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Kinetics of the Atmosphere Modeled with Multi-box Systems

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Abstract

An online simulation application is developed for multi-box simulations of atmospheric carbon dioxide, with different kinetics. Examples are shown with real data. It is shown that a first-order kinetics two-box model is the most adequate for the data. Using real data of CO_2 emissions and concentrations, it is found that the residence time of CO_2 molecules in the atmosphere is 52 years, which is a value about an order larger that ones from literature. On the basis of this value, it is estimated that 60% of anthropogenic CO_2 is still in the atmosphere.

Keywords: Atmosphere, carbon dioxide, residence time, kinetics

Introduction

One of the major issues in the climate discussion is the kinetics of molecules. How fast do molecules go to what places? It is relevant because if we can determine the speed at which the carbon dioxide that is injected in the atmosphere by our emissions disappears from where it can do harm, we will get an idea of the magnitude of the problem, if there exists a problem at all.

It is often intrinsically assumed that the concentrations at various places (atmosphere, oceans, biomass, etc.) are the result of earlier fluxes in the past. For instance, if we imagine that the atmosphere retains all carbon dioxide injected into it, the concentration in the atmosphere would be the integral of these injections (or sum of yearly injections if the analysis is done on basis of a discrete model). Or, generally, if we consider a ’box’ B, for instance the atmosphere, and imagine it of constant volume and well-mixed and into which fluxes come in and from where fluxes go out, once again the concentration change up to a certain point in time is the result of cumulative fluxes of the past, in other words, the concentration C_n in box B at year n given by the concentration at year 0 plus the sum of flux balances since then,

$$C_n = C_0 + \sum_{i=1}^{n} \left(F_{\text{in},i} - F_{\text{out},i}\right)$$

This is the mental picture most climate researchers probably have of the atmospheric system. Research then consists of these main questions: 1) How much CO_2 was injected each year into the system in the past? 2) Effectively how many boxes exist in the system? In other words, how many homogeneously fixed-volume subsystems exist? 3) What are the kinetics of exchange of molecules between these boxes? The author himself having presented one such analysis, in which the conclusion was that, a) Most anthropogenic CO_2 has already left the atmosphere, b) The idea of a constant 280 ppm CO_2 concentration in pre-industrial times is untenable (Stallinga 2023). Yet, before we continue, it has to be pointed out that the system might also be thought of as one in which the concentrations in a box, including that of the atmosphere, are simply a function of many parameters of nature, for instance ocean temperature, cosmic radiation, etc., *except* past fluxes. The fluxes then simply those that are needed to make this programmed concentration occur. In other words,

$$F_{\text{in},n} - F_{\text{out},n} = C_n - C_{n-1}$$

In terms of mathematical modeling this makes no difference; the above two equations are identical. Yet, the mental picture is completely different; causes are commonly imagined on the right side of the equation while effects on the left side. Take for example the effect of adding an amount of CO_2 to a certain box, for instance by emissions into the atmosphere. In the first picture it will take time to process this added CO_2, the time it takes to reach a new equilibrium called the adjustment time. In the second picture the removal of the surplus CO_2 might be considered effectively instantaneous. An amount is simply added to $F_{\text{out},n}$ equal to the emissions in order to result in the ’correct’ C_n that year. C_n is given by nature’s desire, the fluxes are to make it so.

After having made this important observation we are anyway going to do a classic analysis of the system by continuing earlier work. In the discussion on the atmosphere and its effect on the climate, it is very important to use good definitions, in order to have scientists able to communicate between each other. One of the important parameters of the atmospheric system is the residence time, the average time a molecule of carbon-dioxide (CO_2) stays on average in the atmosphere. Or in general in a box that represents the cycle of CO_2 in the system. Another parameter to describe the system is the adjustment time, generally described as the characteristic time it takes for a disturbance, an off-equilibrium situation, to be processed and to settle back in an equilibrium.

In a recent publication (Stallinga 2023) we showed that for a system described by two boxes – for instance the atmosphere and a sink, maybe the ocean – with linear kinetics (so-called ’first-order kinetics’, with fluxes out of a box proportional to the contents in a box), the adjustment time is always shorter than the residence time of CO_2 molecules in any particular box. As such, since the residence time of CO_2 in the atmosphere was found to be about a decade – the IPCC uses a value of 4 years – the time to process off-equilibrium CO_2 – for instance our emission caused by fossil fuels – is less than a decade, and thus most of our CO_2 emissions have already disappeared from the atmosphere.

However, the question arises, if similar statements can still be made if we add boxes to the system (for instance dividing the ’ocean-box’ into a ’shallow-ocean-box’ and a ’deep-ocean-box’. Moreover, what if the kinetics are not linear? Even though very strange and hard to explain, what would happen if the flux out of a box is not a linear function of the contents of the box? How do residence time and adjustment time correlate then? In this work we will try to work it out, as a response to criticism to our over-simplified previous work. Examples will be given to substantiate the conclusions. Table 1 shows the meaning of the parameters used in this text.

Table 1. Important parameters used in this work

Parameter	Meaning	Unit
B_i	Name of box i	N/A
B_i	Amount in box i	Gt, ppm*, mol
F_{ij}	Flux from B_i to B_j	Gt/a
τ_i	Residence time in box B_i	a
τ	Adjustment time	a
E	Emissions	Gt/a

(*: 1 ppm = 7.76 Gt)

Multi-box systems

In these simulations we use a system that has n boxes B_i of equal and constant volume with fully-mixed homogeneous contents B_i (i ranges from 1 to n), and these are connected by fluxes F_{ij} that are given by kinetic constants k_{ij} and B_i, see Figure 1. In chemistry notation,

$$B_i \underset{k_{ji}}{\overset{k_{ij}}{\rightleftharpoons}} B_j$$

which represents the fluxes as,

$$F_{ij} = k_{ij} B_i^{\alpha}$$

[Eq:1]

The first index i is the 'from'-box and the second index j the 'to'-box. Here α is the order of the kinetics, assumed to be equal for all fluxes. (A note about the units at the end). For instance, $\alpha = 1$ for first-order (linear) kinetics (Summers and Scott 1988). The residence time τ_i of molecules in a box B_i, the average time an average molecules stays in that box before leaving to another, is given by the amount in that box divided by all the fluxes going out of it. Using the above equation we find,

$$\tau_i = \frac{B_i}{\sum_j F_{ij}} = \frac{1}{\sum_j k_{ij}} \times B_i^{1-\alpha}$$

[Eq:2]

which is a constant only for first-order kinetics ($\alpha = 1$). In which case the residence times are given by,

$$\tau_i = \frac{1}{\sum_j k_{ij}}$$

[Eq:3]

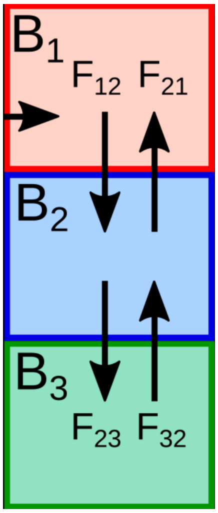


Figure 1: The 3-box general kinetics model used in this work. $\mathrm{B_i}$ are the amounts of molecules in the three boxes, $\mathrm{F_{ij}}$ are the fluxes between the boxes, and E represents external injections ('emissions').

With a conservation of quantity – no matter is lost according to Lavoisier – as our boundary condition, the total amount of matter, $\sum B_i =$ constant, and $\sum_{ij} F_{ij} - F_{ji} = 0$, the changes of the boxes B_i over time are then given by

$$\frac{\mathrm{d}B_i(t)}{\mathrm{d}t} = \sum_{j \neq i}^{n} \left(F_{ji} - F_{ij}\right) = \sum_{j \neq i}^{n} k_{ji}\left[B_j(t)\right]^{\alpha} - k_{ij}[B_i(t)]^{\alpha}$$

[Eq:4]

This indeed follows the above mentioned matter conservation boundary condition; what leaves one box enters others. To simplify the expression we will eliminate self-flux by setting all F_{ii} to zero. Also note that in equilibrium we not only require to not have changes in B_i but also to have partial balances; no net flux between any two boxes, there where theoretically we might cycle around molecules forever without changing their contents (box 1 → box 2 → box 3 → box 1, etc.). To avoid this cycling, we set for equilibrium: $F_{ij} = F_{ji}$. In that case equilibrium is defined by a situation in which,

$$\frac{B_i}{B_j} = \left(\frac{k_{ji}}{k_{ij}}\right)^{1/\alpha}$$

[Eq:5]

We have now defined all necessary math to do an analysis of various (simplified) models of the system. In what comes various models will be described. They are limited to three-box systems because they suffice to highlight the various intricacies of multi-box systems with general kinetics. Using more than three boxes would not have any added value here, while of course in reality nature consists of a multitude of boxes. Any non-infinite number of boxes is a simplification of reality. We also limit ourselves here to a system where fluxes occur only to numerically neighboring boxes. There is no flux between box 1 and box 3. We can imagine box 1 being the atmosphere, box 2 being the shallow ocean, and box 3 the deep ocean; no flux between atmosphere and deep ocean. Or a system where box 2 is the atmosphere, box 1 is the biosphere, and box 3 is the ocean; no flux between biosphere and ocean.

A first-order-kinetics two-box system

We start with the simplest model resembling simple nuclear decay, namely a two-box system $n = 2$ with a linear flux function ($\alpha = 1$) and $k_{12} \neq 0$ and $k_{21} = 0$. Once a molecule has left box 1 and entered box 2, it never comes back to box 1.

$$F_{12} = -k_{12}B_1$$
$$F_{21} = 0$$

The differential equation (Eq. [Eq:4]) with the above substitutions is

$$\frac{\mathrm{d}B_1(t)}{\mathrm{d}t} = -k_{12}B_1$$
$$\frac{\mathrm{d}B_2(t)}{\mathrm{d}t} = k_{12}B_1$$

and the solution is a simple exponential transient. For instance after creating at $t = 0$ an initial amount $\Delta B = 1$ (to be multiplied with any quantity; the scale is relative to this quantity) in box 1:

$$B_1(t) = 1 \times \exp(-k_{12}t)$$

$$B_2(t) = 1 \times [1 - \exp(-k_{12}t)]$$

Generally speaking, the characteristic time constant – 'adjustment time' τ – of a transient $B_i(t)$ of box B_i is given by the characteristic speed to reach the new equilibrium in that box, and can be expressed at any moment t in time as the total amplitude of the transient still to come divided by the instantaneous change in the box's contents,

$$\tau(t) = \frac{B_i(t) - B_i(\infty)}{\mathrm{d}B_i(t)/\mathrm{d}t} \tag{Eq:6}$$

which is constant for exponential transients (only). In this case $\tau = 1/k_{12}$.

