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

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Many systems in nature are modeled using compartmental systems (reservoir/pool/box systems) expressed as a set of first-order differential equations that describe 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 (nonautonomous) 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 nonautonomous system with the same solution trajectory. We demonstrate how to exploit this general 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 compartmental ages. Our results generalize available density formulas for the linear time-independent (autonomous) case and mean-age formulas for the linear nonautonomous 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.

well-mixed | compartmental | nonlinear | nonautonomous | transit-time distribution | age distribution | mean transit time | mean age

Compartmental systems are used in a large variety of scientific fields such as systems biology, toxicology, ecology, hydrology, and biogeochemistry (1–6). To understand the dynamics of such systems, it is useful to know 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 the age structure of compartmental systems was to establish density formulas in dependence on a not explicitly known system response function (9). The 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 formulas have been derived using a probabilistic approach (12). Most importantly, all these approaches were concerned with linear autonomous models in steady state. This restriction is very unrealistic in most applications, for example when time-depending environmental factors influence the behavior of the system. For systems out of steady state, formulas have been developed for one-reservoir hydrological systems, but the theory has not been expanded to include networks of multiple interconnected compartments (13–15). The first result on the age structure of nonautonomous models was the mean age system, a skew product system of linear ordinary differential equations (linear ODEs) that describes the time evolution of mean compartment ages of linear systems with time-dependent coefficients (16).

In this manuscript, we derive formulas for the transit-time, compartment-ages, and system-age densities of possibly nonlinear nonautonomous models. We further extend the mean age system to higher order moments and provide ODEs to describe the time evolution of compartment age quantiles. These results generalize many earlier results from different scientific fields such as atmospheric sciences, ecology, and hydrology. In appendix S6 we explain in detail how our framework relates to these fields.

As an example application of our theoretical results, we apply our results to a simple nonlinear nonautonomous global carbon cycle model and ask the questions: *How old is the carbon in the atmosphere? How long will newly added carbon by fossil fuel combustion remain in the system?* We also compare the results to the corresponding results of a linear version of the considered model and see that transit-time and age distributions are very different in the two scenarios.

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

Significance Statement

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 only be answered for systems in steady state. 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. One application is provided to answer questions of high scientific and societal interest such as the age of fossil fuel derived carbon in the atmosphere.

Author contributions: The contributions of the authors are as follows. The formulas and numerical algorithms describing the evolution of the transit-time and age distributions have been proved and implemented by the two first authors based on a seminar talk describing their derivation and exemplary implementation in R by M.M. H.M. additionally derived and proved the age-moment system and the quantile ODEs, produced the plots, the Jupyter notebook, and wrote the paper. C.S. conceived the research and designed the example application.

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is 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 $t_0 \in \mathbb{R}$, a finite time horizon $T > t_0$, and consider the d-dimensional initial value problem

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

Here, $\mathbf{x}(t) \in \mathbb{R}^d$ is the vector of compartment contents at time t, $\mathrm{B} = (B_{ij})_{i,j=1,2,\dots,d}$ is a matrix-valued function depending on the current system content and time, $\mathbf{u}$ is a vector-valued function that depends on time, and $\mathbf{x^0}$ 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 $[t_0, T]$.

This system describes how mass flows into it 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 additional restrictions on B which require it 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).

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 it in general *nonlinear* and *nonautonomous*. 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 *autonomous*.

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 nonautonomous compartmental system

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

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 semi-analytical formulas for many properties of nonlinear systems. The "semi" reflects here the fact, that all the theory works under the assumption that $\mathbf{x}$ is already known.

General Solution of the Linear System. We consider the linear nonautonomous compartmental system

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

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

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

where Φ denotes the state transition matrix (S1). 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 $t - \tau$.

Age Distribution

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

$$\mathbf{p}(a, t) = \mathbf{g}(a, t) + \mathbf{h}(a, t), \tag{5}$$

where

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

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

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

is the age density of the mass that has entered the system after t_0. The indicator function $\mathbb{1}_S(a)$ 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 (see S2), for $a > 0$ and $t > t_0$,

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

with boundary condition

$$\mathbf{p}(0, t) = \mathbf{u}(t), \quad t > t_0, \tag{9}$$

and initial condition

$$\mathbf{p}(a, t_0) = \mathbf{p^0}(a), \quad a \geq 0. \tag{10}$$

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

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

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

Cumulative Compartment Age Distribution. We denote by capital letters the cumulative age distributions that correspond to age densities. This means for the initial age density $\mathbf{p^0}$ and $\xi \geq 0$ that $\mathbf{P^0}(\xi) = \int_0^{\xi} \mathbf{p^0}(a)\,da$ is the vector of initial compartment contents with age $a \leq \xi$. Then, by Eq. (5),

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

where

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

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

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

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 the mass that was already in the system at time $t-\xi$ and survived until time t.

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

Moments of the Age Distribution

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

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

to be the *kth moment of the density* $\mathbf{p}$, where $\mathrm{X} = \mathrm{diag}(x_1, x_2, \ldots, x_d)$. 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).

Moments of Compartment Ages.

Semi-Explicit Formula for Compartment Age Moments. To circumvent this problem, we can use the McKendrick-von Foerster equation (8) to compute (see S3) 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

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

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 $\bar{\mathbf{a}}^{0,k}$, $k = 1, 2, \ldots, n$ denotes the moments of the initial age distribution. Note that the integral involved is now over the half-open but finite interval $[0, t-t_0)$.

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 (16). It is given by (see S4)

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

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

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

Notice that we occasionally omitted the time-dependencies to simplify notation and that $\mathbf{v}^T$ denotes the transpose of the vector $\mathbf{v}$. The age moment system (17) is a so-called skew product system. It comes with the advantage of solving the compartments' age moments alongside the compartments' contents. This procedure is both fast and numerically robust.

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

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

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

Age Quantiles

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

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

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

In general, the computation of the quantile relies on the computationally expensive inverse of the cumulative age distribution. It is numerically much faster and easier to instead use the fact (see S5) that ξ_i solves the initial value problem, for $t > t_0$,

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

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.

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

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

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

Transit-Time Distribution

Backward Transit Time. Following (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 $\rho(t_e)$ of outflow rates (unit: time^{-1}) from the system at time $t_e \geq t_0$ 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 $t_e \geq t_0$ 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 $t_e \geq t_0$ 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

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

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

Forward Transit Time. For a particle that enters 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)

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

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

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.

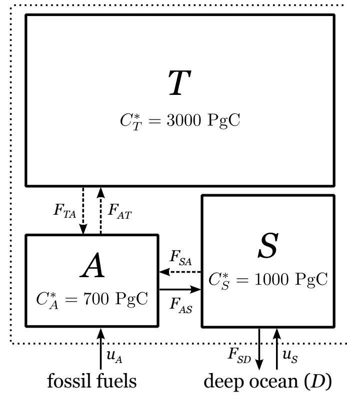

Fig. 1. Simple global carbon cycle model with three compartments (solid boxes within dashed square): atmosphere (A), terrestrial biosphere (T), and surface ocean (S). The indicated carbon contents are the respective steady-state values. External to the modeled system are fossil fuel sources and the deep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

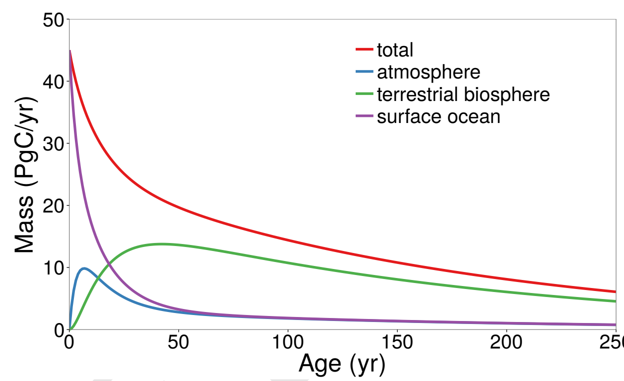

Fig. 2. Pre-industrial age densities of the three compartments: atmosphere (blue), terrestrial biosphere (green), and surface ocean (purple). The red curve shows the age density of the entire system.

