

Reservoir timescales for anthropogenic CO₂ in the atmosphere

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ABSTRACT

Non-steady state timescales are complicated and their application to specific geophysical systems requires a common theoretical foundation. We first extend reservoir theory by quantifying the difference between turnover time and transit time (or residence time) for time-dependent systems under any mixing conditions. We explicitly demonstrate the errors which result from assuming these timescales are equal, which is only true at steady state. We also derive a new response function which allows the calculation of age distributions and timescales for well-mixed reservoirs away from steady state, and differentiate between timescales based on gross and net fluxes. These theoretical results are particularly important to tracer-calibrated "box models" currently used to study the carbon cycle, which usually approximate reservoirs as well-mixed. We then apply the results to the important case of anthropogenic CO₂ in the atmosphere, since timescales describing its behavior are commonly used but ambiguously defined. All relevant timescales, including lifetime, transit time, and adjustment time, are precisely defined and calculated from data and models. Apparent discrepancies between the current, empirically determined turnover time of 30–60 years and longer model-derived estimates of expected lifetime and adjustment time are explained within this theoretical framework. We also discuss the results in light of policy issues related to global warming, in particular since any comparisons of the "lifetimes" of different greenhouse gases (CO₂, CH₄, N₂O, CFC's etc.) must use a consistent definition to be meaningful.

1. Introduction

A number of timescales are being used in both scientific and policy contexts to describe the behavior of greenhouse gases in the atmosphere. However, precise definitions of timescales such as lifetime, age, and residence time are not being used, leading to confusion over how to calculate these numbers, what they mean, and how they relate to each other. For example, the report of the Intergovernmental Panel on Climate Change (IPCC, Houghton et al., 1990) defines lifetime for CO₂ as the time required for the atmosphere to adjust to a future equilibrium state if emissions

change abruptly. However, since models indicate this adjustment does not take place with a single *e*-folding time, some studies conclude that CO₂ has several different lifetimes (Edmonds et al., 1992), while others have used different methods to derive "best estimates" (Rodhe, 1990; Lashof and Ahuja, 1990), generally on the order of centuries.

In addition it has been pointed out (Victor, 1990; Moore and Braswell, 1994) that these "lifetimes", which are on the order of a century, are not consistent with data on emissions and atmospheric concentrations. The current removal timescale of CO₂ must be on the order of a few decades to achieve a mass balance based on such data; this seeming contradiction has not been adequately explained in the carbon cycle literature.

In order to clarify such issues, we turn to age

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distribution theory, which offers the framework for deriving and understanding all reservoir timescales. Early research (Eriksson, 1961, 1971; Bolin and Rodhe, 1973; Nir and Lewis, 1975) laid the theoretical foundation by concentrating on steady state systems. However, since greenhouse gas levels are rising, non-steady state concepts are required. Although some important initial work (Lewis and Nir, 1978; Schwartz, 1979; Jacquez, 1985; Zuber, 1986) outlined essential features of non-steady state theory, much remains to be done in order to apply these concepts to particular geophysical systems.

In this paper we extend the previous limits of reservoir theory by deriving a new equation for the response of well-mixed reservoirs under time-dependent conditions which makes possible the calculation of age distributions and timescales for such systems. We also derive an equation expressing the relationship between fundamental timescales for any system in or out of steady state under any type of mixing conditions. This treatment offers new insight into non-steady state reservoir theory; e.g., we show the turnover time does not generally equal the transit time, a fact noted by Schwartz (1979), but explicitly quantified here. In addition, we differentiate between timescales applicable to reservoirs through which material passes only once and reservoirs which, as part of a cycle, continuously exchange material with other parts of the system.

These results are applicable to any well-mixed reservoir which is part of a non-steady state system model, making them potentially useful in the analysis of many geophysical systems. We apply them here to the specific case of anthropogenic CO₂ in the atmosphere, precisely defining all commonly used timescales and showing how they are related. We then calculate the turnover time directly from historical emissions and concentration data. We estimate the transit time, mean age, lifetime and adjustment time from data and model results.

These calculations are useful in policy contexts related to global warming. We discuss their application to the question of responsibility for past emissions and to the interpretation of Global Warming Potentials. Furthermore, comparisons of the lifetimes of different greenhouse gases (CH₄, N₂O, CFCs, CO₂, etc.) must use consistent, rigorous definitions to avoid erroneous con-

clusions. These results also aid in interpreting timescales derived from model investigations of the carbon cycle.