Figure 2 shows such an exponential decay for k_{12} is 0.02/it, in other words, every iteration (it, representing a simulation time period, for instance a year) 2% of the molecules in box 1 (colored red) are transferred to box 2 (colored blue), where they stay forever. Box 2 is a 'mirror' version of box 1, and the sum of the two boxes is always equal to the sum of the starting values, in this case $B_1(t = 0) = 1$ and $B_2(t = 0) = 0$. The exponential function is shown as a dashed black curve that coincides with $B_1(t)$. The time constant is $\tau = 1/k_{12}$ = 50 it. This is also called the residence time of molecules in box 1, $\tau_1 = 1/k_{12}$ = 50 it, there where the residence time in box 2 is infinite, $\tau_2 = 1/k_{21} = \infty$. In this simulation we can imagine that the red box 1 represents the atmosphere and blue box 2 represents the (ocean) sink. At $t = 0$ we inject 1 unit of CO_2 into the atmosphere, which is captured by the ocean in a characteristic time scale of 50 it.

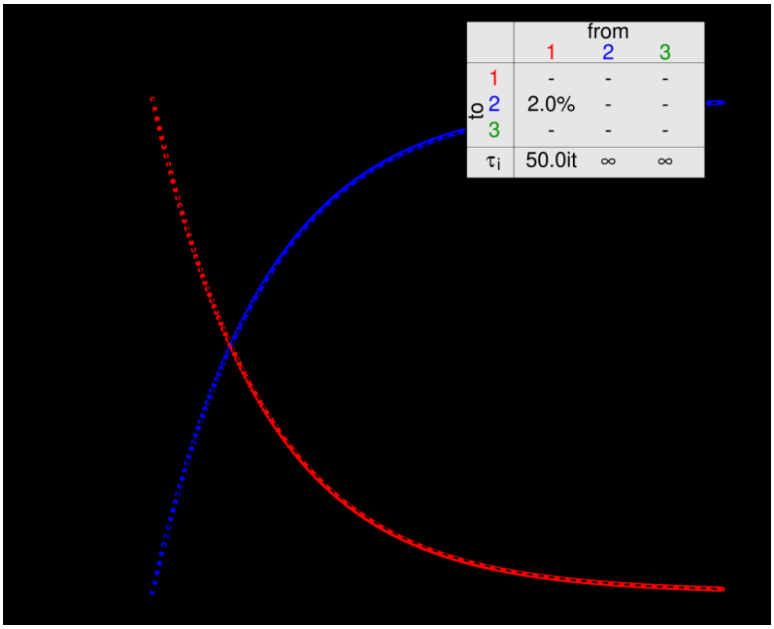


Figure 2: A two-box simulation (third box is not connected) with first-order kinetics (α = 1). This is similar to nuclear decay. Every iteration ('it') 2% of molecules in (red) box 1 are captured by the sink blue box 2 (k_{12} = 0.02/it), where they'll stay forever. The kinetic constants are shown in the legend, which also shows the residence times τ_i in each box. Box 3 (not shown), although existing, is not connected to the other boxes by any fluxes $\forall i\colon k_{i3} = 0$, and thus remains in its start value, $B_3(t) = 0$.

In the next simulation we permit molecules from the sink (blue box 2) to flow back to the atmosphere (red box 1) by setting a kinetic constant $k_{21} \neq 0$. In this specific case we chose it to be equal to k_{12}. See Figure 3. We used for both kinetic constants a value of 0.02/it, representing a residence time of molecules in the atmosphere as well as in the sink of $\tau_1 = \tau_2 = 50$ it. We see in the figure that the adjustment time is now no longer equal to the residence times, but remarkably shorter. It is easy to show (Stallinga 2023) that the adjustment time τ in a two-box first-order-kinetics model is given by the 'reciprocal sum',

$$\tau = \frac{1}{k_{12} + k_{21}}$$

in this case, $\tau = 25$ it. For such a simple two-box system the residence time in box 1 ('atmosphere') is given by the reciprocal of the kinetic constant associated to the flux going out of box 1 into box 2 ('ocean'); using Equation [Eq:3] we find,

$$\tau_1 = \frac{1}{k_{12}}$$

and by comparing these two, we see that the adjustment time of the atmosphere-ocean system is indeed shorter than the residence time of molecules in the atmosphere; $\tau \leq \tau_1$. In the limit of no reflux from the ocean to the atmosphere ($k_{21} = 0$), as in the case of a nuclear-decay-like system, the two are equal. For comparison, in the figure exponential curves with a time constant of 50 it are shown. The curves of both atmosphere (red) and sink (blue) settle twice as fast compared to the situation of exponential decay. By adding relaxation paths we speed up the relaxation, which is a general property of relaxation.

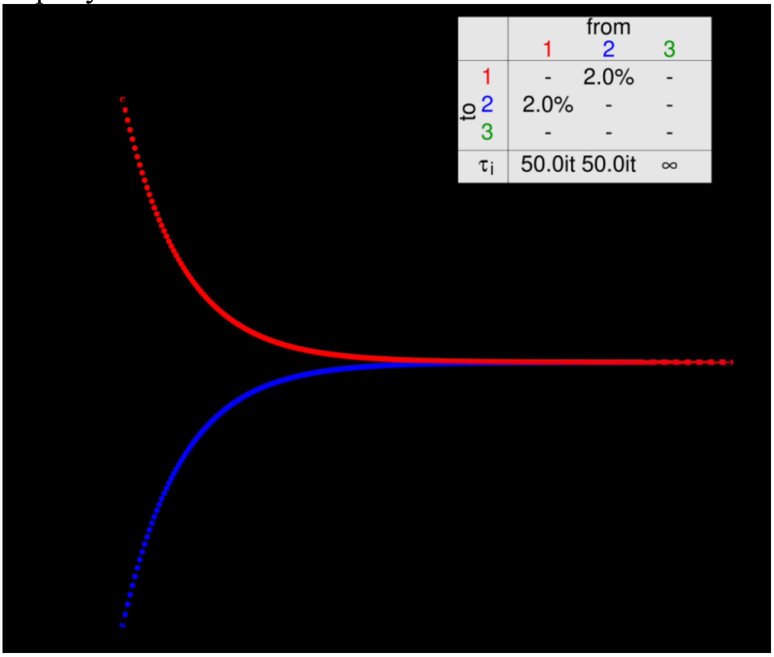


Figure 3: A two-box first-order (α = 1) simulation with reflux, i.e., molecules going back from box 2 to box 1. Every iteration 2% of molecules in the atmosphere (red box 1) are captured by the sink (k_{12} = 0.02/it), and 2% of molecules in the sink (blue box 2) are released into the atmosphere (k_{21} = 0.02/it). The kinetic constants are shown in the legend, which also shows the residence times τ_i.

Note also that it does not matter in which box the disturbance occurs, or how much will eventually wind up where. To illustrate that we make a simulation with asymmetric residence times. Box 1 keeps a residence time of τ_1 = 50 it, but for the reflux from box 2 into box 1 we use a four times smaller kinetic rate k_{21} = 0.005/it, box 2 has a four times longer residence time, τ_2 = 200 it. See Figure 4. We see that the adjustment time is equal in both boxes and still faster than the 50 it that was found in the exponential decay of Figure 2, and shorter than the residence time in box 1 as well as box 2; the adjustment time τ is 40 it (Eq. [Eq:7]) compared to the residence times of 50 it and 200 it. We also see that the final fraction in box 1 is given by $k_{21}/(k_{12} + k_{21})$, in this case 0.5%/(2%+0.5%) = 0.2.

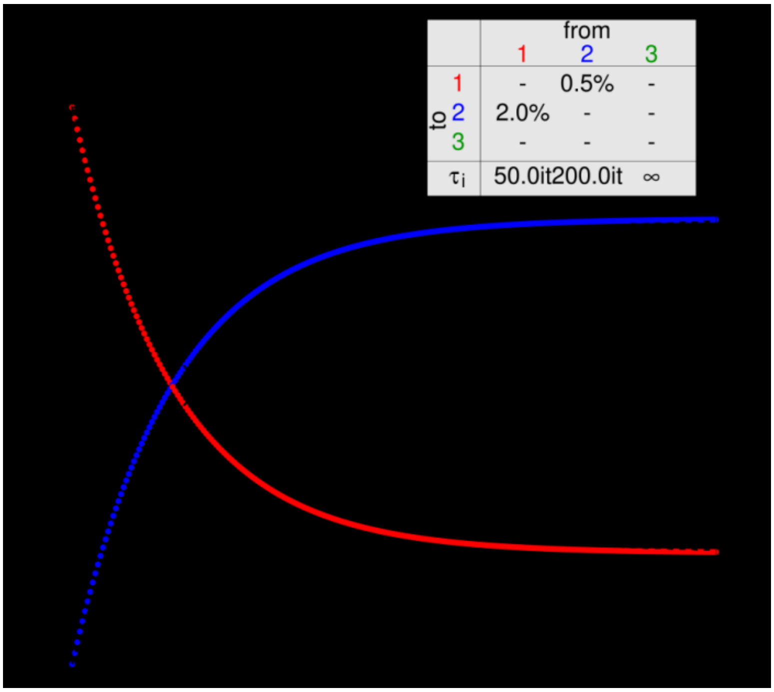


Figure 4: A two-box first-order (α = 1) simulation with reflux. Every iteration 2% of molecules in the atmosphere (red box 1) are captured by the sink (k_{12} = 0.02/it), and 0.5% of molecules in the sink (blue box 2) are released into the atmosphere (k_{21} = 0.005/it). The kinetic constants are shown in the legend, which also shows the residence times τ_i.