Unfortunately, we cannot profit from the close link (28) between FTT and BTT, since the exit time $t_e = t_a + a$ depends on a.

We consider the simple global carbon cycle model introduced in (3) which consists of three compartments: atmosphere (A), terrestrial biosphere (T), and surface ocean (S). The model is depicted in Fig. 1. Furthermore, the model depends on the 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: (1) $(\alpha, \beta) = (0.2, 10)$ and (2) $(\alpha, \beta) = (1, 1)$. Parameter set (1) is taken from the original publication and describes a nonlinear scenario. Parameter set (2) has the effect that the model becomes linear and we use this scenario as a reference measure for the nonlinear model (1). A more detailed description and how the notation from the original publication can be transformed to fit Eq. (1) can be found in S7. In S8 we explain in more detail how our derived formulas can be used to produce the results we present in this section.

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 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 through 2100, the data correspond to the Representative Concentration Pathways 8.5 Scenario (RCP8.5) (20),

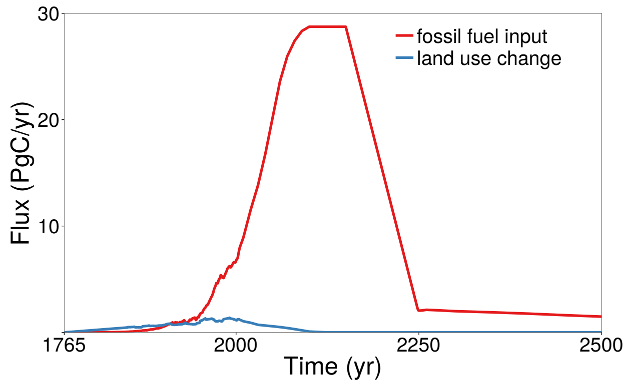

Fig. 3. Anthropogenic perturbations of the global carbon cycle by fossil fuel emissions (u_A, red) and land use change (f_{TA}, blue) according to RCP/ECP8.5.

whereas the data for the interval 2100 through 2500 stem from the Extended Concentration Pathways Scenario 8.5 (ECP8.5). Constant emissions after 2100 are assumed, 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 they can be depicted by two-dimensional surfaces in a three-dimensional space (Fig. 4).

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

Origin of Current Effects: How Old Is the Current Atmospheric Burden?. The entire time evolution of the atmosphere's age density derived from the nonlinear model is depicted in the left panel of Fig. 4, and so we can answer the question of origin of current effects for all times between 1765 and 2500. In the year 2017, the mean age of carbon in the atmosphere is 126.35 yr (linear: 128.32 yr) and the median age is equal to 61.76 yr (62.69 yr). The standard deviation equals 161.72 yr (162.92 yr) indicating that the age distribution has a long tail.

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 the left and the middle panel in Fig. 4. The difference consists of two aspects. First, the pure amount of carbon in the atmosphere is much higher in the nonlinear model. Second, the age distributions of carbon in the atmosphere show also different shapes for the two scenarios. This results in the non-flat surface in the right panel which shows the difference between the amounts of carbon in the atmosphere in the nonlinear model and the linear model.

Future Commitment: If We Allow a Pulse of a Gas to Be Emitted Today, How Long Will a Significant Fraction of This Excess Remain in the Atmosphere?. We hypothetically inject 1 Pg C into the atmosphere at different times and want to know how long it will take to remove this carbon from the system. The forward transit time at time t describes how old mass that entered the system at time t will be at the time of its exit. As indicated by the upper panel of Fig. 5, for the nonlinear model the forward transit-time distribution of mass injected between 1765 and 2170 constantly shifts to the right, while it shifts back to the left after 2170. The medians of the forward transit time of mass injected in the years 1800, 1990 (Kyoto Protocol), 2015 (Paris Agreement), 2170, and 2300 are given by 79.85 yr, 82.91 yr, 86.12 yr, 108.91 yr, and 102.61 yr, respectively. As the lower panel of Fig. 5 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.

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 nonautonomous models. Furthermore, they cover the system age as well as separate compartment ages.

The derivation of the formulas for the age densities only relies on the general solution formula for linear nonautonomous systems (Eq. (4)). Nonlinear systems are interpreted linearly, once a solution trajectory is known, and can then be treated as if they were linear. This approach also allows us to consider age densities of subsystems like all mass that entered the system through a specific compartment (see supplementary material).

We also obtained a skew product system of ODEs to compute not only the mean, but also higher order moments of the compartment ages and system age. Furthermore, we derived ODEs for the time evolution of compartment age quantiles of which the median is a special case.

The power of these results is shown in an application to a nonlinear global carbon cycle model. In addition, we show how different the transit-time and age structures of this nonlinear model are from the ones of a linear reference model. We want to stress that the models used here are very simple and used only for illustrative purposes. Nevertheless, for any global carbon cycle model represented as well-mixed compartmental system, we can answer questions of high scientific and societal interest such as the age of the current atmospheric content or 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 the theoretical results and makes them usable by just a few simple commands (https://github.com/MPIBGC-TEE/CompartmentalSystems). This package also includes a demonstration (Jupyter) notebook and an html file with all the code to reproduce the figures and to show additional characteristics of the model (see supplementary material).

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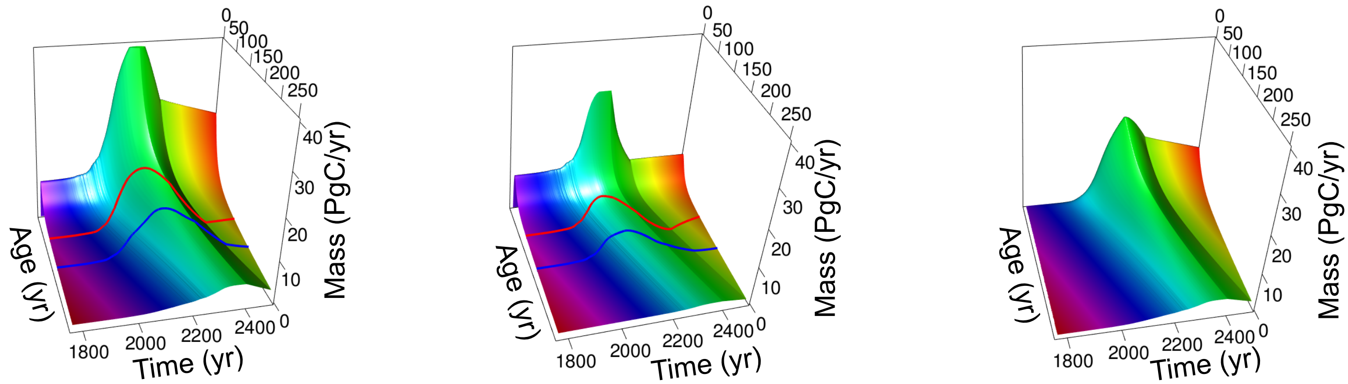

Fig. 4. Time evolution of the atmosphere's carbon age density. The left panel is for the nonlinear model, the middle panel for the linear model, and the right panel shows the difference between the two (panel 1 minus panel 2). Furthermore, the red curves show the respective median age and the blue curves the respective mean age. The surface color is constant along the time-age diagonal such that the color reflects the moment of entry into the system. At the very left edge of the first two panels (time $= 1765\,\mathrm{yr}$) we can identify the equilibrium age density of the atmosphere's carbon (cf. Fig. 2), whereas the front edge (age $= 250\,\mathrm{yr}$) shows how much mass is in the system with age equal to $250\,\mathrm{yr}$ from the year 1765 through the year 2500.