Symbols and definitions

t	chronological time
τ	age (time since entry into a reservoir)
T	transit time (time between entry into and exit from a reservoir)
$M(t)$	total mass in a reservoir
$F(t)$	total flux exiting a reservoir
$I(t)$	total flux entering a reservoir
$\Psi(\tau, t)$	age distribution of mass in a reservoir (particles/time)
$\Phi(T, t)$	transit time distribution of flux exiting a reservoir (particles/time ²)
$\Theta(T, t)$	transit time distribution of flux entering a reservoir (particles/time ²)
$\tau_o(t)$	turnover time
$\tau_a(t)$	mean age
$\tau_i(t)$	mean transit time of flux exiting a reservoir
$\tau_{exp}(t)$	expected lifetime (mean transit time) of flux entering a reservoir
$\tau_{adj}(t)$	adjustment time; average time necessary for reservoir to reach equilibrium after emissions are shut off following some emissions scenario
$H(\tau, t)$	impulse response function
$G(\tau)$	Green function (impulse response function for a linear system)

2. Reservoir timescales

To derive the timescales of interest, we first define three related variables which aid in describing the passage of material through a reservoir: “ t ” refers to chronological time; “ τ ” to age, or time elapsed since entry into a reservoir; and “ T ” to transit time, or total time between entry into and exit from a reservoir. Using these variables, three fundamental distributions can be defined for any population: (1) the distribution with respect to age of all particles within the reservoir, denoted $\Psi(\tau, t)$; (2) the distribution with respect to transit time of particles leaving the reservoir, denoted $\Phi(T, t)$; and (3) the distribution with respect to transit time of particles entering the reservoir, denoted $\Theta(T, t)$. Since the $\Theta(T, t)$ distribution

indicates the number of particles entering a reservoir at time t with transit time T , it requires knowledge of the future response of the system to the input flux. Note also that since the age of a particle at exit is by definition equal to its transit time, the transit time distribution of flux out of a reservoir, $\Phi(T, t)$, may also be written as the age distribution of flux out, $\Phi(\tau, t)$.

As an example, for the human population $\Psi(30, 1993)$ is the total number of 30 year olds alive in 1993, $\Phi(30, 1993)$ is the total number of 30 year olds that die in 1993, and $\Theta(30, 1993)$ is the total number of people born in 1993 who will die when they are 30 years old. From these distributions the total mass of a reservoir $M(t)$, total outgoing flux $F(t)$, and total input flux $I(t)$ are:

$$M(t) = \int_0^\infty \Psi(\tau, t) d\tau,$$

$$F(t) = \int_0^\infty \Phi(T, t) dT,$$

$$I(t) = \int_0^\infty \Theta(T, t) dT.$$

Four basic reservoir timescales can now be defined:

turnover time

$$\tau_o(t) = \frac{1}{k_o(t)} = \frac{M(t)}{F(t)} \quad (1a)$$

mean age

$$\tau_a(t) = \int_0^\infty \tau \frac{\Psi(\tau, t)}{M(t)} d\tau \quad (1b)$$

mean transit time

$$\tau_t(t) = \int_0^\infty T \frac{\Phi(T, t)}{F(t)} dT \quad (1c)$$

expected lifetime

$$\tau_{\text{exp}}(t) = \int_0^\infty T \frac{\Theta(T, t)}{I(t)} dT. \quad (1d)$$

The turnover time as defined here is best interpreted as the inverse of the fraction of material removed from the reservoir per unit time, $k_o(t)$. It may therefore be thought of as the instantaneous removal timescale. The mean age is calculated by

averaging τ over the normalized age distribution, $\Psi(\tau, t)/M(t)$. The mean transit time (sometimes referred to as "residence time") is defined here as the average transit time of the outgoing flux and is calculated by averaging T over the normalized distribution $\Phi(T, t)/F(t)$. In terms of the human population, these timescales correspond to the average age of all people alive today and the average age of all people dying today, respectively. The expected lifetime indicates the average time necessary for removal of a single pulse of input into a reservoir at time t , and is analogous in population dynamics to the average expected lifetime of a group of people born at the same time (Kamerud, 1989). Note that both mean transit time and expected lifetime are average transit times; the first measures the average transit time of the outgoing flux, while the second is averaged over the incoming flux (Lewis and Nir, 1978).

The four timescales defined in eq. (1) are completely general. Their relations to one another, however, depend on whether the system is in a steady or non-steady state. For example, Eriksson (1961, 1971) and Bolin and Rodhe (1973) showed that $\tau_t = \tau_o$ for stationary states, i.e.,

$$\Psi(\tau, t) = \Phi(\tau, t) = \dot{M}(t) = 0.$$

However, for non-steady state systems a more general expression relating τ_t , τ_o , and τ_a is required. We derive such an expression by considering the mass continuity equation, fundamental to population dynamics (Rubinow, 1975):

$$\frac{\partial \Psi(\tau, t)}{\partial t} = -\frac{\partial \Psi(\tau, t)}{\partial \tau} - \Phi(\tau, t). \quad (2)$$