This system represents the atmospheric kinetics very well. As mentioned before, because the kinetics are first-order, with fluxes proportional to the contents of the boxes, we can take any subset of molecules and do the kinetics on them. For instance the analysis of CO_2 with carbon isotopes produced after above-ground nuclear bomb tests. These stopped abruptly and this is the engineer's dream data. The data show two things: 1) the transients are of the type exponential decay indicating first-order kinetics, 2) the final level of CO_2 remaining in the atmosphere (B_1) is tiny compared to what winds up in the oceans (B_2). It is difficult to estimate exactly how much, but from the equilibrium equation (Eq. [Eq:5]) we know that $k_{12} \gg k_{21}$, and the adjustment time τ of the two-box system is close to the residence time τ_1 of molecules in the atmosphere.

Adding boxes

In first-order kinetics, the fluxes are linear functions of quantities in the boxes. As we know from mathematics, a function f is linear if $f(ax) = af(x)$, and $f(a+b) = f(a) + f(b)$. This implies that any combination of solutions of the differential equation is also a solution. In this way we can *imagine* the carbon-dioxide in the atmosphere as composed of identifiable natural CO_2 and ’anthropogenic’ CO_2. As if they are ’colored’ yellow and red. Then separately solve the differential equation for these two subsets of CO_2, and then add the solutions to find the total behavior of the atmosphere. More specifically, we can imagine the atmosphere in steady state, and then analyze only the disturbance caused by human behavior. This is what was done in Ref. (Stallinga 2023) and it was found that already 90% of the carbon dioxide has disappeared from the atmosphere if we use a 2-box first-order model assuming a atmospheric residence time of 5 years.

If we add boxes, while the system is still first-order, we can keep on analyzing the system in this way, by only analyzing the disturbance and then adding it to the natural steady state. For two boxes it was done in the previous section. We will now add a third box and, to see the kinetics, we first go back to a system resembling nuclear decay, in the sense that there is flux from box 1 to box 2 and from box 2 to box 3, but never a flux in the other directions. A nuclear decay cascade. We see that adding a third box does not change the kinetics of the atmosphere. See Figure 5. The adjustment time of the atmosphere (red box 1) remains at 50 it and the transient of box 1 purely exponential. The same cannot be said for the other two boxes. To find the adjustment times for these two boxes we must place the disturbance at those boxes. If we, for instance, place the disturbance in box 2, the system resembles simple nuclear of Fig. 2 from blue box 2 to green box 3. The reader can imagine the same plot with the colors exchanged. Note also that a nuclear decay cascade system with all the disturbance in the final box 3 would yield three straight lines: the green box-3 line at fraction = 1 and the other two (box-1 red and box-2 blue) at fraction = 0. To save space these plots are omitted here.

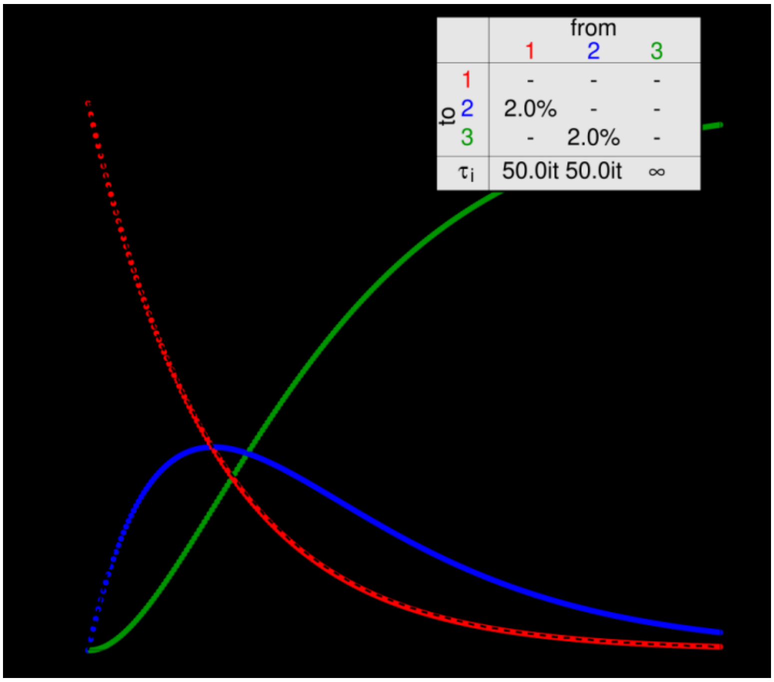


Figure 5: A three-box first-order (α = 1) simulation without reflux, resembling a nuclear decay cascade. Every iteration 2% of molecules in the atmosphere (red box 1) are captured by the shallow ocean (k_{12} = 0.02/it), and 2% of molecules in the shallow ocean (blue box 2) are removed to the deep-ocean sink (k_{23} = 0.02/it). The kinetic constants are shown in the legend, which also shows the residence times τ_i.

More interesting is the case of multiple boxes with reflux. Full exchange between neighbors (box 1 is connected to box 2, box 2 is connected to boxes 1 and 3, box 3 is connected to box 2). For simplicity all constants are chosen symmetric at 0.02/it between boxes 1 and 2 and five times slower at 0.004/it between boxes 2 and 3. In equilibrium boxes 1 and 2 must thus have equal amount, and boxes 2 and 3 must also have equal amount, therefore all boxes must be equal in equilibrium. This is shown in Figure 6. Note that the residence time in the middle box is now given by a sum. In general, any box B_i that has outflux to more than 1 box, has a residence time give by Eq. [Eq:3],

$$\frac{1}{\tau_i} = \sum_j k_{ij} = \sum_j 1/\tau_{ij}$$

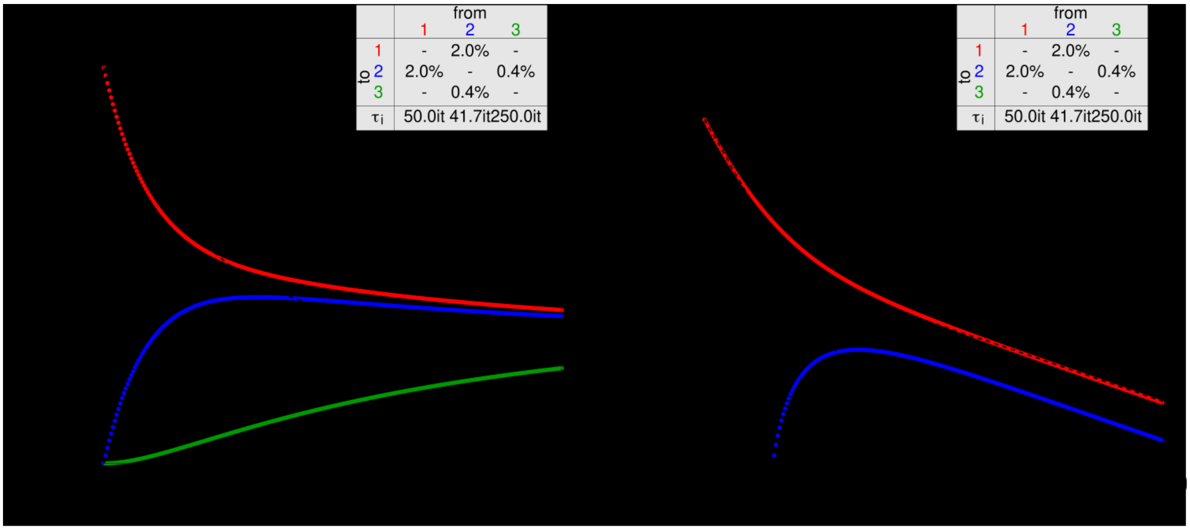


Figure 6: Left: A three-box first-order ($\alpha = 1$) simulation with exchange between neighbors. Every iteration 2% of molecules in the atmosphere (red box 1) are captured by the shallow ocean (k_{12} = 0.02/it), and 0.25% of molecules in the shallow ocean (blue box 2) are removed to the deep-ocean sink (k_{23} = 0.004/it). Similar constants are for the way back: k_{32} = 0.004/it and k_{21} = 0.02/it. The kinetic constants are shown in the legend, which also shows the residence times τ_i. Right: Logarithmic scale of the same curves after subtracting offsets (final values F_{final}) of 1/3. Fitting exponent with two relaxation constants τ: 35 it and 180 it.

We immediately see that the transient is no longer exponential and no adjustment time τ can be defined. Initially the transient is faster, but then becomes slows down over time; compare the red curve of the atmosphere to the exponential curve with a relaxation time of 50 it, shown as a dashed line. It at first decays at an adjustment time equal to one as if the deep ocean did not exist and a (pseudo)equilibrium is established between atmosphere (red box one) and shallow ocean (blue box 2); the time scale which is shorter than the residence time in the atmosphere and shallow ocean. On a longer time scale they together establish equilibrium with the deep ocean (green box 3). It is a classic double-exponential transient. Plotting the logarithm of the curves relative to the final value, $\log(B_1(t) - B_{\text{final}})$ vs. time will show two slopes, an initial steep one and later a moderate one. See the right panel of the figure.

It is also interesting to see what happens if the disturbance is in the middle box 2. This is shown in Figure 7. It has the exact same parameters as the system above, but the disturbance is added to the middle box 2. This might then represent a system where blue box 2 is the atmosphere, red box 1 is the biosphere, and green box 3 is the oceans.