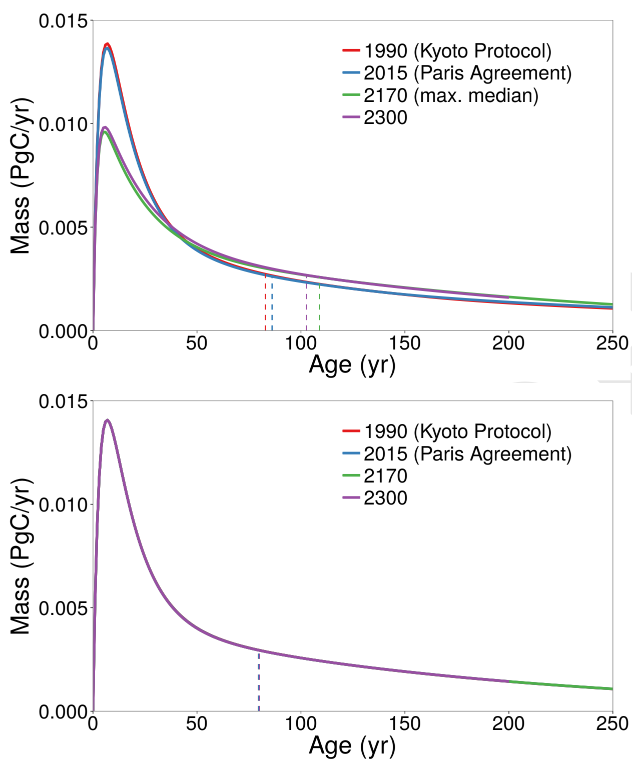

Fig. 5. Forward transit-time densities of $1\,\mathrm{Pg\,C}$ hypothetically injected into the atmosphere in the years 1990 (red), 2015 (blue), 2170 (green), and 2300 (purple). The upper panel is for the nonlinear model and the lower panel for the linear one. The orange curves end at the age of 200, because our simulation only lasts until the year 2500. The medians (dashed vertical lines) in the nonlinear model increase until the year 2170 and then start decreasing, while in the linear model the distributions and with them the medians remain constant.

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S1. The State Transition Matrix Φ

In contrast to the one-dimensional or the autonomous case, the state transition matrix for a nonautonomous multi-dimensional system can in general not be computed analytically. It has nevertheless some useful properties, some of which we collect here. They can be found in (23, 24).

The state transition matrix of the system described by Eq. (3) is the solution of the matrix equation

$$\begin{aligned}\frac{d}{dt}\Phi(t,t_0) &= \mathrm{B}(t)\,\Phi(t,t_0), \quad t>t_0,\\ \Phi(t_0,t_0) &= \mathrm{I},\end{aligned} \tag{S1.1}$$

and is given by the *Peano-Baker series*

$$\Phi(t,t_0) = \mathrm{I} + \int_{t_0}^{t} \mathrm{B}(\tau_1)\,d\tau_1 + \int_{t_0}^{t} \mathrm{B}(\tau_1) \int_{t_0}^{t} \mathrm{B}(\tau_2)\,d\tau_2\,d\tau_1 \cdots . \tag{S1.2}$$

Here, $\mathrm{I}\in\mathbb{R}^{d\times d}$ is the identity matrix.

If $\mathrm{B}(t)=b(t)$ is a scalar, then the Peano-Baker series can be summed to

$$\Phi(t,t_0) = \exp\left(\int_{t_0}^{t} b(\tau)\,d\tau\right), \tag{S1.3}$$

and if $\mathrm{B}(t)=\mathrm{B}$ is a real constant $d\times d$-matrix, then

$$\begin{aligned}\Phi(t,t_0) &= \mathrm{I} + \frac{1}{1!}\,\mathrm{B}^1\cdot(t-t_0)^1 + \frac{1}{2!}\cdot\mathrm{B}^2\,(t-t_0)^2+\cdots\\ &= e^{(t-t_0)\,\mathrm{B}},\end{aligned} \tag{S1.4}$$

where for a $d\times d$-matrix Q the expression e^{Q} denotes the matrix exponential. In many cases this matrix exponential can be computed explicitly. If B is compartmental and invertible, then $(\Phi(t,t_0))_{t\geq t_0}$ is a semigroup of contractions, meaning that

$$\|\Phi(t,t_0)\,\mathbf{u}\| \leq e^{\lambda\,(t-t_0)}\,\|\mathbf{u}\|, \quad t\geq t_0, \mathbf{u}\in\mathbb{R}^d. \tag{S1.5}$$

Here, $\lambda<0$ is the largest real part of the eigenvalues of B. More information on matrix exponentials and semigroups can be found in (25).

If $\mathrm{B}(t)=b(t)\,\mathrm{M}$ is a scalar multiplied with a constant matrix, then

$$\Phi(t,t_0) = \exp\left(\int_{t_0}^{t} b(\tau)\,d\tau\,\mathrm{M}\right). \tag{S1.6}$$

If $\mathrm{B}(t)$ is compartmental for all $t\geq t_0$, then its logarithmic norm $\mu(\mathrm{B}(t))$ is nonpositive. Consequently,

$$\sup_{\|\mathbf{x}\|=1}\|\Phi(t,t_0)\,\mathbf{x}\| \leq \exp\left(\int_{t_0}^{t}\mu(\mathrm{B}(\tau))\,d\tau\right)\leq 1, \tag{S1.7}$$

where the norm $\|\mathbf{v}\|$ of a vector $\mathbf{v}\in\mathbb{R}^d$ is given by

$$\|\mathbf{v}\| = \sum_{i=1}^{d}|v_i|. \tag{S1.8}$$

S2. The McKendrick-von Foerster Equation

Eq. (8) can be interpreted as a multidimensional McKendrick-von Foerster equation, because for the ith compartment

$$\left(\frac{\partial}{\partial a}+\frac{\partial}{\partial t}\right)\,p_i(a,t) = \gamma_i(t)\,p_i(a,t), \tag{S2.1}$$

where $\gamma_i(t)=\sum_{j\neq i}b_{ij}(t)+b_{ii}(t)$ is the combination of the incoming and outgoing rates of mass with age a at time t.

We prove now that our density function satisfies the McKendrick-von Foerster equation Eq. (8). To that end, we compute the total differential of the density function along the characteristics $a(t)=a^0+t$ by

$$\begin{aligned}\frac{d}{dt}\,\mathbf{p}(a,t) &= \frac{\partial}{\partial a}\,\mathbf{p}(a,t)\,\frac{d}{dt}\,a(t) + \frac{\partial}{\partial t}\,\mathbf{p}(a,t)\,\frac{d}{dt}\,t\\ &= \left(\frac{\partial}{\partial a}+\frac{\partial}{\partial t}\right)\,\mathbf{p}(a,t),\end{aligned} \tag{S2.2}$$

where $a^0\geq 0$ is some initial age. We continue in two steps. In the first step we show that Eq. (8) holds on $S_1:=\{(a,t):t\geq t_0, a\geq t-t_0\}$ with initial condition (10). In the second step we show that Eq. (8) holds on $S_2:=\{(a,t):a\geq 0, t\geq t_0, a<t-t_0\}$ with boundary condition (9).

