distributions: The master equation. *Geophys Res Lett* 38:L11403.
14. Calabrese S, Porporato A (2015) Linking age, survival, and transit time distributions. *Water Resour Res* 51:8316–8330.
15. Harman CJ (2015) Time-variable transit time distributions and transport: Theory and application to storage-dependent transport of chloride in a watershed. *Water Resour Res* 51:1–30.
16. Rasmussen M, et al. (2016) Transit times and mean ages for nonautonomous and autonomous compartmental systems. *J Math Biol* 73:1379–1398.
17. McKendrick AG (1926) Applications of mathematics to medical problems. *Proceedings of the Edinburgh Mathematical Society* 44:98–130.
18. von Foerster H (1959) Some remarks on changing populations. *Kinetics Cell Prolif*, ed Stohlman, JF (Grune and Stratton, New York), pp 382–407.
19. Niemi AJ (1977) Residence time distributions of variable flow processes. *Int J Appl Radiat Isotopes* 28:855–860.
20. Fujino J, Nair R, Kainuma M, Masui T, Matsuoka Y (2006) Multi-gas mitigation analysis on stabilization scenarios using AIM global model. *Energy J* 27:343–353.
21. Meinshausen M, et al. (2011) The RCP greenhouse gas concentrations and their extensions from 1765 to 2300. *Clim Change* 109:213–241.
22. O'Neill BC, Oppenheimer M, Gaffin SR (1997) Measuring time in the greenhouse; an editorial essay. *Clim Change* 37:491–505.
23. Archer D, Brovkin V (2008) The millennial atmospheric lifetime of anthropogenic CO_2. *Clim Change* 90:283–297.
24. Brockett RW (2015) *Finite Dimensional Linear Systems* (SIAM, New York), Vol 74.
25. Desoer CA, Vidyasagar M (2009) *Feedback Systems: Input-Output Properties* (SIAM, New York), Vol 55.
26. Engel KJ, Nagel R (2000) *One-Parameter Semigroups for Linear Evolution Equations* (Springer Science & Business Media, New York), Vol 194.
27. Waugh D, Hall TM (2002) Age of stratospheric air: Theory, observations, and models. *Rev Geophys* 40:1-1–1-26.
28. Lewis S, Nir A (1978) On tracer theory in geophysical systems in the steady and non-steady state. Part II. Non-steady state–theoretical introduction. *Tellus* 30: 260–271.
29. Kida H (1983) General circulation of air parcels and transport characteristics derived from a hemispheric GCM. Part 2. Very long-term motions of air parcels in the troposphere and stratosphere. *J Meteorol Soc Jpn* 61:510–522.
30. Hall TM, Plumb RA (1994) Age as a diagnostic of stratospheric transport. *J Geophys Res Atmos* 99:1059–1070.
31. Holzer M, Hall TM (2000) Transit-time and tracer-age distributions in geophysical flows. *J Atmos Sci* 57:3539–3558.
32. Strogatz SH (1994) *Nonlinear Dynamics and Chaos: With Applications to Physics, Biology, Chemistry, and Engineering* (Perseus, Reading, MA).
33. McDonnell JJ (2017) Beyond the water balance. *Nat Geosci* 10:396–396.
34. Porporato A, Calabrese S (2015) On the probabilistic structure of water age. *Water Resour Res* 51:3588–3600.
35. Asmussen S (2003) *Applied Probability and Queues* (Springer Science & Business Media, New York), 2nd Ed.