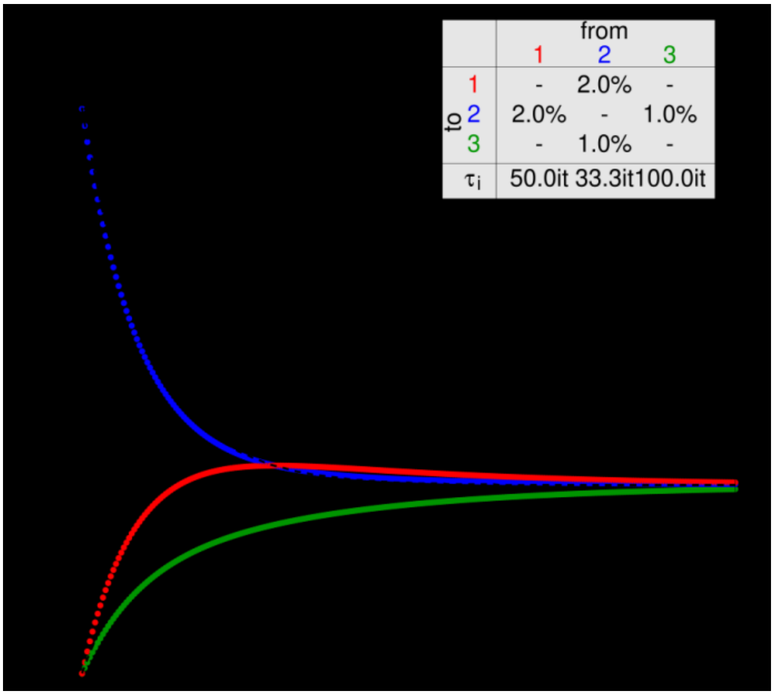


Figure 7: A three-box first-order (α = 1) simulation with exchange between neighbors with an initial disturbance at the middle (blue) box 2. Every iteration 2% of molecules in the atmosphere (red box 1) are captured by the shallow ocean (k_{12} = 0.02/it), and 2% of molecules in the shallow ocean (blue box 2) are removed to the deep-ocean sink (k_{23} = 0.02/it). Similar constants are for the way back: k_{32} = 0.02/it and k_{21} = 0.02/it. The kinetic constants are shown in the legend, which also shows the residence times τ_i. This might represent a system where blue box 2 is the atmosphere, red box 1 is the biosphere, and green box 3 is the oceans.

Unequal boxes

Now let's go back to our two-box first-order-kinetics system and relax the condition that boxes are of equal volume. What happens if one box is bigger than the other? For simplicity's sake we will take a system with box 2 half the volume compared to box 1. There is now no longer a direct equivalence between contents (unit: mol) and concentrations (unit: mol/L).

Concentrations are only half in the double-sized box 1 compared to the same amount in box 2. We have a problem that a Chemistry analysis of the system is no longer valid because the sum of concentrations is no longer constant, seemingly violating the law of Lavoisier; when 1 mol/L leaves box 1 it enters box 2 as 0.5 mol/L and seemingly 0.5 mol/L was destroyed. We must speak of Physics quantities only (mol, and not mol/L). Yet, how to explain it to people more familiar with Chemistry textbooks?

We must assume that the driving mechanism behind the kinetics is diffusion, and according to Fick's first law such diffusion is proportional to the gradient of *concentration*. Applied to our system, flux between two boxes is the result of difference in concentrations and not amounts in the boxes, and also proportional to the area (unit: m^2) of interface between the boxes, which we keep constant. We get a system that could hopefully be conventionally described in Chemistry terms as an equation with concentrations (concentration of chemical species X in box i given by $[X]_i = B_i/V_i$) rather than amounts on both sides:

$$B_1 \underset{k_{21}}{\overset{k_{12}}{\rightleftharpoons}} B_2$$

(Note the non-italics $\mathrm{B_1}$ and $\mathrm{B_2}$ representing species 1 and 2). And then translate that into our Physics system

$$\frac{B_1}{V} \underset{k_{21}}{\overset{k_{12}}{\rightleftharpoons}} \frac{B_2}{V/2}$$

(Note the italics B_1 and B_2 representing species 1 and 2, representing quantities of $\mathrm{B_1}$ and $\mathrm{B_2}$ respectively). With volume V canceling, but don't forget that they are needed for a chemistry type of analysis that talks in terms of concentrations. (Chemical equations are normally used to describe concentrations in the same single closed box B; here is where our physics system deviates from Chemistry). Where the B_i still represent physical amounts (moles), this so that $\sum_i B_i$ = constant, while the kinetic constants conventionally pertain to concentrations. However, whether a kinetic constant is expressed in units of mol/s per mol, or mol/Ls per mol/L is mathematically irrelevant, since it winds up in a unit 1/s. It is only relevant for our mental picture. In this Physics analysis, fluxes F_{ij} between boxes are in terms of moles per second and not moles per liter per second, and we prefer to speak in terms of amounts and kinetic constants are rather mol/s per mol.

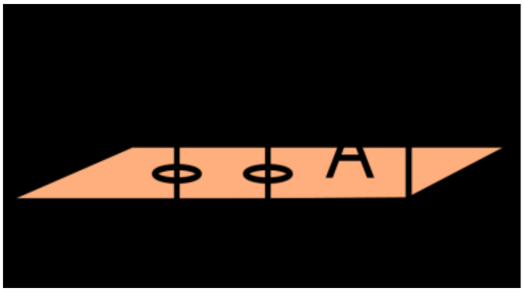


Figure 8: When boxes are not of equal volume, fluxes F (mol/s) are proportional to concentrations B/V and area A. Here an example is shown for two boxes.

Continuing. What it means in practice is that the above reaction equation reduces to the format in terms of amounts:

$$B_1 \underset{k_{21}}{\overset{k_{12}}{\rightleftharpoons}} 2B_2$$

which is aesthetically more pleasing since fluxes are then directly described in terms of amounts per second. However, we would then be laboring under the misapprehension of dealing here with a chemical reaction, which would result in a second-order kinetics (to be discussed in the next section). We must therefore insist on following a Physics reasoning. And the solution is quite simple. At the interface between the boxes molecules are exchanged with fluxes proportional to *concentrations* B/V, the kinetic constants k, and the interface area A, see Figure 8. In other words, for fluxes between boxes i and j Equation [Eq:1] is replaced by

$$F_{ij} = k'_{ij} A_{ij} \frac{B_i}{V_i}$$

with k'_{ij} having units m/s, and necessarily $A_{ij} = A_{ji}$. With thus a total effect of,

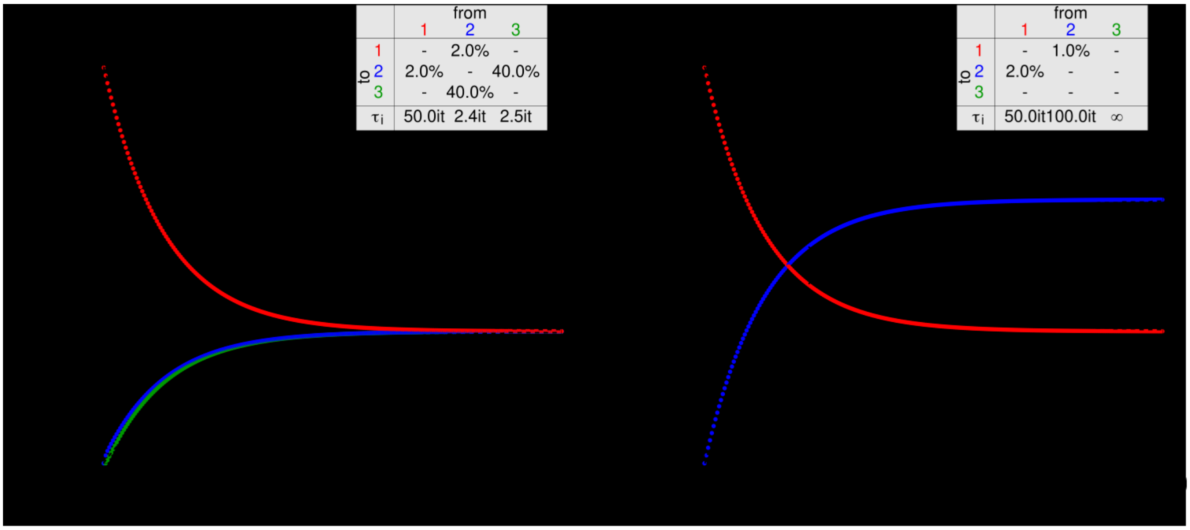
$$\frac{\mathrm{d}B_1(t)}{\mathrm{d}t} = -F_{12} + F_{21} = -k'_{12} B_1 A_{12}/V_1 + k'_{21} B_2 A_{21}/V_2$$

$$\frac{\mathrm{d}B_2(t)}{\mathrm{d}t} = F_{12} - F_{21} = k'_{12} B_1 A_{12}/V_1 - k'_{21} B_2 A_{21}/V_2$$

We see that this is a normal system as described above for two equal boxes, but with the kinetic constants scaled proportional to volume, $k_{ij} = k'_{ij} A_{ij}/V_i$. Boxes having different volumes does not change anything about the behavior of the system, it only changes the kinetic constants. We could actually simulate this with *equal* boxes with normal kinetics, but with the kinetics constants adequately scaled with dimensions of the boxes.

To show that this works, we repeat the two-box first-order kinetics system, now with box 2 twice the volume. We do this by simulating it by three boxes of equal volume. Between box 1 and 2 we have normal kinetics parameters, but there are ultra-fast kinetics between box 2 and 3, $k_{23} = k_{32} = \infty$ to represent a well-mixed situation in the meta-box 2/3. This is shown in Figure 9, which indeed mimics Figure 4 if we join boxes 2 (blue) and 3 (green) together. Noteworthy, the transients are still exponential and the adjustment time τ is still shorter than the residence times.