Step 1. On S_1 we have $\mathbf{p}(a,t)=\mathbf{g}(a,t)$. Consequently, we prove the initial condition (10) by

$$\mathbf{p}(a,t_0) = \Phi(t_0,t_0)\,\mathbf{p^0}(a-(t_0-t_0)) = \mathbf{p^0}(a), \tag{S2.3}$$

Furthermore,

$$\mathbf{p}(a,t) = \Phi(t,t_0)\,\mathbf{p^0}(a^0), \tag{S2.4}$$

where $a^0=a-(t-t_0)$ does not change with time on the characteristics. Consequently,

$$\begin{aligned}\left(\frac{\partial}{\partial a}+\frac{\partial}{\partial t}\right)\,\mathbf{p}(a,t) &= \frac{d}{dt}\,\mathbf{p}(a,t)\\ &= \frac{d}{dt}\,\Phi(t,t_0)\,\mathbf{p^0}(a^0)\\ &= \mathrm{B}(t)\,\Phi(t,t_0)\,\mathbf{p^0}(a^0)\\ &= \mathrm{B}(t)\,\mathbf{p}(a,t),\end{aligned} \tag{S2.5}$$

which proves Eq. (8) on S_1.

Step 2. On S_2 we have $\mathbf{p}(a,t)=\mathbf{h}(a,t)$ and $a^0=0$. Consequently, we prove the boundary condition (9) by

$$\mathbf{p}(0,t) = \Phi(t,t-0)\,\mathbf{u}(t-0) = \mathbf{u}(t). \tag{S2.6}$$

Furthermore,

$$\mathbf{p}(a,t) = \Phi(t,s)\,\mathbf{u}(s), \tag{S2.7}$$

where $s=t-a$ does not change with time on the characteristics. Consequently,

$$\begin{aligned}\left(\frac{\partial}{\partial a}+\frac{\partial}{\partial t}\right)\,\mathbf{p}(a,t) &= \frac{d}{dt}\,\mathbf{p}(a,t)\\ &= \frac{d}{dt}\,\Phi(t,s)\,\mathbf{u}(s)\\ &= \mathrm{B}(t)\,\Phi(t,s)\,\mathbf{u}(s)\\ &= \mathrm{B}(t)\,\mathbf{p}(a,t),\end{aligned} \tag{S2.8}$$

which proves Eq. (8) on S_2.

S3. The Semi-Explicit Formula for Compartment Age Moments

We assume that the initial age density $\mathbf{p^0}$ admits finite moments up to order n and denote them by $\bar{\mathbf{a}}^{0,k}$, $k=1,2,\ldots,n$. Additionally, we define

$$\mathbf{y}(t) := \Phi(t,t_0)\,\mathbf{x^0}, \quad t\geq t_0, \tag{S3.1}$$

and

$$\mathbf{z}(t) := \int_{t_0}^{t}\Phi(t,\tau)\,\mathbf{u}(\tau)\,d\tau, \quad t\geq t_0. \tag{S3.2}$$

Consequently, $\mathbf{x}=\mathbf{y}+\mathbf{z}$, where $\mathbf{y}$ describes the evolution of the initial mass and $\mathbf{z}$ describes the evolution of mass that comes later into the system. We use the shorthand $\bar{\mathbf{a}}^n$ for $\bar{\mathbf{a}}^n(t):=\bar{\mathbf{a}}^{\mathbf{x}(t),n}$ and note that we can compute the nth moment of the age density of $\mathbf{x}$ by the corresponding moments of the age densities of $\mathbf{y}$ and $\mathbf{z}$ by

$$\bar{a}_i^n = \frac{y_i\,\bar{a}_i^{\mathbf{y},n}+z_i\,\bar{a}_i^{\mathbf{z},n}}{x_i}, \quad i=1,2,\ldots,d, \tag{S3.3}$$

or in vector notation

$$\bar{\mathbf{a}}^n(t) = \mathrm{X}(t)^{-1}\,\left[\mathrm{Y}(t)\,\bar{\mathbf{a}}^{\mathbf{y},n}(t)+\mathrm{Z}(t)\,\bar{\mathbf{a}}^{\mathbf{z},n}(t)\right]. \tag{S3.4}$$

We see from Eq. (15) that

$$\mathrm{Y}(t)\,\bar{\mathbf{a}}^{\mathbf{y},n}(t) = \int_0^\infty a^n\,\mathbf{g}(a,t)\,da, \qquad [S3.5]$$

which can be transformed by

$$\mathbf{g}(a,t) = \mathbb{1}_{[t-t_0,\infty)}(a)\,\Phi(t,t_0)\,\mathbf{p^0}(a-(t-t_0)) \qquad [S3.6]$$

and a change of variables from a to $\tau := a-(t-t_0)$ into

$$\mathrm{Y}(t)\,\bar{\mathbf{a}}^{\mathbf{y},n}(t) = \Phi(t,t_0)\int_0^\infty [\tau+(t-t_0)]^n\,\mathbf{p^0}(\tau)\,d\tau. \qquad [S3.7]$$

An application of the binomial theorem and Eq. (15) leads to

$$\mathrm{Y}(t)\,\bar{\mathbf{a}}^{\mathbf{y},n}(t) = \sum_{k=0}^{n}\binom{n}{k}\,(t-t_0)^{n-k}\,\Phi(t,t_0)\,\mathrm{X}^0\,\bar{\mathbf{a}}^{0,k}. \qquad [S3.8]$$

Furthermore, again by Eq. (15),

$$\mathrm{Z}(t)\,\bar{\mathbf{a}}^{\mathbf{z},n}(t) = \int_0^\infty a^n\,\mathbf{h}(a,t)\,da = \int_0^{t-t_0} a^n\,\mathbf{h}(a,t)\,da. \qquad [S3.9]$$

We plug the sum of Eq. (S3.8) and Eq. (S3.9) into Eq. (S3.4) to establish Eq. (16).

S4. The Compartment Age Moment System

We assume that the initial age density $\mathbf{p^0}$ admits finite moments up to order n and denote them by $\bar{\mathbf{a}}^{0,k}$, $k=1,2,\dots,n$. Furthermore, we assume y and z as in (S3.1, S3.2).

Our goal is to derive a system of ordinary differential equations for the moments up to order n for the age distributions of $\mathbf{x}$. To this end, we try to represent the time derivative of the kth moment by known quantities. For that purpose, we need some auxiliary results.

Auxiliary Results.

Lemma S4.1. *For $k=1,2,\dots,n$, and $t>t_0$,*

$$\frac{d}{dt}\int_0^\infty a^k\,g_i(a,t)\,da = \sum_{j=1}^{d} B_{ij}(t)\,y_j(t)\,\bar{a}_j^{\mathbf{y},k}(t) + k\,y_i(t)\,\bar{a}_i^{\mathbf{y},k-1}(t). \qquad [S4.1]$$

Proof. For simplicity of notation, we do not consider the component i, but the entire vector. We start with

$$\frac{d}{dt}\int_0^\infty a^k\,\mathbf{g}(a,t)\,da \qquad [S4.2]$$

and use

$$\mathbf{g}(a,t) = \mathbb{1}_{[t-t_0,\infty)}\,\Phi(t,t_0)\,\mathbf{p^0}(a-(t-t_0)) \qquad [S4.3]$$

to obtain

$$\frac{d}{dt}\,\Phi(t,t_0)\int_{t-t_0}^\infty a^k\,\mathbf{p^0}(a-(t-t_0))\,da, \qquad [S4.4]$$

which by the product rule turns into

$$\begin{aligned}&\mathrm{B}(t)\,\Phi(t,t_0)\int_{t-t_0}^\infty a^k\,\mathbf{p^0}(a-(t-t_0))\,da\\ &\quad+\Phi(t,t_0)\,\frac{d}{dt}\int_{t-t_0}^\infty a^k\,\mathbf{p^0}(a-(t-t_0))\,da.\end{aligned} \qquad [S4.5]$$