The left-hand-side of (2) is the time rate of change of the population of any age group τ . This must equal the divergence of the population "flow" at that age minus the death rate of that age group. (Note that integrating (2) over all ages τ from 0 to ∞ yields the familiar continuity equation for the total reservoir: $\dot{M}(t) = I(t) - F(t)$.) Multiplying the continuity equation by τ and integrating over all ages gives an expression that relates turnover time, mean age and mean transit time for non-equilibrium as well as equilibrium systems. We find

$$\tau_o(t) - \tau_t(t) = \frac{\dot{M}(t) \tau_a(t) + \dot{\tau}_a(t) M(t)}{F(t)}. \quad (3)$$

For stationary systems, the mean transit time of the outgoing flux and the turnover time are rigorously equal, as expected. However for non-stationary systems (i.e., $\dot{M}$ and/or $\tau_a \neq 0$), they are not equal, a fact which can have important geophysical consequences (Gaffin and O'Neil, 1994). Although this inequality has been pointed out by Schwartz (1979), eq. (3) is, to our knowledge, the first equation explicitly quantifying the relationship between timescales away from steady state.

3. Distributions for well-mixed reservoirs without return fluxes

In order to calculate the timescales defined in eq. (1) for a particular reservoir, one must be able to calculate the Ψ , Φ , and Θ distributions. These distributions require convolving an emissions history with a response function describing the fate of material in the reservoir (Niemi, 1977; Lewis and Nir, 1978; Schwartz, 1979; Zuber, 1986). We therefore seek a general form for such a function, which we denote $H(\tau, t)$, defined as the fraction of an emission at time t remaining in a reservoir as a function of age. Since we intend to investigate the behavior of CO₂ in the atmosphere, which may be considered well-mixed (Ekdahl and Keeling, 1973), we can use the definition of a well-mixed reservoir to derive $H(\tau, t)$.

For the sake of clarity, we will begin by ignoring the possibility that a gas molecule may return to the atmosphere after exiting it, as is the case for CO₂. The existence of a return flux complicates the concept of the age of a molecule, and we will consider first only irreversible removal. These results will be of interest in themselves and will also provide the basis for defining timescales for a reservoir with return fluxes, discussed in Section 4.

A key point related to well-mixed reservoirs is that removal processes cannot distinguish between particles, i.e., all particles, regardless of age, have an equal probability of being removed per unit time (Bolin and Rodhe, 1973). This probability per unit time is given by $k_o(t)$, the inverse of the turnover time, which is the fraction of the mass in the entire reservoir removed per unit time. Therefore the removal of any particular emission into a well-mixed reservoir without return fluxes is dictated by the turnover time, a quantity which may be

estimated from mass and flux measurements (eq. (1a)). The fractional removal of an emission can be written in terms of H as $-H'(\tau, t)/H(\tau, t)$, where the prime indicates a derivative with respect to age. The numerator is the fraction of an emission which exits the reservoir as a function of age, while the denominator is the fraction which remains. Thus the ratio gives the probability of removal of a particular emission; since the reservoir is well-mixed this ratio must equal the removal probability of the whole reservoir:

$$\frac{-H'(\tau, t)}{H(\tau, t)} = k_o(t + \tau). \quad (4)$$

Note that eq. (4) does not necessarily imply a linear system; the form of $k_o(t + \tau)$ may be such that it incorporates nonlinearities. Solving (4) for $H(\tau, t)$,

$$H(\tau, t) = \exp \left[- \int_t^{t+\tau} k_o(t') dt' \right]. \quad (5)$$

Eq. (5) is the generalized response function for well-mixed reservoirs without return fluxes, and indicates that the fraction of an emission at time t which will remain τ years later is dictated by the turnover time operating on the reservoir over the time period t to $t + \tau$. The turnover time is therefore a critical parameter which expresses the action of the system on both the bulk mass and on any particular emission. Note for a constant turnover time, eq. (5) reduces to $H(\tau, t) = \exp[-k\tau]$, as would be expected for a simple, single-process decay. Importantly, since the turnover time is indirectly measurable (through mass and flux measurements), the response function and all distributions and timescales describing past and present behavior may be calculated for well-mixed reservoirs even when direct age measurements cannot be made.

This finding makes it possible to use $H(\tau, t)$ in conjunction with any emissions history $I(t)$ to define the reservoir distributions and calculate the timescales presented in Section 1. The age distribution of mass in a reservoir is

$$\begin{aligned} \Psi(\tau, t) &= I(t - \tau) H(\tau, t - \tau) \\ &= I(t - \tau) \exp \left[- \int_{t-\tau}^t k_o(t') dt' \right]. \end{aligned} \quad (6)$$

Eq. (6) indicates that the number of particles in the

reservoir with age τ is just the number of particles emitted τ years ago which remain today. The transit time (or age) distribution of mass leaving a reservoir can be derived from eq. (6) and the continuity equation (eq. (2)):

$$\begin{aligned}\Phi(T, t) &= -I(t - T) H'(T, t - T) \\ &= I(t - T) k_o(t) \exp \left[- \int_{t-T}^t k_o(t') dt' \right].\end{aligned}\quad (7)$$