We can therefore safely continue with our analysis as if our boxes are of equal volume, keeping in mind that the effective kinetic constants scale with the respective volumes.



		from	
	1	2	3
to 1	-	2.0%	-
to 2	2.0%	-	40.0%
to 3	-	40.0%	-
τ_i	50.0it	2.4it	2.5it

		from	
	1	2	3
to 1	-	1.0%	-
to 2	2.0%	-	-
to 3	-	-	-
τ_i	50.0it	100.0it	∞

Figure 9: Left: A (pseudo)three-box first-order (α = 1). A simulation with symmetric exchange ($k_{21} = k_{12}/2 = 0.02$/it) between box 1 (red) and box 2 (blue), and with very rapid exchange between box 2 (blue) and box 3 (green); effectively the two are one box. Exchange between box 2 and 3 is here chosen to be 20 times faster than between box 1 and box 2. The blue and green curves nearly coincide. If we sum them together into one box we get the system of two boxes, with $k_{21} = k_{12}/2 = 0.01$/it, shown on the right. These two systems are identical in behavior, which demonstrates that kinetic parameters effectively scale with volume.

Non-linear kinetics

In this section we analyze non-first-order kinetics. In that case, the linearity condition of the functions is no longer met, and we cannot analyze subsets of molecules separately. The possible combinations of systems is then infinite. We limit ourselves to some specific cases. First of all, what is presented here is only systems where at $t = 0$ molecules are injected in a box of an hitherto empty system.

The first system to analyze is a two-box decay system without reflux. F_{12} is given by Equation (2) and F_{21} is equal to zero. For first order ($\alpha = 1$) the solution was given in an earlier section and is exponential decay like a nuclear decay system (Figure 2). First order is a special case. The general solution for order α, of such a two-box system with differential equation

$$\frac{\mathrm{d}B_1(t)}{\mathrm{d}t} = -F_{12} = -k_{12}B_1(t)^{\alpha}$$

after setting the starting value of box 1 to $B_{1,0}$ at $t = 0$, is

$$B_1(t) = [\text{unit}] \times \left[\left(\frac{B_{1,0}}{[\text{unit}]}\right)^{1-\alpha} + (\alpha - 1)\frac{t}{\tau_{12}}\right]^{1/(1-\alpha)}$$

with $\tau = 1/k_{12}$. Note we had to factor the unit/scale out of the equation (for instance [unit] = [1 mol]) otherwise it would not make sense,

B_1 has units mol and raising units to a fractional power $(1-\alpha)$ does not make physical sense.

A special case is the second-order-kinetics ($\alpha = 2$) two-box decay model. It results in a so-called delayed-power-law function,

$$B_1(t) = B_{1,0} \times \frac{\tau_{12}}{\tau_{12} + (B_{1,0}/[\text{unit}])t}$$

An example is shown in Figure 10. It is called delayed-power law because after initially being constant it turns into a power law (with power equal to -1) after a characteristic time τ_{12}. This is easiest seen in a double-logarithmic scale (right panel of Figure 10). Notice the red curve becoming linear after 50 it ($\log_{10}(\text{it}) = 1.7$), also the point where half of the original amount in box 1 has disappeared from it and entered box 2; at $t = \tau$: $B_2 = B_1$.

These transients do not have an adjustment time constant; the speed of adjustment slows down over time. For comparison, the figure also shows a normal exponential transient (dashed line). At the start they coincide but start ever-more diverging when the value of B_1 drops. This behavior is also called long-tail, something made famous by the book of Nassim Taleb, *The Black Swan* (Taleb 2011). In chemistry such transients are observed when two reactants on one side of a reaction result in one product on the other side, for instance this reaction without reflux:

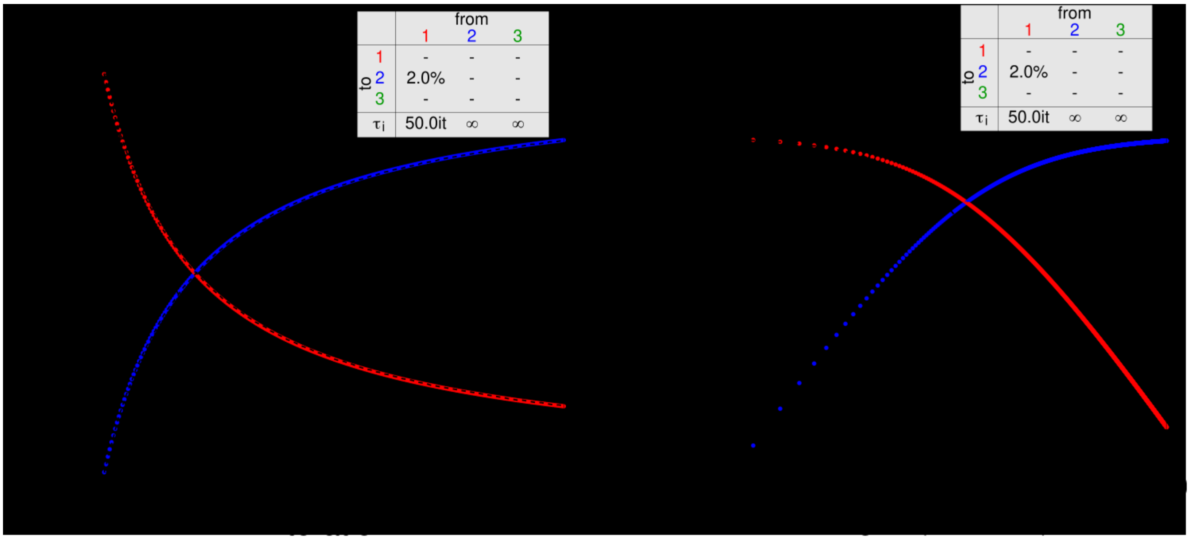
$$R + R \overset{k}{\rightarrow} P$$

The differential equation is then

$$\frac{\text{d[R]}}{\text{d}t} = -k[\text{R}]^2$$

resulting in kinetic curves as described above. Similar kinetics we find for two-photon absorption processes, whose magnitude also depends quadratically on the light intensity. Although mathematically it can make sense, it seems highly unlikely that this type of behavior is applicable to the atmosphere. Molecules of CO_2 that do not react with anything are only by diffusion changing from one state (box) to another. These processes are rather have linear kinetics, *viz.* Fick's law.

Careful atmospheric studies of carbon-isotope concentrations after above-ground atomic-bombs experiments (Solomon 2008) have revealed that the kinetics are first order, moreover with only a very tiny reflux, making the system nearly similar to one of nuclear decay described in the beginning. First-order kinetics seems to be the most reasonable candidate for the atmosphere, since the underlying process seems to be diffusion, which is essentially first order. Non-first-order kinetics systems are only described here for completeness' sake.



to \ from	1	2	3
1	-	-	-
2	2.0%	-	-
3	-	-	-
τ_i	50.0it	∞	∞

to \ from	1	2	3
1	-	-	-
2	2.0%	-	-
3	-	-	-
τ_i	50.0it	∞	∞

Figure 10: Left: A two-box simulation (third box is not connected) with second-order kinetics ($\alpha = 2$). The decay is a delayed-power function of Equation [Eq:8]. Box 3 (green), although existing, is not connected to the other boxes by any fluxes $\forall i{:}\, k_{i3} = k_{3i} = 0$, and thus remains in its start value, $B_3(t) = 0$. Right: same plots on double-logarithmic scale.

Another often-mentioned scheme is zeroth-order kinetics, implying a flux that does not depend on concentrations. In this case the solution is quite simple. As an example may serve the two-box system with no reflux; F_{12} is constant and $F_{21} = 0$,

$$\frac{\mathrm{d}B_1(t)}{\mathrm{d}t} = -F_{12}$$

$$\frac{\mathrm{d}B_2(t)}{\mathrm{d}t} = +F_{12}$$

This is also called the bathtub model, molecules like CO_2 are removed from the atmosphere at a constant rate. The model was created by Sterman and coworkers (Sterman and Sweeney 2007; Carter 2015), who wanted to ”create the interactive simulation with children and youth in mind” (Interactive 2025). How zeroth-order kinetics would work in nature is difficult to understand, but remember that also our anthropogenic injections into the atmosphere are of this type, independent of concentrations.

In this case we can talk about a *removal* time $\tau_{1,\mathrm{r}}$, the time it will take to remove the rest of (all) molecules from box B_1, given at any time by

$$\tau_{1,\mathrm{r}}(t) = \frac{B_1(t)}{F_{12}}$$

The residence time could then be defined as the *average* time a molecule stays in the atmosphere and is then half the removal time,

$$\tau_1 = \frac{\tau_\mathrm{r}}{2} = \frac{B_1}{2F_{12}}$$

While this is the easiest system to model, it remains to be explained how this can make sense in physics terms. Moreover, our emissions might be needed to keep the planet alive by offsetting the constant leaking of CO_2 out the atmosphere and depleting it to below the life-sustaining level of 180 ppm. In any case, the transients are shown in Figure 11.

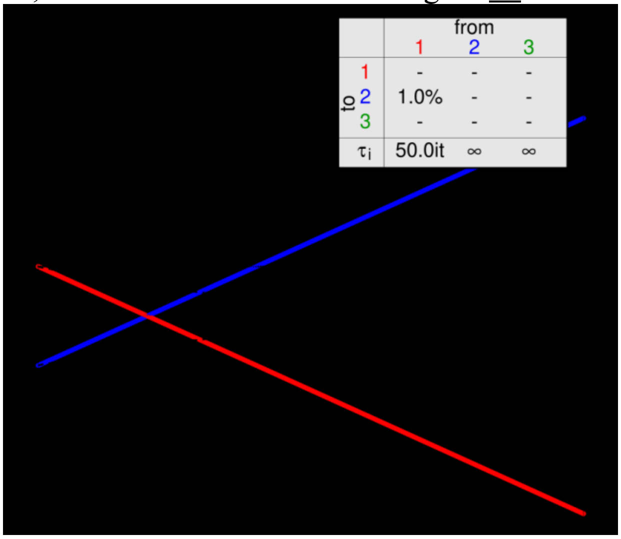


Figure 11: Left: A two-box simulation with zeroth-order kinetics (α = 0), the so-called bathtub model: a constant removal of CO_2 from the atmosphere into the sinks. The residence time (50 it) is half the removal time (100 it) of all the molecules. Note that mathematically the removal continues after everything has been removed.