We transform the first term back. In addition, a change of variables in the second term from a to $\tau := a-(t-t_0)$ brings

$$\mathrm{B}(t)\int_0^\infty a^k\,\mathbf{g}(a,t)\,da + \Phi(t,t_0)\,\frac{d}{dt}\int_0^\infty (\tau+(t-t_0))^k\,\mathbf{p^0}(\tau)\,d\tau. \qquad [S4.6]$$

We use Eq. (15) in the first term and in the second term we bring the derivative under the integral by means of the dominated convergence theorem to get

$$\mathrm{B}(t)\,\mathrm{Y}(t)\,\bar{\mathbf{a}}^{\mathbf{y},k}(t) + \Phi(t,t_0)\int_0^\infty k\,(\tau+(t-t_0))^{k-1}\,\mathbf{p^0}(\tau)\,d\tau. \qquad [S4.7]$$

We undo the change of variables in the second term and transform it back to obtain

$$\mathrm{B}(t)\,\mathrm{Y}(t)\bar{\mathbf{a}}^{\mathbf{y},k}(t) + k\int_{t-t_0}^\infty a^{k-1}\,\mathbf{g}(a,t)\,da, \qquad [S4.8]$$

which equals

$$\mathrm{B}(t)\,\mathrm{Y}(t)\bar{\mathbf{a}}^{\mathbf{y},k}(t) + k\,\mathrm{Y}(t)\,\bar{\mathbf{a}}^{\mathbf{y},k-1}. \qquad [S4.9]$$

Computing the ith component, we get

$$\sum_{j=1}^{d} B_{ij}(t)\,y_j(t)\,\bar{a}_j^{\mathbf{y},k} + k\,y_i(t)\,\bar{a}_i^{\mathbf{y},k-1}(t) \qquad [S4.10]$$

and we are finished with the proof. $\square$

Lemma S4.2. *For $k=1,2,\dots,n$, and $t>t_0$,*

$$\begin{aligned}\frac{d}{dt}\int_0^\infty a^k\,h_i(a,t)\,da &= \sum_{j=1}^{d} B_{ij}(t)\,z_j(t)\,\bar{a}_i^{\mathbf{z},k}(t)\\ &\quad+k\,z_i(t)\,\bar{a}_i^{\mathbf{z},k-1}(t).\end{aligned} \qquad [S4.11]$$

Proof. Again, for simplicity of notation, we do not consider the component i, but the entire vector. From

$$\mathbf{h}(a,t) = \mathbb{1}_{[0,t-t_0)}(a)\,\Phi(t,t-a)\,\mathbf{u}(t-a), \qquad [S4.12]$$

we get

$$\int_0^\infty a^k\,\mathbf{h}(a,t)\,da = \lim_{\varepsilon\searrow 0}\int_0^{t-t_0-\varepsilon} a^k\,\mathbf{h}(a,t)\,da. \qquad [S4.13]$$

We can interchange the limit and the derivative to see

$$\frac{d}{dt}\int_0^\infty a^k\,\mathbf{h}(a,t)\,da = \lim_{\varepsilon\to 0}\frac{d}{dt}\int_0^{t-t_0-\varepsilon} a^k\,\mathbf{h}(a,t)\,da. \qquad [S4.14]$$

By an application of the Leibniz rule to the right hand side we obtain

$$\lim_{\varepsilon\searrow 0}\int_0^{t-t_0-\varepsilon} a^k\,\frac{\partial}{\partial t}\,\mathbf{h}(a,t)\,da + (t-t_0-\varepsilon)^k\,\mathbf{h}(t-t_0-\varepsilon,t). \qquad [S4.15]$$

In S2 (Step 2) we derived that for $a\in[0,t-t_0-\varepsilon]$,

$$\frac{\partial}{\partial t}\,\mathbf{h}(a,t) = \mathrm{B}(t)\,\mathbf{h}(a,t) - \frac{\partial}{\partial a}\,\mathbf{h}(a,t), \qquad [S4.16]$$

which we plug into the first term of Eq. (S4.15) and turn it into

$$\lim_{\varepsilon\searrow 0}\int_0^{t-t_0-\varepsilon} a^k\,\left[\mathrm{B}(t)\,\mathbf{h}(a,t) - \frac{\partial}{\partial a}\,\mathbf{h}(a,t)\right]\,da, \qquad [S4.17]$$

which equals by Eq. (15)

$$\mathrm{B}(t)\,\mathrm{Z}(t)\,\bar{\mathbf{a}}^{\mathbf{z},k} - \lim_{\varepsilon\searrow 0} \int_0^{t-t_0-\varepsilon} a^k\,\frac{\partial}{\partial a}\,\mathbf{h}(a,t)\,da. \qquad \text{[S4.18]}$$

We integrate by parts and use again Eq. (15) to get

$$\begin{aligned}\mathrm{B}(t)\,\mathrm{Z}(t)\,\bar{\mathbf{a}}^{\mathbf{z},k} - \lim_{\varepsilon\searrow 0}\,(t-t_0-\varepsilon)^k\,\mathbf{h}(t-t_0-\varepsilon,t)\\ + k\,\mathrm{Z}(t)\,\bar{\mathbf{a}}^{\mathbf{z},k-1}.\end{aligned} \qquad \text{[S4.19]}$$

Together with Eq. (S4.15), we have

$$\frac{d}{dt}\int_0^\infty a^k\,\mathbf{h}(a,t)\,da = \mathrm{B}(t)\,\mathrm{Z}(t)\,\bar{\mathbf{a}}^{\mathbf{z},k} + k\,\mathrm{Z}(t)\,\bar{\mathbf{a}}^{\mathbf{z},k-1}, \qquad \text{[S4.20]}$$

which completes the proof by considering the ith component. □

Lemma S4.3. *For $k = 1,2,\ldots,n$, and $t > t_0$,*

$$\frac{d}{dt}\,\left(x_i(t)\,\bar{a}_i^k(t)\right) = \sum_{j=1}^d B_{ij}(t)\,x_j(t)\,\bar{a}_j^k(t) + k\,x_i(t)\,\bar{a}_i^{k-1}(t). \qquad \text{[S4.21]}$$

Proof. From Eq. (15) and $\mathbf{p}(a,t) = \mathbf{g}(a,t) + \mathbf{h}(a,t)$, we know

$$\begin{aligned}\frac{d}{dt}\,\left[x_i(t)\,\bar{a}_i^k(t)\right] &= \frac{d}{dt}\int_0^\infty a^k\,p_i(a,t)\,da\\ &= \frac{d}{dt}\int_0^\infty a^k\,g_i(a,t)\,da + \frac{d}{dt}\int_0^\infty a^k\,h_i(a,t)\,da.\end{aligned} \qquad \text{[S4.22]}$$

Consequently, we can apply Lemmas S4.1 and S4.2, and use

$$x_i\,\bar{a}_i{}^k = y_i\,\bar{a}_i^{\mathbf{y},k} + z_j\,\bar{a}_i^{\mathbf{z},k} \qquad \text{[S4.23]}$$

from Eq. (S3.3) to obtain

$$\begin{aligned}\frac{d}{dt}\,\left(x_i\,\bar{a}_i^k\right) &= \sum_{j=1}^d B_{ij}\,y_j\,\bar{a}_j^{\mathbf{y},k} + k\,y_i\,\bar{a}_i^{\mathbf{y},k-1}\\ &\quad + \sum_{j=1}^d B_{ij}\,z_j\,\bar{a}_j^{\mathbf{z},k} + k\,z_i\,\bar{a}_i^{\mathbf{z},k-1}\\ &= \sum_{j=1}^d B_{ij}\,\left(y_j\,\bar{a}_j^{\mathbf{y},k} + z_j\,\bar{a}_j^{\mathbf{z},k}\right)\\ &\quad + k\,\left(y_j\,\bar{a}_i^{\mathbf{y},k-1} + z_j\,\bar{a}_i^{\mathbf{z},k-1}\right)\\ &= \sum_{j=1}^d B_{ij}\,x_j\,\bar{a}_j^k + k\,\bar{a}_i^{k-1}.\end{aligned} \qquad \text{[S4.24]}$$