Eq. (7) indicates that the number of particles exiting the reservoir at time t with transit time (or age) T is just the number of particles which entered T years ago with expected transit time T . Also, the $\Phi(T, t)$ distribution differs from the $\Psi(\tau, t)$ distribution only by a factor of $k_o(t)$. The normalized distributions, $\Phi(T, t)/F(t)$ and $\Psi(\tau, t)/M(t)$, are therefore equal, since $F(t) = k_o(t) M(t)$. Thus, as is seen from eqs. (1b) and (1c), for well-mixed reservoirs without return fluxes $\tau_a(t) = \tau_i(t)$ (Schwartz, 1979). This reflects the fact that the removal processes acting on a well-mixed reservoir cannot discriminate between particles of different ages and so the gross flux out must have the same average age as that of the bulk mass.

The transit time distribution of the input flux is:

$$\begin{aligned}\Theta(T, t) &= -I(t) H'(T, t) \\ &= I(t) k_o(t + T) \exp \left[- \int_t^{t+T} k_o(t') dt' \right].\end{aligned}\quad (8)$$

This equation is similar to eq. (7), but relates to the present and future; it indicates the number of particles entering the reservoir at time t with transit time T , and requires knowledge of the future removal rate of the system.

The distributions defined in eqs. (6), (7) and (8) can be used to calculate the timescales defined in eq. (1), as long as the turnover time of the system as a function of time is known.

4. Distributions for well-mixed reservoirs with return fluxes

The equations defining distributions presented in Section 3 apply to reservoirs through which material passes once and does not return after exiting. Carbon reservoirs, on the other hand, are part of a cycle and continuously exchange their con-

tents. Carbon added to the atmosphere does not therefore simply exit never to return, but redistributes among all the reservoirs, during which time it may pass through the atmosphere many times.

Because of this behavior, the addition of an amount of CO_2 to the atmosphere presents a choice in timescales. Eqs. (4)–(8) may be applied directly and return fluxes ignored, yielding timescales describing the behavior of extra CO_2 molecules passing irreversibly through the atmosphere. Or, the impulse response function $H(\tau, t)$ may be redefined to allow for return fluxes. In this case, the resulting distributions and timescales describe the transition of the atmospheric CO_2 mass from its perturbed to its equilibrium level as the extra molecules distribute throughout the system.

If return fluxes are ignored and the first method chosen, the turnover time used in eqs. (4) and (5) to derive the mass response function $H(\tau, t)$ is calculated from the total CO_2 mass ($M(t)$) and the gross flux out of the atmosphere ($F(t)$) as described in Section 3. As pointed out in the IPCC report (Watson et al., 1990), this turnover time for atmospheric CO_2 is about 4 years. The expected lifetime (reflecting an average turnover time) is therefore also about four years and answers the question: how long, on average, does it take for CO_2 molecules to pass once through the atmosphere before being taken up by the oceans or terrestrial biosphere?

However, the more interesting question is: how long does it take an amount of CO_2 added to the atmosphere to distribute throughout all the reservoirs in the system? Answering this question requires considering the *net* flux of CO_2 out of the atmosphere, thereby accounting for CO_2 which may have previously left the atmosphere but is now returning. The impulse response function $H(\tau, t)$ must be redefined as the fraction of a pulse of CO_2 mass added to the atmosphere remaining as a function of age, regardless of how many times individual molecules making up that extra mass have passed through the atmospheric reservoir since they were emitted.

This function cannot be derived using eqs. (4) and (5) as can the removal function for reservoirs without return fluxes since the turnover time of the reservoir reflects only gross removal. Instead, $H(\tau, t)$ must be estimated from model experiments

(Maier-Reimer and Hasselmann, 1987; Sarmiento and Orr, 1992; Caldeira and Kasting, 1993). In general, $H(\tau, t)$ is a function of time, since the carbon cycle is nonlinear. However, if a linear approximation is made, it is constant in time and is termed the Green's function of the system. With the response function the distributions in eqs. (6)–(8) and the relevant timescales may be calculated.

Interpreting $H(\tau, t)$ for the redistribution of CO₂ in terms of a transit time distribution of specific molecules requires examining the processes governing transfer between reservoirs. Consider, for example, the addition of a pulse of CO₂ to the atmosphere. Assume the molecules making up this pulse are all labeled so that their individual fates may be tracked (e.g., they may be radioactive ¹⁴CO₂ molecules if fractionation effects during transfers between reservoirs are ignored). If the carbon cycle were linear, with all fluxes proportional to the reservoir masses, the decay of the labeled molecules as they redistributed throughout the system would be exactly the same as the decay of the excess CO₂ mass in the atmosphere (Jacquez, 1985). This is a consequence of the linearity (and proportionality) of the fluxes, which allows one to consider the unlabeled (equilibrium) molecules separately from the labeled (excess) molecules. The net change in the number of unlabeled molecules in the atmosphere will always be zero; thus the decay of the excess mass is entirely attributable to the decay of the labeled molecules.