Finally, important for these non-first-order systems is to remember that we can no longer make a partial analysis as was done in the beginning by ’coloring’ the molecules and solving the system for each subsystem of a specific color molecules and then adding it all together. Only in the last case of zeroth-order kinetics can we do an attempt. The removal time for a subsystem b_1 in box B_1 (imagine these molecules to be colored yellow, contrasting the natural molecules colored red, all well mixed) is equal to the removal time of the entire amount B_1. Therefore, also the residence time of those ’b’ molecules is equal to that of all molecules,

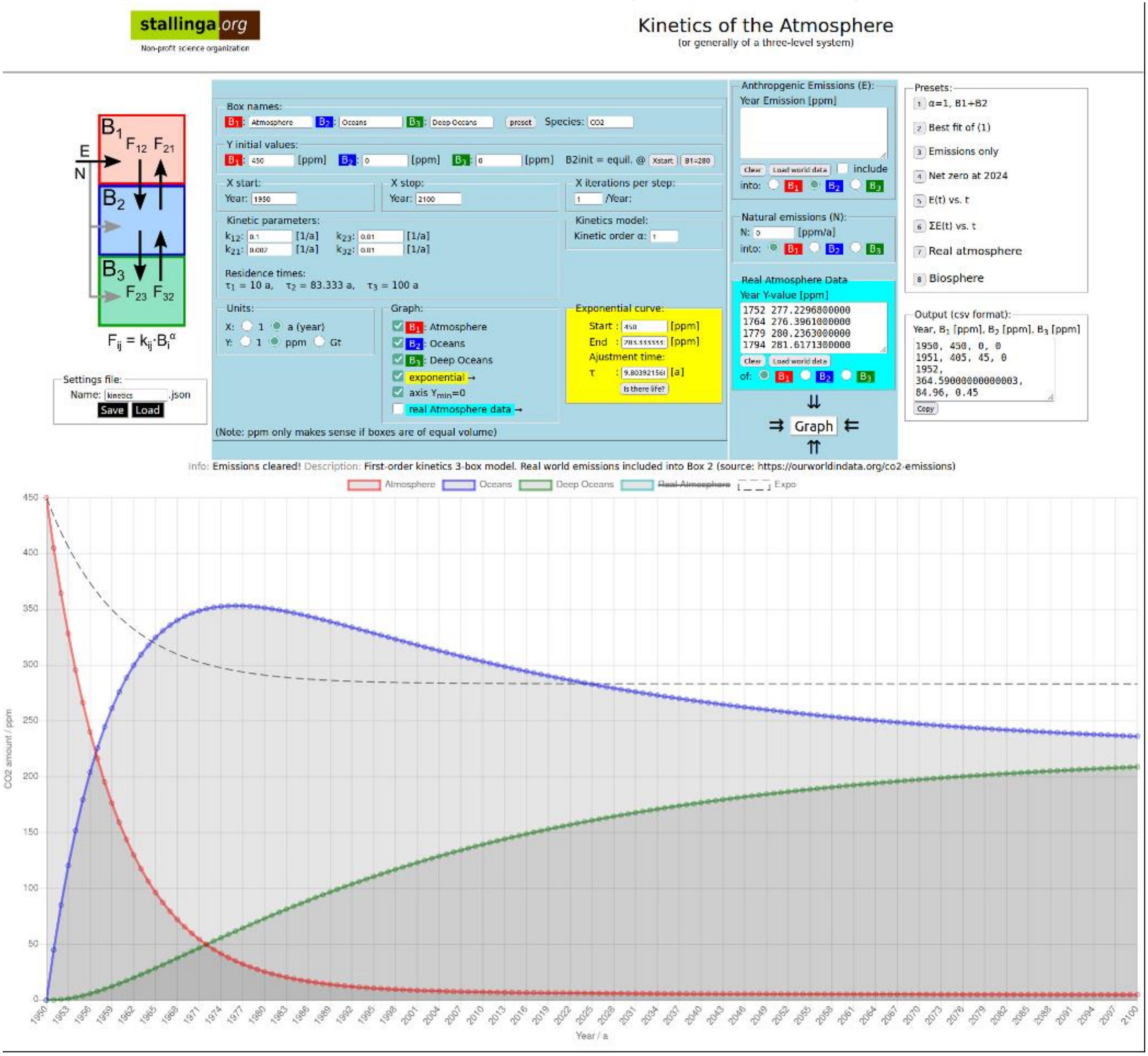
$$\tau_{1,\mathrm{b}} = \tau_{1,\mathrm{B}} = \frac{B_1}{2F_{12}}$$

If the flux F_{12} were a trickle, this indeed would take centuries. Maybe more interesting is to know what the ’re-adjustment time’ is, the time it takes to remove a certain amount b_1 of molecules from the box, for instance an amount of molecules equal to the amount b_1 added by humans. This is given by,

$$\tau_\mathrm{b} = \frac{b_1}{F_{12}}$$

Online kinetics lab tool

An application was written in open-source Javascript and can be found at stallinga.org (Peter Stallinga for stallinga.org 2025). Figure 12 shows a screenshot of the current version. It has all the ingredients discussed in the current work and the simulations here presented in the next section were based on output from this app. (The theoretical simulations of the previous section were done in Lazarus using Free Pascal, though).



Kinetics of the Atmosphere
(or generally of a three-level system)

Settings file:
Name: kinetics .json

Box names:
B_1: Atmosphere B_2: Oceans B_3: Deep Oceans Species: CO2

Y initial values:
B_1: 450 [ppm] B_2: 0 [ppm] B_3: 0 [ppm] B2init = equil. @

X start:
Year: 1950

X stop:
Year: 2100

X iterations per step:
1 /Year:

Kinetic parameters:
k_{12}: 0.1 [1/a] k_{23}: 0.01 [1/a]
k_{21}: 0.002 [1/a] k_{32}: 0.01 [1/a]

Kinetics model:
Kinetic order α: 1

Residence times:
τ_1 = 10 a, τ_2 = 83.333 a, τ_3 = 100 a

Units:
X: ○ 1 ● a (year)
Y: ○ 1 ● ppm ○ Gt

Graph:
☑ B_1: Atmosphere
☑ B_2: Oceans
☑ B_3: Deep Oceans
☑ exponential →
☑ axis Y_{min}=0
☐ real Atmosphere data →

Exponential curve:
Start : 450 [ppm]
End : 283.333333 [ppm]
Ajustment time:
τ : 9.80392156 [a]

(Note: ppm only makes sense if boxes are of equal volume)

Anthropgenic Emissions (E):
Year Emission [ppm]
☐ include
into: ○ B_1 ● B_2 ○ B_3

Natural emissions (N):
N: 0 [ppm/a]
into: ● B_1 ○ B_2 ○ B_3

Real Atmosphere Data
Year Y-value [ppm]
1752 277.2296800000
1764 276.3961000000
1779 280.2363000000
1794 281.6171300000
of: ● B_1 ○ B_2 ○ B_3

Presets:

Output (csv format):
Year, B_1 [ppm], B_2 [ppm], B_3 [ppm]
1950, 450, 0, 0
1951, 405, 45, 0
1952,
364.59000000000003,
84.96, 0.45

Info: Emissions cleared! Description: First-order kinetics 3-box model. Real world emissions included into Box 2 (source: https://ourworldindata.org/co2-emissions)

Figure 12: Online javascript application of a kinetics 3-box lab in which readers can test their own models. Available at stallinga.org (Peter Stallinga for stallinga.org 2025).

At each iteration the current state of the boxes is saved, then *all* changes $\Delta B_i = \sum_j \left(k_{ji}B_j - k_{ij}B_i\right) \times \Delta t$ are calculated on basis of the current state and *afterwards* the changes are applied to the boxes. This procedure avoids a loss of molecules that might occur if we immediately apply changes to the current state the moment we calculate a change (flux).

Because these are numerical simulations, and not analytical solutions, an additional feature has to be added in order to make sure that they do not suffer from classic numerical simulations problems described by Runge and Kutta (Chapter 17.1 of Ref. (Press et al. 2011)). The derivative of our function y depends on time and the function value itself and it is not expected to be constant; a change in function value, Δy, in a step is only to a certain degree equal to the derivative of the function, $y'(t)$, and the time step, Δt:

$$\Delta y(t) = \int_t^{t+\Delta t} y'\,(t')\mathrm{d}t' \approx y'(t) \times \Delta t$$

Moreover, the formula is ”unsymmetrical” as it is evaluated at the beginning of the interval (Press et al. 2011). In our case the function steps are the ΔB_i, and the derivatives the fluxes formed by the F_{ij} which depend on the B_i, and the simulation system indeed suffers from this problem. The change of contents in box B_i in a time step is given by

$$\Delta B_i(t) \approx \sum_j \left(F_{ji}(t) - F_{ij}(t)\right)\Delta t = \sum_j \left[k_{ji}B_j(t) - k_{ij}B_i(t)\right]\Delta t$$

The solution in the simulation app is to subdivide each data step into many smaller iteration steps, implementing the Euler approach, instead of following the full Runge-Kutta method or Simpson’s approximation. Instead of one step with full k parameters, n iterations with parameters k scaled down by a factor n are done. The user should increase the number of iterations per step until the point where simulations do not change significantly anymore. The iteration-step problems become apparent when the kinetics parameters start approaching unity. For parameters smaller than about 0.5/step these considerations are not relevant. For simplicity’s sake, all simulations done in the next section were done with 1 iteration per step.

Applying it to the real atmosphere

Let us now apply the above kinetics models to the real atmosphere by adding emission E to it and see if we can simulate the real CO_2 data. In what follows, box B_1 represents the atmosphere and B_2 and B_3 the sinks, for instance the shallow and deep oceans. For the concentrations and emissions we use as source OurWorldInData, Refs. (Our World In Data 2025a) and (Our World In Data 2025b), respectively. The former based on data from the NOAA Global Monitoring Laboratory.