□

Proof of the Age Moment System. Let $k \in \{1,2,\ldots,n\}$. We compute the time derivative of $\bar{a}_i^k$ at $t > t_0$ by

$$\frac{d}{dt}\,\bar{a}_i^k(t) = \frac{d}{dt}\,\left[\frac{x_i(t)\,\bar{a}_i^k(t)}{x_i(t)}\right]. \qquad \text{[S4.25]}$$

We apply the quotient rule and Lemma S4.3 to get

$$\begin{aligned}\frac{d}{dt}\,\bar{a}_i^k &= \frac{1}{x_i^2}\,\Bigg[\left(\sum_{j=1}^d B_{ij}\,x_j\,\bar{a}_j^k + k\,x_i\,\bar{a}_i^{k-1}\right)\,x_i\\ &\qquad - x_i\,\bar{a}_i^k\,\frac{d}{dt}\,x_i\Bigg]\\ &= k\,\bar{a}_i^{k-1}\\ &\quad + \frac{1}{x_i}\,\left[\sum_{j=1}^d B_{ij}\,x_j\,\bar{a}_j^k - \bar{a}_i^k\,\left(\sum_{j=1}^d B_{ij}\,x_j + u_i\right)\right]\\ &= k\,\bar{a}_i^{k-1} + \frac{1}{x_i}\,\left[\sum_{j=1}^d B_{ij}\,x_j\,\left(\bar{a}_j^k - \bar{a}_i^k\right) - \bar{a}_i^k\,u_i\right].\end{aligned} \qquad \text{[S4.26]}$$

Now, we can bring all components $i = 1,2,\ldots,d$ into one vector and the proof is complete.

S5. The Age Quantiles

We want to show that the compartment age quantiles solve the initial value problem given in the main text. The time evolution of the kth n-quantile $\xi_i(t)$ of the age of compartment i can be described by taking the time derivative in both sides of Eq. (20), which gives

$$\frac{d}{dt}\int_0^{\xi_i(t)} p_i(a,t)\,da = q\,\left[\mathrm{B}(t)\,\mathbf{x}(t)\right]_i + q\,u_i(t). \qquad \text{[S5.1]}$$

Using the Leibniz rule, we can rewrite the left hand side to

$$\int_0^{\xi_i(t)} \frac{\partial}{\partial t}\,p_i(a,t)\,da + p_i(\xi_i(t),t)\,\frac{d}{dt}\,\xi_i(t). \qquad \text{[S5.2]}$$

Outside the null set $\{a \geq 0 : a = t - t_0\}$ the McKendrick-von Foerster equation (8) holds. Consequently,

$$\int_0^{\xi_i(t)} \frac{\partial}{\partial t}\,p_i(a,t)\,da = \int_0^{\xi_i(t)} \left(\left[\mathrm{B}(t)\,\mathbf{p}(a,t)\right]_i - \frac{\partial}{\partial a}\,p_i(a,t)\right)\,da, \qquad \text{[S5.3]}$$

which turns by integration by parts and $p_i(0,t) = u_i(t)$ into

$$\left[\mathrm{B}(t)\,\mathbf{P}(\xi_i(t),t)\right]_i - p_i(\xi_i(t),t) + u_i(t). \qquad \text{[S5.4]}$$

We plug it into Eq. (S5.2) and the result into Eq. (S5.1) to prove, for $t > t_0$,

$$\frac{d}{dt}\xi_i(t) = 1 + \frac{u_i(t)\,(q-1) + \left[\mathrm{B}(t)\,\left(q\,\mathbf{x}(t) - \mathbf{P}(\xi_i,t)\right)\right]_i}{p_i(\xi_i,t)}. \qquad \text{[S5.5]}$$

The initial value ξ_i^0 is just the kth n-quantile of the initial age of the content of compartment i. The proof for the system age quantile ξ follows analogously from $\|\mathbf{P}(\xi(t),t)\| = q\,\|\mathbf{x}(t)\|$.

S6. The Relations to Earlier Results and Possible Applications

Linear Autonomous Systems. Our results generalize earlier results on explicit formulas for ages and transit times of linear autonomous compartmental systems in steady state. To arrive at this conclusion, we consider such a system given by

$$\begin{aligned}\frac{d}{dt}\mathbf{x}(t) &= \mathrm{B}\,\mathbf{x}(t) + \mathbf{u}, \quad t > t_0,\\ \mathbf{x}(t_0) &= \mathbf{x^0},\end{aligned} \qquad \text{[S6.1]}$$

where B is an invertible compartmental matrix. Recall that in autonomous systems both B and $\mathbf{u}$ are independent of time. The state transition matrix $\Phi(t,s) = e^{(t-s)\,\mathrm{B}}$ is then given by the matrix exponential. Therefore, for $a \geq 0$ and $t \geq t_0$,

$$\mathbf{p}(a,t) = e^{(t-t_0)\,\mathrm{B}}\,\mathbf{p^0}(a-(t-t_0)) + e^{a\,\mathrm{B}}\,\mathbf{u}. \qquad \text{[S6.2]}$$

Since $\lim_{t\to\infty} e^{t\,\mathrm{B}} = 0$ if B is compartmental and invertible, $\lim_{t\to\infty} \mathbf{p}(a,t) = e^{a\,\mathrm{B}}\,\mathbf{u}$. This vector contains the compartment age densities of system (S6.1) after it has run for an infinite time. Hence, they belong to the steady state $\mathbf{x}^* = -\mathrm{B}^{-1}\,\mathbf{u}$ of the system. If we divide each component of the steady-state age density vector by the steady-state content of the corresponding compartment, we obtain the normalized age density vector from (12), namely

$$\mathbf{f_a}(a) = (\mathrm{X}^*)^{-1}\,e^{a\,\mathrm{B}}\,\mathbf{u}, \quad a \geq 0, \qquad \text{[S6.3]}$$

where $\mathrm{X}^* = \mathrm{diag}(x_1^*, x_2^*, \ldots, x_d^*)$. This means that along with the compartment contents also the age distributions converge to their steady state.

If we start the system with the steady-state age structure by choosing $\mathbf{p^0}(a) = e^{a\,\mathrm{B}}\,\mathbf{u}$, then

$$\mathbf{x^0} = \int_0^\infty e^{a\,\mathrm{B}}\,\mathbf{u}\,da = -\mathrm{B}^{-1}\,\mathbf{u} = \mathbf{x}^*. \qquad \text{[S6.4]}$$

For $t \geq t_0$ and $a > t - t_0$ we have

$$\mathbf{p}(a,t) = e^{(t-t_0)\,\mathrm{B}}\,e^{[a-(t-t_0)]\,\mathrm{B}}\,\mathbf{u} = e^{a\,\mathrm{B}}\,\mathbf{u}, \qquad \text{[S6.5]}$$

and for $t \geq t_0$ and $a \leq t - t_0$ we have also

$$\mathbf{p}(a,t) = e^{[t-(t-a)]\,\mathrm{B}}\,\mathbf{u} = e^{a\,\mathrm{B}}\,\mathbf{u}. \qquad \text{[S6.6]}$$

Consequently, both the system's content and its age structure remain constant for all time if the system is in steady state.

Since the backward transit time is the age of a particle at the moment when it leaves the system, in steady state and after normalization Eq. (24) coincides with the formula given in (12). The same formula holds also for the forward transit time density in steady state, since by Eq. (28) the forward transit time is only a time-shifted backward transit time.