However the carbon cycle is nonlinear, in particular through the oceanic buffer factor. Even if the approximation of a constant buffer factor is made, the flux from the ocean to the atmosphere is linear but not proportional to the total carbon mass in the ocean (Rodhe and Björkström, 1979). As a result, the equilibrium partitioning of carbon among all reservoirs is a function of the total carbon in the system in such a way that the addition of CO₂ to the system shifts proportionately more carbon to the atmosphere. Therefore when a labeled pulse of CO₂ is added to the atmosphere, the number of unlabeled molecules increases as carbon shifts from the oceans to the atmosphere. At the same time, the number of labeled molecules decreases as they distribute throughout the other reservoirs. The result is that the decay of the excess atmospheric mass will be slower than the decay of

labeled molecules since it will also include the effect of molecules shifting from the oceans to the atmosphere as result of the increase in total carbon mass in the oceans. It is for this reason that the removal of atmospheric ¹⁴CO₂ cannot be equated with the decay of anthropogenic CO₂.

Thus any impulse response function derived from a carbon cycle model including the buffer factor and/or any other nonlinearities must be defined as the fraction of a particular emission remaining in the atmosphere as a function of age, where that fraction may be made up not only of molecules which were part of the original emission but also of molecules which have transferred to the atmosphere as a result of that emission. The concepts of "age" and "transit time" of molecules making up this response function must take on a more liberal definition. The age of a labeled molecule remains straightforward: the time since emission into the atmosphere, regardless of the number of times it has passed through the atmospheric reservoir. The age of unlabeled excess CO₂ which has shifted to the atmosphere as a result of an emission is the time since that emission. In this way, the transit time distribution of a pulse of CO₂ (eq. (8)) consists of the net number of molecules in the atmosphere as a result of the pulse which are exiting the atmosphere with age τ .

5. Timescales for atmospheric CO₂ from data and models

The results of the preceding sections may be applied to CO₂ by using historical emissions and concentration data and impulse response functions derived from carbon cycle models.

As pointed out in the previous section, the turnover time for the entire mass of atmospheric CO₂ is about 4 years and corresponds to considering only the gross flux out of the atmosphere. However, in relation to global warming it is the behavior of the excess CO₂ above the pre-industrial value (M_{eq}) of 280 ± 10 ppm (Watson et al., 1990) which is of more interest. Following eq. (1a), we therefore define the turnover time of excess CO₂ as $\Delta M(t)/\Delta F(t)$, where $\Delta M(t) = M(t) - M_{eq}$ is the excess CO₂ and $\Delta F(t) = F(t) - R(t)$ is the net flux of CO₂ out of the atmosphere ($F(t)$ is the gross flux out and $R(t)$ the return flux from all other reservoirs). In practice,

$\Delta F(t)$ can be computed from the mass conservation equation for anthropogenic CO_2 , $\Delta F(t) = I_a(t) - \Delta \dot{M}(t)$, where $I_a(t)$ is the anthropogenic emissions data.

Fig. 1 shows the turnover time, ($\tau_{o,\text{data}}$, plotted as a dotted line over the period 1959–1989) calculated from data on anthropogenic CO_2 emissions from industrial activity (Keeling, 1991; Marland and Boden, 1991) and land use change (Houghton, 1991), and atmospheric levels (Keeling and Whorf, 1991). It has been relatively con-

stant at about 30 years since at least 1960, with superimposed interannual fluctuations corresponding to El Niño events (Quinn et al., 1987) reflecting a decrease in net CO_2 uptake by oceanic and terrestrial processes combined (Keeling et al., 1989). This constancy mirrors the behavior of the airborne fraction, a measure of excess CO_2 removal closely related to the turnover time (Ekdahl and Keeling, 1973). Using industrial emissions alone yields a turnover time of about 60 years over the same period. This 30–60 year