It has to be noted that, regardless of the order of kinetics, the overall system is ’linear’. Meaning that if we double the total amount of molecules, all boxes will be doubled in equilibrium. That is the result of Equation [Eq:5], the ratio of any two boxes is a function of the kinetics constants that connect them. This except for zeroth-order ($\alpha = 0$) which mathematically and physically does not make sense and has to be treated separately.

Figure 13 shows real data of solubility of CO_2 in water (Haynes et al. 2017). As we can see, even for extreme temperatures (100 °C) and pressures (100 kPa partial pressure of CO_2) the water/CO_2 behaves linear. The ratio between CO_2 in gas form above water and the concentration dissolved in the water is constant. And this is called Henry’s Law, which states that ”the amount of gas dissolved in a liquid is directly proportional to the partial pressure of that gas above the liquid at equilibrium”. This gives us confidence that our system is adequate for the atmosphere/oceans. While we do not delve into the temperature dependence here, it is important to point out that Henry’s Law proportionality coefficient is depending on temperature and that real measured dependence of real salty ocean water by Al Anezi and coworkers (Al-Anezi et al. 2008) revealed a dependence that can perfectly explain the correlation of data of [CO_2] and temperature on a time scale of 800 ka (Stallinga 2018). Moreover, Indermühle and coworkers made a full statistical analysis (Indermühle et al. 2000) and find that [CO_2] was lagging behind the temperature by some 900 years and note that ”This value is roughly in agreement with findings by Fischer et al. who reported a time lag of CO_2 to the Vostok temperature of (600 ± 400) year during early deglacial changes in the last 3 transitions glacial-interglacial” (Fischer et al. 1999). Fischer and coworkers attribute the delay to the ocean outgassing effects, i.e., Henry’s Law, and even find that at colder times the delay is longer, which is itself consistent with Arrhenius-like behavior of thermally-activated processes, such as most in nature (Stallinga 2018).

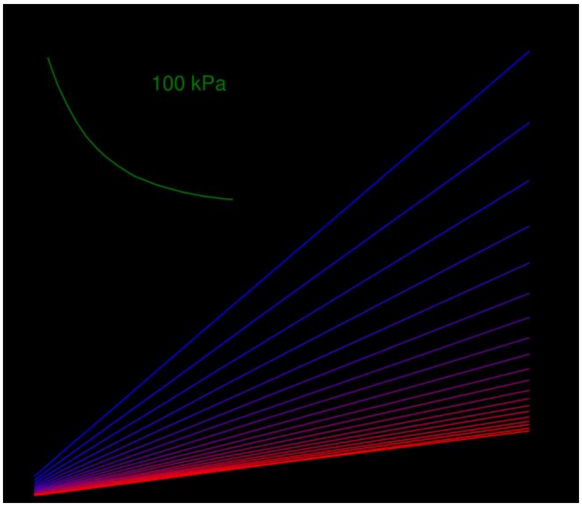


Figure 13: Mole fraction of CO_2 in water as a function of partial gas pressure above it. Curves for temperatures from 0 °C (blue) to 100 °C (red). The linearity shows this is following Equation [Eq:5]; in our system we can imagine horizontal B_1 and vertical B_2, with the ratio of the two constant, with the kinetic constants, and thus ratio, depending on temperature. This ratio is called Henry's Law. The inset shows the dependence on temperature for 100 kPa partial pressure. Data from Ref. (Haynes et al. 2017).

Zeroth order kinetics

Let us first consider the easiest to analyze case of zeroth-order – constant flux – bathtub model described above. It can be rejected on terms of its non-physical nature, as observed above. Zeroth order processes are difficult to imagine in nature when not in some way very ordered, something diffusion processes certainly are not.

Moreover, where the bathtub model assumes that only a tiny trickle of outflow F_{12} exists, by far outweighed by the anthropogenic emissions E into the atmosphere, contemporary data have shown that the yearly removal of CO_2 from the atmosphere is equal to about 50% of these emissions (Stallinga and Khmelinskii 2018), see also Figure 14; it is literally gushing out of the atmosphere. The net outflow can be estimated by,

$$F_{12}(t) - F_{21}(t) = E(t) - \frac{\mathrm{d}B_1}{\mathrm{d}t}$$

and is shown in Figure 14 in green. What is more, this removal fraction is in lockstep with the emissions trend, it is a constant 50% for all the years reported. It has grown from 5 Gt/a to about 20 Gt/a in the sixty years from 1960 to 2020. This proves the bathtub model wrong, since the fluxes are obviously *not* constant.

Moreover, if all these natural fluxes are zeroth-order, independent of conditions, and continue to keep rising at the current rate, we must increase our emissions too, or the planet is doomed. To give an example, the simulation in Figure 15 shows that life would cease to exist – with a CO_2 concentration below 180 ppm – before the year 2040 had we adopted net-zero policies already back in 1955. This model seems very unlikely, but should we take a gamble with our planet? Isn't it better to be safe than sorry? After all, we do not have a Planet B.

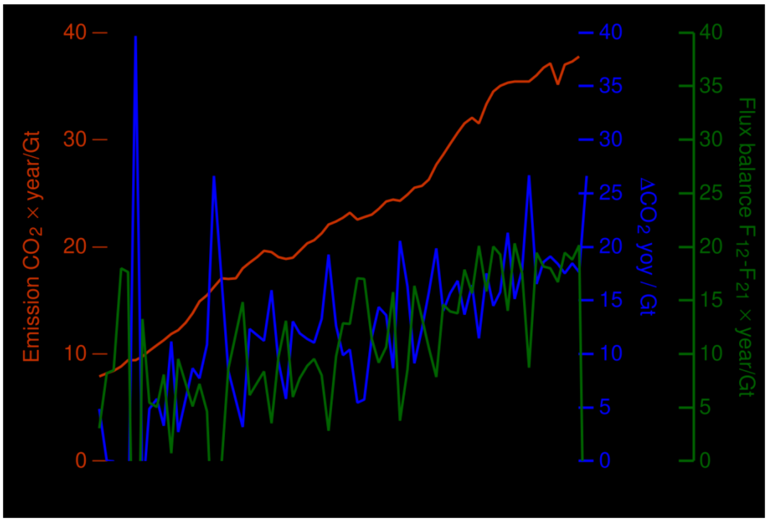


Figure 14: Red: Emission of carbon dioxide in the atmosphere by anthropogenic sources. Source: Ref. (Our World In Data 2025b). Blue: Year-on-year (yoy) growth of CO_2 in the atmosphere. Source: Ref. (Our World In Data 2025a). Green: the difference between the two, which is the net outflow from box B_1. (1 ppm of $[CO_2]$ equals 7.76 Gt of CO_2). Note that the data have become of much noisier character since the publication of Ref. (Stallinga and Khmelinskii 2018). The reason for this data manipulation not clear to the author.

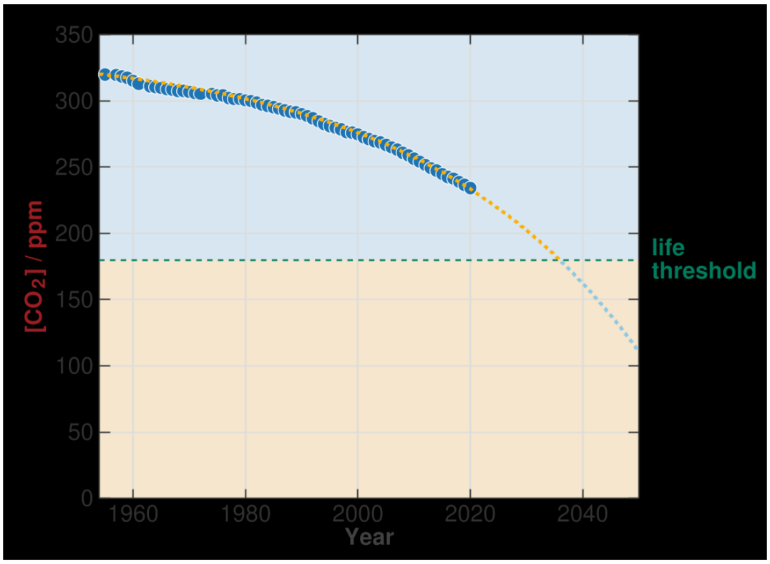


Figure 15: Based on the real data of Figure 14 and a zeroth-order model: If we had stopped emissions in 1955, considering the measured zeroth-order fluxes $F_{12} - F_{21}$, we can predict that life would cease to exist before the year 2040.

First order kinetics

For the moment we would rather reject the zeroth-order model and continue with a physically more sensible model of first-order kinetics that is consistent with relevant processes such as diffusion. The simplest first-order kinetics model is one with only two boxes. In such a system, when at this moment half of the amount of injected CO_2 immediately disappears from the atmosphere (box B_1), we can understand this as the sink having a large capacity with a large residence time, and a residence time of molecules in the atmosphere of about 2 years. To be more precise, if the residence time in the oceans (box B_2) is much larger, then the residence time τ_1 in the atmosphere (box B_1) is equal to the adjustment time of the system (Eq. [Eq:6]). It is then given by

$$\tau_1(t) = \frac{B_1(t) - B_{1,\text{eq}}}{F_{12} - F_{21}}$$

This can serve as a first estimation. As an example, for the year 2020, where B_1 = 412.44 ppm (3201 Gt), and from Figure 14 we find $F_{12} - F_{21}$ = 20 Gt/year, while for the equilibrium value we use the often-heard value of $B_{1,\text{eq}}$ = 280 ppm (2173 Gt). This results in an atmospheric residence time of τ_1 = 50 years (k_{12} = 0.02/a). This is much longer than the value commonly accepted in the literature (and the one used by us in a previous work (Stallinga 2023)) but it can replicate contemporary data to a rather good degree.