Different Approaches for Different Scenarios. Depending on the structure of the system, there exist many different approaches to obtain transit-time and age distributions, most of which are special cases of our present results.

Linear Autonomous Systems. If the compartmental system is linear and autonomous, the response function approach is very useful. A first step in this direction was done by (9) who established formulas for transit-time and age densities in dependence on a system response function, which was not explicitly known. This system response function returns the proportion $h(\tau)$ of the input that leaves the system when time τ has passed by. The concept of response functions was the basis for (11) to compute the desired densities numerically by long-term simulations in two carbon-cycle models by computing the system response to impulsive inputs. The resulting impulse response function ψ depends in the first place on the fixed and constant impulse. Then $\psi(\tau)$ is the vector of mass leaving the system after time τ has elapsed, where each component of the vector belongs to a compartment. The impulse response approach was later investigated theoretically by (10) to obtain transit-time and age densities explicitly for a set of carbon-cycle models of very simple structure, using Laplace transforms. As shown by (12), both the transit-time and the age density are simply the probability density functions of a phase-type distribution and the impulse response function is a matrix exponential. Its argument is a time τ and it maps a vector of incoming mass to a vector of mass leaving the system τ units of time later. If furthermore, the corresponding compartmental matrix $\mathrm{B} \in \mathbb{R}^{d\times d}$ is invertible, which holds true for trap-free open systems (12), then by Eq. (S1.6) it is obvious that the age densities decay exponentially. The phase-type distribution (12) is known to behave asymptotically exponential under condition S1.6 (26). The exponential decay rate λ is exactly the e-folding time of the longest-lived mode of stratospheric transport, as stated in (27).

Consequently, our present work generalizes the response function approach by allowing for time-dependent parameters and nonlinear dependencies.

Linear Nonautonomous Systems. Response theory for linear systems with time-dependent parameters was already presented by (28). Our present work generalizes their results to a multi-dimensional, possibly nonlinear setting. In addition, we provide semi-explicit formulas to compute the time-dependent system response function.

Green's Function. The green's function approach is very common, for example, in atmospheric sciences. Regarding the age of stratospheric air, (29) coined the term "age spectrum" for the age density of a fixed box in the stratosphere. (30) identified the age spectrum as a Green's function which governs the transport of particles from the tropical tropopause to the stratosphere. The stratosphere is modeled as $\mathbb{R}^n$, for $n = 1, 2, 3$. Mostly, the transport is considered to be stationary, which makes the Green's function depend only on one time variable. Consequently, the corresponding Green's function G belongs to a partial differential equation and $G(\tau, x_1, x_2)$ is the mass or concentration that moved from x_1 to x_2 in time τ. The main difference between this approach and ours is the different interpretation of space, since we consider the $\mathbb{R}^n$ discretized into d compartments. After this discretization, the Green's function for linear autonomous systems is a matrix exponential.

(31) consider the age spectrum as the transit-time probability density function from a region Ω to a point space. This is in our context the age density and we point out, that we instead denote as transit time the time a particle needs from its entry into the system until its exit. They do not simplify to a stationary transport, consequently their Green's function depends on two time variables. Consequently, our Green's function Φ does not belong to a partial differential equation anymore, but to a linear ordinary differential equation. This differential equation is nonautonomous and lives on $\mathbb{R}^d$, consequently Φ depends on two time variables and is matrix-valued. Such a Green's function is better known as state transition matrix.

Nonlinear Systems. A classical approach to treat nonlinear systems is the linearization of the system in the neighborhood of a fixed point (32). On the one hand, this approach has the advantage that now the simpler linear steady-state theory can be applied. On the other hand, the resulting outcomes are valid only in the vicinity of this particular fixed point and also only after an infinite amount of time has passed. Our approach, however, requires neither the existence of fixed points nor an infinite history.

Mixed Compartments. A different approach than ours is needed when the well-mixed assumption of the compartments is dropped. The fluxes could be age-dependent, which is a very common case in hydrology, where the focus mostly lies on the annual water balance of catchments (33). Such catchments are usually modeled as one compartment with one influx (precipitation) and two age-dependent outfluxes (evaporation, runoff) (13, 15, 34). Even though this case does not fit directly in our framework, it is possible to approximate the one-compartment system with age-dependent outflow by a multiple-compartment well-mixed system. For time-independent systems, this approximation bases on the fact that every nonnegative probability distribution can be approximated arbitrarily well by a phase-type distribution (35). Doing a similar kind of approximation for a nonautonomous single-catchment model allows the full application of the theory presented here. The recent commentary by (33) emphasizes the restrictions of single-catchment models and highlights the need for splitting the single catchment into several compartments. Our results deliver the demanded "theoretical framework that includes both flow and the age distribution of these flowing and stored waters".

S7. The Detailed Model Description

The model consists of three compartments: atmosphere (A), terrestrial biosphere (T), and surface ocean (S). The letter D stands for the external compartment deep ocean with infinite content. We denote by $C_A = C_A(t)$, $C_T = C_T(t)$, and $C_S = C_S(t)$ the respective carbon contents in Pg C at time t in yr. Two external fluxes add carbon to the system. The first one, u_S, is constant and goes from the deep ocean to the surface ocean, whereas the second one, $u_A = u_A(t)$, is time-dependent and represents carbon added to the atmosphere by the burning of fossil fuels. Carbon can leave the system only if it moves from the surface ocean to the deep ocean. A flux from compartment X to compartment Y is denoted by F_{XY} and the following fluxes exist in the model, all given in $\mathrm{Pg\,C\,yr^{-1}}$:

$$\begin{aligned} F_{AT} &= 60\,(C_A/700)^{\alpha}, & F_{AS} &= 100\,C_A/700, \\ F_{TA} &= 60\,C_T/3000 + f_{TA}, & F_{SA} &= 100\,(C_S/1000)^{\beta} \\ F_{SD} &= 45\,C_S/1000, & u_S &= 45. \end{aligned} \qquad [S7.1]$$

Here, $f_{TA} = f_{TA}(t)$ represents an internal flux from the terrestrial biosphere to the atmosphere caused by land use change (deforestation). Its values and also the values of the external inputs through fossil fuel emissions, $u_A(t)$, are taken as time series data from the RCP/ECP8.5 scenario (see supplementary material). The two parameters α and β control the fluxes from the atmosphere to the terrestrial biosphere and from the surface ocean to the atmosphere, respectively. If both parameters are equal to 1, then the model is linear, otherwise it is nonlinear.

The model can now be described by the three ordinary differential equations, for $t > t_0 = 1765$,

$$\begin{aligned} \frac{d}{dt}\,C_A(t) &= F_{TA}(t) + F_{SA}(t) - F_{AT}(t) - F_{AS}(t) + u_A(t), \\ \frac{d}{dt}\,C_T(t) &= F_{AT}(t) - F_{TA}(t), \\ \frac{d}{dt}\,C_S(t) &= F_{AS}(t) - F_{SA}(t) - F_{SD}(t) + u_S(t), \end{aligned} \qquad [S7.2]$$

with initial conditions $C_A(1765) = 700\,\mathrm{Pg\,C}$, $C_T(1765) = 3000\,\mathrm{Pg\,C}$, $C_S(1765) = 1000\,\mathrm{Pg\,C}$. Note that the right hand side of Eq. (S7.2) depends through Eq. (S7.1) not only on t, but also on the state vector $\mathbf{x}(t) = (C_A(t), C_T(t), C_S(t))^T$. If we now define the state- and time-dependent compartmental matrix $\mathrm{B} = \mathrm{B}(\mathbf{x}(t), t)$ to equal

$$\begin{pmatrix} -C_A^{-1}\,(F_{AT} + F_{AS}) & C_T^{-1}\,F_{TA} & C_S^{-1}\,F_{SA} \\ C_A^{-1}\,F_{AT} & -C_T^{-1}\,F_{TA} & 0 \\ C_A^{-1}\,F_{AS} & 0 & -C_S^{-1}\,(F_{SA} + F_{SD}) \end{pmatrix} \qquad [S7.3]$$

and $\mathbf{u}(t) := (u_A(t), 0, u_S)^T$, then the model fits in the framework of Eq. (1) describing the nonlinear nonautonomous compartmental system

$$\begin{aligned} \frac{d}{dt}\mathbf{x}(t) &= \mathrm{B}(\mathbf{x}(t), t)\,\mathbf{x}(t) + \mathbf{u}(t), \quad t > t_0, \\ \mathbf{x}(t_0) &= \mathbf{x^0}. \end{aligned} \qquad [S7.4]$$