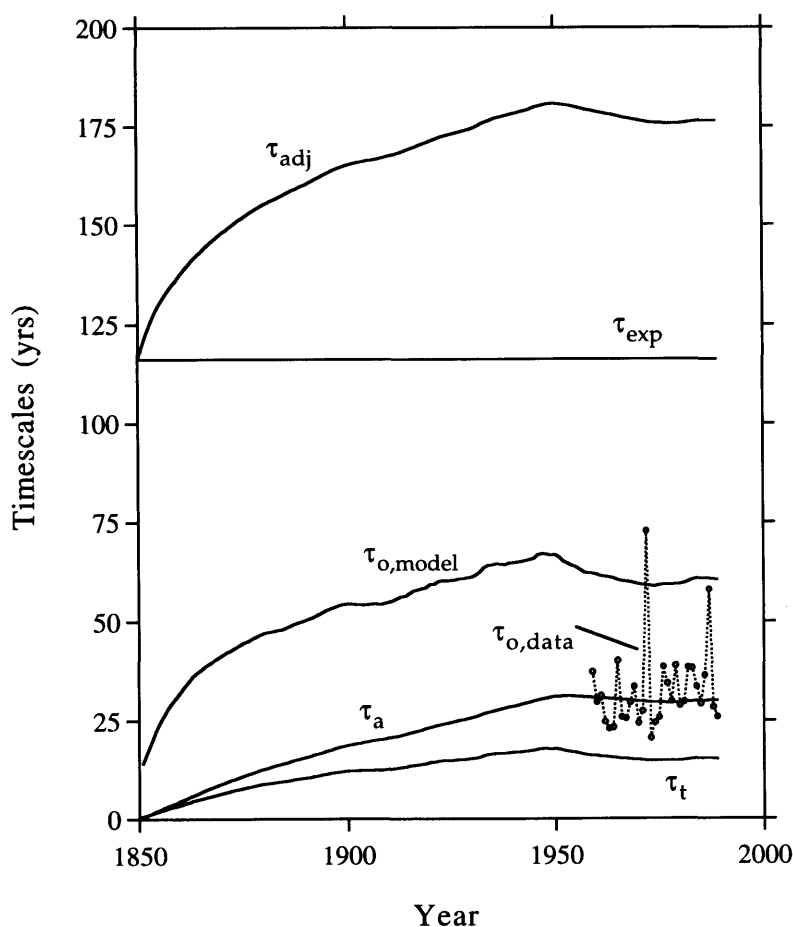


Fig. 1. Open circles connected by a dotted line show the turnover time of excess CO_2 in the atmosphere calculated from concentration data and estimates of emissions from fossil fuel burning and land use change. Solid lines are reservoir timescales calculated by modeling CO_2 removal with the impulse-response function derived from the oceanic GCM of Maier-Reimer and Hasselmann (1987) and using the same anthropogenic emissions data. τ_o , τ_a , and τ_t are the turnover time, mean age, and mean transit time, respectively. τ_{exp} is the expected lifetime of CO_2 entering the atmosphere, while τ_{adj} is the average time necessary for the atmosphere to reach equilibrium after emissions are shut off.

value is a model-independent estimate of the instantaneous net removal rate of excess CO₂.

Although the turnover time of the excess is much larger than the turnover time of the total CO₂ mass, this does not imply that there are two distinct populations of CO₂ molecules with different behaviors. The fraction of CO₂ molecules removed from the atmosphere each year is always given by the inverse of the turnover time of the whole mass, regardless of whether the molecules are anthropogenic or not. The turnover time of the excess mass is larger because it is based on the net removal; net removal is slower because it takes into account the return flux from other reservoirs.

To calculate all other timescales, model-derived impulse response functions are required in order to define the relevant age or transit time distributions. The calculation is, in general, complicated by the nonlinearity of the system; i.e., each emission must be convolved with a different response function due to nonlinearities in ocean uptake (Caldeira and Kasting, 1993) and the terrestrial biosphere (Moore and Braswell, 1994). For simplicity, we will take an impulse-response function derived from the abiotic oceanic general circulation model of Maier-Reimer and Hasselmann (1987) and assume the carbon cycle is linear. While this approximation clearly makes the numerical values of derived timescales only approximate, it allows a more transparent demonstration of timescale calculations. The response function, which was determined by a multiple-exponential fit to the response of the model to a spike in emissions, may be considered a Green's function and is given by:

$$G(\tau) = a_c + \sum_{i=1}^4 a_i e^{-k_i \tau}, \quad (9)$$

where $a_{0-4} = (0.131, 0.201, 0.321, 0.249, 0.098)$ and $k_{0-4} = (0, 1/362.9, 1/73.6, 1/17.3, 1/1.9)$. All distribution-based timescales may be calculated using this function combined with estimates of past CO₂ emissions in eqs. (6)–(8) (assuming $H(\tau, t) = G(\tau)$). In addition, $G(\tau)$ may be used in a convolution integral to calculate the atmospheric CO₂ mass and flux out of the atmosphere (Wigley, 1991), and therefore the turnover time.

Results are shown in Fig. 1. It is apparent that the model's turnover time is too long compared to the real atmosphere; i.e., the model removes a smaller fraction of the anthropogenic CO₂ mass

than does the real carbon cycle and therefore does not reproduce well observed levels of atmospheric CO₂. This fact was discussed by Wigley (1991; see also Wigley and Raper, 1992) and is likely due to the model's lack of biological processes.