Figure 16 shows a first-order-kinetics two-box model simulation with parameters as sown in Table 2. The values of k_{21} and $B_2(1950)$ are difficult to estimate, one can only state that $B_2 \gg B_1$ and $k_{21} \ll k_{12}$, but were chosen such that an atmospheric concentration 280 ppm would be the equilibrium value. Most noteworthy is the kinetic parameter k_{12} = 0.0192, which indicates a residence time of 52 years.

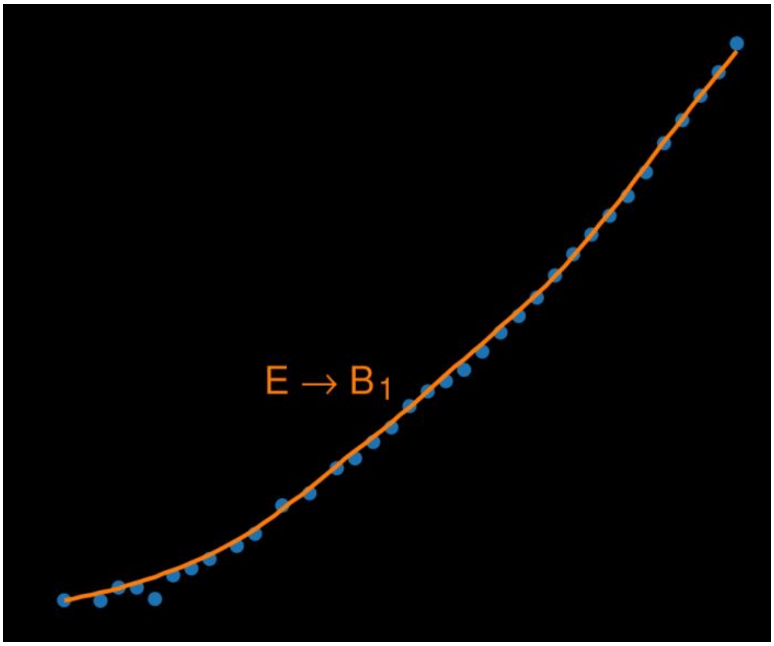


Figure 16: A first-order-kinetics two-box model simulation (solid orange line) with the real emissions (E) of Figure 14 into box B_1, and parameters such as described in Table 2. The blue full circles are real observed concentrations.

Table 2: Parameter values used in the simulation of Figure 16.

Parameter	Value
$B_1(1950)$	312.831 ppm
$B_2(1950)$	14,000 ppm*
k_{12}	0.0192/a
k_{21}	0.0004/a

*: assuming equal volumes

Considering the quality of the simulation, there is no need to add more boxes or to change the order of the kinetics. Ockham's Razor makes us chose the simplest mode with the least number of parameters possible. We could, for instance add a box for the biosphere. It would then seem most obvious to add this above the box of the Atmosphere, which becomes B_2 and the Oceans become B_3. However, best simulations are achieved with the kinetics parameters connecting the atmosphere and the biosphere so fast, compare to those connecting the atmosphere and the oceans, as effectively make them into one box, as described above. In other words, adding a box does not improve the simulation quantity; we can consider the atmosphere/biosphere box as a single well-mixed homogeneous box.

However, the residence found for molecules in the atmosphere is more than an order larger than the 4 years mentioned by the IPCC in its report (Stocker et al. 2013), and also substantially larger compared to the value found by Solomon (Solomon 2008). In a previous work we assumed the value of 5 years to calculate how much of the anthropogenic CO_2 would still be in the atmosphere and reached a conclusion that only some 10% of these still remain in the atmosphere (Stallinga 2023). On basis of the new found value here we can simulate that nearly 60% of anthropogenic CO_2 is still present in the atmosphere. See Figure 17.

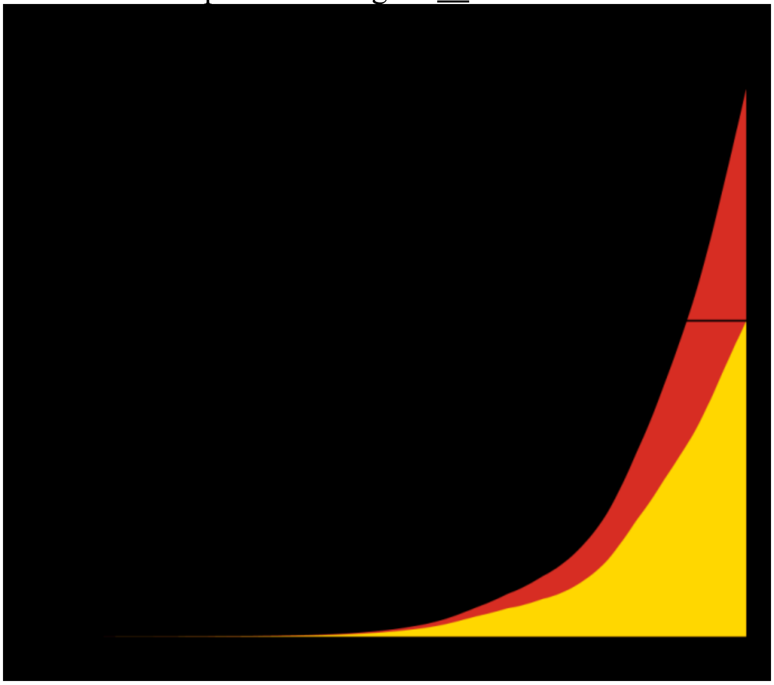


Figure 17: Cumulative emissions (red) and the fraction that is still remaining in the atmosphere (yellow). Based on a residence time in the atmosphere of τ_1 = 52 a.

There is a huge discrepancy between the residence time of 52 years found in this work and the reliable work of Solomon of about an order of magnitude. Especially the isotopes-based methods after the abrupt ending of the atomic bomb tests are very reliable, since they supply ideal stimulus functions for signal analysis. As any signal engineer knows, analysis is easiest done with Heaviside functions or delta-Dirac functions because they can easily be used in Laplace transforms. The isotopes methods are based on Heaviside functions and do deliver a clear-cut analysis. This in contrast to the convoluted method here, where the stimulus signal – the anthropogenic emissions – is not clean. The model seems to be inadequate, in spite of its rather good fitting results.

That makes us look for other features that might have to be added to the system. Natural seems to have kinetic parameters k_{ij} that depend on temperature. This is so-called Henry's Law, where the ratio of concentrations in the atmosphere and in a body of water depends on temperature, represented in temperature-dependent kinetics parameters. This indeed seems a good candidate for effects on the CO_2 concentrations, but also much more complex to simulate, and outside the scope of the current work. We are here trying to find out how good simple multi-box systems can model the carbon-dioxide in the atmosphere.

We can, however, add another feature, namely natural emissions N. This can be in any box, but most reasonable seems the box representing the oceans. This would then also simulate, to some extent, Henry's Law. However, it does not seem possible to make a good fit using this system when we fix the residence time in the atmosphere to the reliable 10 years (k_{12} = 0.1/a). See Figure 18 where injections were tried into B_1 (2 ppm/a) and B_2 (40 ppm/a).

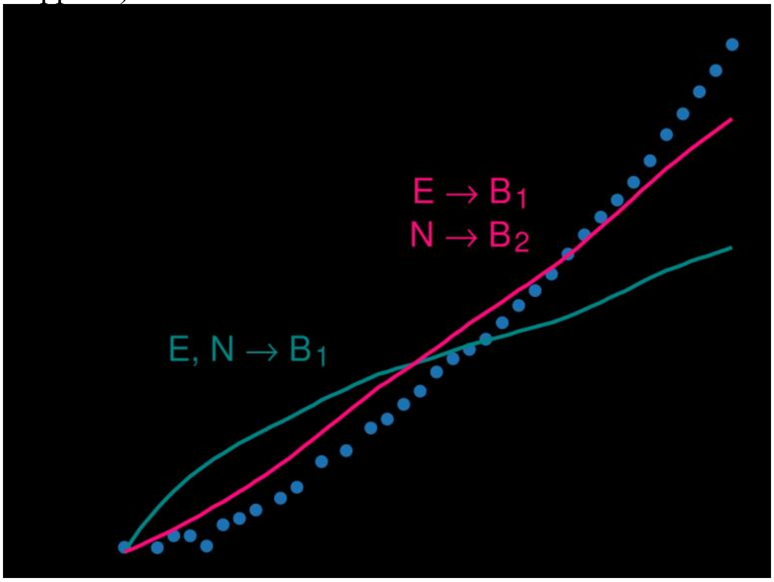


Figure 18: First-order-kinetics two-box model simulations with the real emissions (E) of Figure 14 into box B_1 and natural emissions N into box B_1 (2 ppm/a, cyan solid line) or B_2 (40 ppm/a, solid magenta line) and parameters such as described in Table 2, with fixed τ_1 = 10 a (k_{12} = 0.1/a) and k_{21} 20 times smaller. Equilibrium at 1950. The full circles are real observed concentrations.

Conclusions

In this work multi-box systems for the kinetics of CO_2 in the atmosphere system are described. A good simulation was obtained using a first-order kinetics two-box model. Considering the quality of the simulation, there is no need to add more boxes or to change the order of the kinetics.

Ockam's Razor makes us chose the simplest model with the least number of parameters possible. In this case it is obvious that a first-order two-box model is adequate to simulate the data. The best fit was found with a residence of molecules in the atmosphere equal to 52 years, which is a value about an order of magnitude to high compared to the reliable value based on isotopic measurements, or the one presented by the IPCC and used in our earlier work. Using 52 years, we found that some 60% of anthropogenic CO_2 is still present in the atmosphere.

Using a more reasonable value of 10 years for the residence time, it was not possible to obtain a good fit, not even when (constant) natural emissions were included in the model.

In any case, this work was to present the modeling platform with 3 boxes and general kinetics, and not to have the final word in the values of kinetics. If the reader wants to do private simulations, the author has written an online kinetics ’laboratory’, which is available at stallinga.org.

Conflict of Interest: The author reported no conflict of interest.

Data Availability: All data are included in the content of the paper.

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