Since at time $t_0 = 1765$ the system is supposed to be in equilibrium,

$$\mathbf{x^0} = (700, 3000, 1000)^T. \qquad [S7.5]$$

S8. The Derivation of the Results from the Example Application

First of all, we solve Eq. (S7.4) numerically on the time interval $[1765, 2500]$ and obtain a solution trajectory $\mathbf{x} = \mathbf{x}(t)$, $t \in [1765, 2500]$. With this solution in hand, we can at all times $t \in [1765, 2500]$ compute the compartmental matrix $\mathrm{B} = \mathrm{B}(\mathbf{x}(t), t)$.

Equilibrium Age Densities. At time $t_0 = 1765$ the system is supposed to be in equilibrium and the land use flux $f_{TA}(t_0)$ vanishes. We plug Eq. (S7.5) in Eq. (S7.3) and get

$$\mathrm{B}(\mathbf{x^0}, t_0) = \begin{pmatrix} -160/700 & 60/3000 & 100/1000 \\ 60/700 & -60/3000 & 0 \\ 100/700 & 0 & -145/1000 \end{pmatrix}. \qquad [S8.1]$$

If we set $\mathrm{B}^0 := \mathrm{B}(\mathbf{x^0}, t_0)$ and $\mathbf{u^0} := \mathbf{u}(t_0) = (0, 0, 45)^T$, then $\mathrm{B}^0\,\mathbf{x^0} + \mathbf{u^0} = \mathbf{0}$. We further define $\mathrm{X}^0 = \mathrm{diag}(x_1^0, x_2^0, x_3^0)$ and apply the steady-state formula

$$\mathbf{p^0}(a) = \left(\mathrm{X}^0\right)^{-1}\,e^{a\,\mathrm{B}^0}\,\mathbf{u^0}, \quad a \geq 0, \qquad [S8.2]$$

from (12) to obtain the vector-valued function $\mathbf{p^0}$ of age densities in equilibrium.

Atmospheric Age. Fig. (4) depicts the two-dimensional surface corresponding to $\mathbf{p} = \mathbf{p}(a, t)$ in the time interval 1765 through 2500 and the age interval 0 to 250. The scalar field $\mathbf{p}$ can be obtained by Eq. (5). In Eq. (S8.2) we have already computed the initial age density $\mathbf{p^0}$, and the input vector $\mathbf{u}$ is given by the RCP/ECP8.5 scenario. Consequently, we are only missing the state transition matrix Φ. We compute Φ by numerically solving the matrix ODE (S1.1) on $\{(t_2, t_1) \in [1765, 2500] \times [1765, 2500] : t_2 \geq t_1\}$ and can then proceed to compute $\mathbf{p}$ on $[0, 250] \times [1765, 2500]$.

To obtain a time trajectory of the mean age and the second moment of the atmosphere, we follow Eq. (17) and solve the 9-dimensional ODE system, for $t_0 = 1765$,

$$\begin{aligned} \frac{d}{dt}\begin{pmatrix} \mathbf{x} \\ \bar{\mathbf{a}}^1 \\ \bar{\mathbf{a}}^2 \end{pmatrix}(t) &= \begin{pmatrix} \mathrm{B}(\mathbf{x}(t), t)\,\mathbf{x}(t) + \mathbf{u}(t) \\ \gamma^1(t, \mathbf{x}(t), \mathbf{1}, \bar{\mathbf{a}}^1(t)) \\ \gamma^2(t, \mathbf{x}(t), \bar{\mathbf{a}}^1(t), \bar{\mathbf{a}}^2(t)) \end{pmatrix}, \quad t > t_0, \\ (\mathbf{x}, \bar{\mathbf{a}}^1, \bar{\mathbf{a}}^2)(t_0) &= (\mathbf{x^0}, \bar{\mathbf{a}}^{0,1}, \bar{\mathbf{a}}^{0,2}), \end{aligned} \qquad [S8.3]$$

where, for $k = 1, 2$, $\gamma^k = (\gamma_1^k, \gamma_2^k, \gamma_3^k)^T$ and for $i = 1, 2, 3$,

$$\begin{aligned} \gamma_i^k(t, \mathbf{x}, \bar{\mathbf{a}}^{k-1}, \bar{\mathbf{a}}^k) &= k\,\bar{a}_i^{k-1} \\ &\quad + \frac{1}{x_i}\left[\sum_{j=1}^{d} B_{ij}\,x_j\,\left(\bar{a}_j^k - \bar{a}_i^k\right) - \bar{a}_i^k\,u_i\right]. \end{aligned} \qquad [S8.4]$$

The initial age moments $\bar{\mathbf{a}}^{0,1}$ and $\bar{\mathbf{a}}^{0,2}$ can be obtained using the equilibrium formula

$$(-1)^n\,n!\,\left(\mathrm{X}^0\right)^{-1}\,\left(\mathrm{B}^0\right)^{-n}\,\mathbf{x^0}, \quad n = 1, 2, \qquad [S8.5]$$

from (12). Then $m_1 := \bar{a}_1^1(t)$ is the mean age of the carbon in the atmosphere at time t and $m_2 := \bar{a}_1^2(t)$ its second moment. The standard deviation can be computed as the square root of $m_2 - m_1^2$ from standard probability theory.

The trajectory of the age median of carbon in the atmosphere can be computed by solving Eq. (21) for $q = 0.5$ and $i = 1$. To that end, the cumulative compartment age distribution $\mathbf{P}$ can be obtained by Eq. (12) together with

$$\mathbf{P^0}(a) = \left(\mathrm{X}^0\right)^{-1}\,\left(\mathrm{B}^0\right)^{-1}\,\left(e^{a\,\mathrm{B}^0} - \mathrm{I}\right)\,\mathbf{u}(t_0), \quad a \geq 0, \qquad [S8.6]$$

where I is the 3-dimensional identity matrix. To obtain Eq. (S8.6), we only need to integrate Eq. (S8.2). The initial age median ξ_1^0 of the atmosphere at time t_0 needs to be approximated by a nonlinear optimization algorithm such that $P_i^0(\xi_i^0) = 0.5\,x_i^0$.

Forward Transit Time of Injected Carbon. To compute the forward transit time density of $1\,\mathrm{Pg\,C}$, we simply change the input vector $\mathbf{u} = \mathbf{u}(t)$ to be constantly equal to $(1, 0, 0)^T$. Now, we apply Eq. (27) and are done. Since the state transition matrix maintains the nonlinear behavior of the system and pretends that all other carbon particles are still in the system, we can separately consider the subsystem of $1\,\mathrm{Pg\,C}$ that comes directly into the atmosphere. After the year 1927, when the fossil fuel emissions constantly exceed this $1\,\mathrm{Pg\,C}$, this approach works perfectly.

Quantiles, such as the median, for the forward transit time need to be computed by nonlinear optimization algorithms.