The mean age of anthropogenic CO₂ is about 30 years, where age is defined as the average time since the causative emission for all CO₂ in the atmosphere which was either directly released by human activity or is in the atmosphere as a result of such releases (see Section 4). The mean transit time is about 15 years, and indicates the average age of the net flux of excess CO₂ out of the atmosphere. The transit time is different from the mean age even though the reservoir is well-mixed since these timescales are based on net fluxes. The shorter transit time reflects the form of $G(\tau)$ as given in eq. (9), which indicates that net removal of CO₂ is fastest just after emission and slows with age; thus the transit time distribution of outgoing flux is biased toward "young" CO₂. The age and transit time are 25 and 13 years if emissions from fossil fuel burning alone are used.

Fig. 1 indicates that all reservoir timescales have become asymptotically constant. This behavior is due to the exponential form of the emissions forcing and removal function, which causes $M(t)$ to increase exponentially and the normalized age distribution ($\Psi(\tau, t)/M(t)$) to become asymptotically stable. τ_a reflects the average age of the predominantly "young" anthropogenic CO₂ mass; its value is a function of both the emissions and the Green's function. Likewise, the turnover time becomes constant since the removal rate as a function of age is time-invariant and the age distribution becomes stable.

It is because of the asymptotically constant behavior of the turnover time observed over the past several decades that a single exponential response function with a decay constant approximately equal to the asymptotic value of $\tau_o(t)$ can reproduce the atmospheric data well. Fig. 2 shows the results of a model using such a single-exponential removal function for the years 1850–1989. The model produces the atmospheric CO₂ level ($M(t)$) from the equation $\Delta \dot{M}(t) = I_a(t) - \Delta M(t)/\tau_o$, where $\Delta M(t) = M(t) - M_{eq}$. $I_a(t)$ is anthropogenic CO₂ emissions, taken here as industrial emissions and emissions from land use change. M_{eq} , τ_o , and $M(1850)$ are treated as free parameters and allowed to assume values for which the best least-

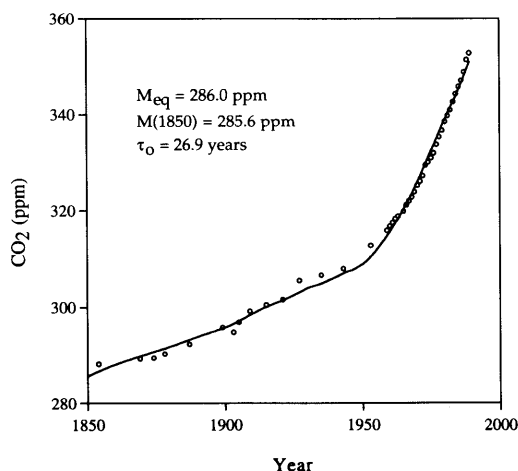


Fig. 2. Open circles are data on atmospheric CO_2 levels from Mauna Loa (1959–1990) and from the Siple ice core (Neftel et al., 1991). The solid line is the output of a model which removes CO_2 from the atmosphere with a single, constant removal rate equal to the inverse of the turnover time. As noted in the text, the quality of the fit does not necessarily imply that there is one constant removal timescale acting on atmospheric CO_2 ; rather, it is more likely that the net action of several removal processes has become constant as a result of the exponential form of emissions (see Fig. 1).

squares fit of $M(t)$ to the data is achieved. This fit does not imply that the Green's function of the real carbon cycle is a single exponential. Rather, as indicated by Fig. 1, it means only that the turnover time of the system has become constant, which is a result of the exponential form of the emissions history. Using industrial emissions alone produces a best-fitting output with $\tau_0 = 42.5$ years and $M_{\text{eq}} = 295.3$ ppm.

6. The atmospheric lifetime of CO_2

The "lifetime" of CO_2 has been estimated by various techniques and yielded values ranging from decades to centuries. Each estimate also defined "lifetime" in different ways.

In reservoir theory, the expected lifetime of a particular emission has a precise definition given by eq. (1d). It is the average transit time of flux entering the reservoir, calculated using the impulse response function as in eq. (8). The impulse response function does not have to be a single

exponential decay in order to calculate this average; the single exponential form is merely convenient since the decay constant turns out to equal the average transit time. The only requirement for calculating a true average is that the integral in eq. (1d) converge. Unfortunately, impulse-response functions from carbon cycle models do not generally converge over timescales of centuries, as indicated by the a_0 term in eq. (9).

Several strategies have been used to circumvent this problem. Lashof and Ahuja (1990) chose to assign the a_0 term an arbitrary decay constant, effectively discounting future CO_2 concentrations, and then calculated a 230-year average transit time implied by this new function. Moore and Braswell (1994) and Rodhe (1990) ignored the long-term response, focusing instead on short-term removal and quoting a "single half-life" and an " e -folding ($1/e$) time" of about 30 and about 120 years, respectively. Edmonds et al. (1990) concluded that because impulse response functions indicate that CO_2 is removed more and more slowly with age, the "lifetime" (which they define as the instantaneous net removal rate of a particular emission) is a function of time and ranges from decades to centuries. The IPCC report (Watson et al., 1990; Shine et al., 1990) offers no rigorous definition of lifetime; for the purpose of defining Global Warming Potentials, it instead presents integrations of impulse-response functions over several finite time intervals. Each of these estimates has its own strengths and weaknesses. Taken together, however, they create confusion over what "lifetime" means, how to calculate it, and how it relates to other timescales.

Eq. (1d) suggests an alternative definition which is consistent with the traditional concept of lifetime as an average transit time. The distribution of transit times used in its calculation given by eq. (8) automatically excludes that fraction of material which never leaves the reservoir. Thus, the expected lifetime of CO_2 may be calculated as the average transit time of all CO_2 which eventually leaves the atmosphere. For the function $G(\tau)$ given by eq. (9), this average is 116 years. We may therefore say that according to eq. (9), for any emission of CO_2 to the atmosphere, a fraction equal to a_0 will effectively remain indefinitely and the rest will stay in the atmosphere an average of 116 years.

It is important to note that this expected lifetime calculated from the Green's function of a carbon

cycle model is not equal to the time required for the excess atmospheric CO₂ mass to adjust to a new equilibrium after emissions are shut off. This "adjustment time" may be defined similarly to the expected lifetime: it is the average transit time of excess CO₂ already in the atmosphere when emissions cease, measured from the moment emissions are shut off, and excluding that fraction which remains indefinitely. The IPCC report describes lifetime as equivalent to the adjustment time (Watson et al., 1990); however, it can be shown (Tubiello et al., submitted) that even if the carbon cycle is assumed linear, its adjustment time may be 50–90% greater than the expected lifetime of a single emission added at equilibrium. Fig. 1 shows the evolution of the adjustment time of the model represented by eq. (9) as a function of the emissions history. Since it is an average transit time, this timescale is based on eqs. (1d) and (8), except that the response function must be the response of the entire excess CO₂ mass to the emissions shut-off, not an impulse response function. For a detailed discussion of adjustment time, see Tubiello et al. (submitted). Fig. 1 indicates that according to eq. (9), if emissions ceased in 1990 it would take an average of 175 years (not 116 years) to adjust to the new equilibrium level.

7. Discussion and conclusions

A growing array of timescales are being extracted from carbon cycle models and data and their relationships have not been clear. In particular, model atmospheres forced with historical emissions data require a 30–60 year turnover time to match data on atmospheric levels, while much longer timescales are being extracted from multiple exponential impulse-response functions derived from carbon cycle models. This discrepancy has not been adequately explained and is causing confusion in literature concerned with the atmospheric "lifetime" of anthropogenic CO₂ (Victor, 1990; Edmonds et al., 1992). Considering the policy implications of such numbers, it is important that their meanings and relationships be fully clarified.

We suggest that the only basis for such a clarification is age-distribution theory for reservoir timescales away from steady state. This formalism can unify seemingly diverse timescales and clearly distinguishes between timescales calculated from gross and net fluxes. We have also used reservoir theory to derive a more general result which quantifies the difference between turnover time and transit time away from steady state for any mixing process. Many geophysical studies implicitly equate the two numbers and use the turnover time when they actually need the transit time. Our eq. (3) will be useful in assessing the error introduced by such an assumption.

Clearly defined timescales should also be useful in addressing policy issues related to global warming. For example, it has been suggested that cumulative surviving emissions disaggregated by region be used to proportion responsibility for the present accumulations of greenhouse gases (Smith, 1991; Parikh, 1992). The IPCC "lifetime" has been used to choose the necessary period of integration. However, the most appropriate period for determining surviving emissions is given by the average age. For CO₂, regional emissions data over the past 30 years may suffice.

In addition, a major program determining the atmospheric "lifetimes" of halocarbons (Prinn et al., 1983; WMO, 1992) has long been underway. Reservoir timescale theory applies equally well to halocarbons, nitrous oxide (Watson et al., 1990) and methane (Prather, 1994; Osborn and Wigley, 1994), and any comparison of the atmospheric lifetimes of different greenhouse gases must use a consistent definition to be meaningful. Also, careful consideration will need to be given to the choice and use of a particular timescale for the purpose of developing "Global Warming Potential" indices. The model-independent turnover time (τ_o), as a measure of the short-term removal rate of CO₂, may prove useful as part of an alternative measure of warming potentials. Additionally, timescale dynamics in response to changes in emission patterns and nonlinear climate effects must be understood to ensure their proper use in any economic index.

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