

On the Atmospheric Residence Time of Anthropogenically Sourced Carbon Dioxide

Gavin C. Cawley*

School of Computing Sciences, University of East Anglia, Norwich, U.K.

E-mail: gcc@cmp.uea.ac.uk

Abstract

A recent paper by Essenhigh¹ (hereafter ES09) concludes that the relatively short residence time of CO₂ in the atmosphere (5 – 15 years) establishes that the long term (\approx 100 year) rise in atmospheric concentration is not due to anthropogenic emissions, but is instead caused by an environmental response to rising atmospheric temperature, which is attributed in ES09 to “other natural factors”. Clearly, if true, the economic and political significance of that conclusion would be self-evident, and indeed most welcome. Unfortunately however, the conclusion is false; it is straightforward to show, with considerable certainty, that the natural environment has acted as a net carbon sink throughout the industrial era, taking in significantly more carbon than it has emitted, and so the observed rise in atmospheric CO₂ cannot be a natural phenomenon. The carbon cycle includes exchange fluxes that constantly redistribute vast quantities of CO₂ each year between the atmospheric, oceanic and terrestrial reservoirs. As a result, the residence time, which depends on the total volume of these fluxes is short. However the rate at which atmospheric concentrations rise or fall depends on the net difference between fluxes into and out of the atmosphere, rather than their total volume, and so the long term rise is essentially independent of the residence time. The aim of this paper is to provide an accessible explanation of why the short residence time of CO₂ in the atmosphere is completely

*To whom correspondence should be addressed

consistent with the generally accepted anthropogenic origin of the observed post-industrial rise in atmospheric concentration. Furthermore, we demonstrate that the one-box model of the carbon cycle used in ES09 directly gives rise to: (i) a short residence time of ≈ 4 years, (ii) a long adjustment time of ≈ 74 years, (iii) a constant airborne fraction, of $\approx 58\%$, in response to exponential growth in anthropogenic emissions and (iv) a very low value for the expected proportion of anthropogenic CO_2 in the atmosphere. This is achieved without environmental uptake ever falling below environmental emissions, and hence is consistent with the generally accepted anthropogenic origin of the post industrial increase in atmospheric carbon dioxide.

Introduction

The residence time (RT) of carbon dioxide in the atmosphere is defined as the average length of time a molecule of CO_2 remains in the atmosphere before being taken up by the oceans or terrestrial biosphere. The argument presented in ES09 is essentially that anthropogenic emissions cannot be the cause of the observed rise in atmospheric CO_2 as the residence time is short, of the order of only 4-15 years, and hence rather than accumulate in the atmosphere, anthropogenic emissions are rapidly taken up by the oceans and terrestrial biota. The error in this argument lies not in the premise, it is widely accepted (and indeed clearly stated in the reports published by the IPCC^{2,3}) that the residence time (RT) is only about 5 years; but the conclusion drawn does not follow from the premise. The aim of this paper is to explain why a short residence time is completely consistent with the generally accepted anthropogenic origin of the observed rise in atmospheric CO_2 . The error arises due to a confusion of residence time with the *adjustment time*, which describes the time taken for the atmospheric CO_2 concentration to substantially recover towards its original concentration following a perturbation;⁴ unlike other atmospheric gasses, the residence time and adjustment time are not the same for carbon dioxide. In the remainder of this section, we provide an overview of the global carbon cycle, and then provide intuitive arguments that demonstrate that the observed rise in atmospheric CO_2 cannot be a natural phenomenon and that the rate at which CO_2 accumulates in the atmosphere is essentially independent of the residence time. These arguments

demonstrate that while the residence time calculations of ES09 are correct, the conclusion of the paper, that anthropogenic carbon emissions are not responsible for the observed increase, must be false. In subsequent sections, we perform a more detailed analysis using a one-box model of the carbon cycle, similar to that used in ES09, to show that the rise in atmospheric CO₂ is completely consistent with an anthropogenic origin.

Overview of the Global Carbon Cycle

The global carbon cycle describes the large-scale transport of carbon between atmospheric, terrestrial, oceanic and lithospheric reservoirs. A schematic representation of the global carbon cycle, taken from chapter 7⁵ of the Working Group 1 contribution to the IPCC Fourth Assessment Report³ (hereafter AR4), is shown in Figure 1. The carbon cycle is dominated by vast exchanges of carbon dioxide between the atmosphere and surface ocean and between the atmosphere and terrestrial biota. As shown in Figure 1 the surface ocean emits some 90.6 $GtC\ yr^{-1}$ into the atmosphere, but also takes in approximately 92.2 $GtC\ yr^{-1}$. The net flux of -1.6 $GtCyr^{-1}$ is negative, indicating that the surface ocean acts a net carbon sink, taking in more carbon each year than it emits.⁶ Likewise, the terrestrial biosphere is also a net sink, emitting some 121.2 $GtC\ yr^{-1}$ each year through respiration, but taking up approximately 122.6 $GtC\ yr^{-1}$ each year in gross primary production (GPP). The exchange fluxes are very substantial, exchanging approximately 20% of the total atmospheric reservoir each year; hence the residence time, which can be calculated as the ratio of the mass of the atmospheric reservoir and the volume of the flux out of the atmosphere, is short $762/(92.2 + 122.6) \approx 3.5$ years. However, it is important to note that the residence time is short because of an *exchange* of carbon between the atmospheric, oceanic and terrestrial reservoirs, and a simple exchange of carbon does not affect the mass of carbon in any of the reservoirs concerned. In principle, it would be possible to have arbitrarily large exchange fluxes, and hence an arbitrarily short residence time, while at the same time the mass of the atmospheric reservoir remained constant. Like many common misunderstandings regarding the global carbon cycle, the residence time argument put forward in ES09 fails to consider the fact that natural fluxes into and

out of the atmosphere are closely balanced, and hence comparatively small anthropogenic fluxes can have a substantial effect on atmospheric concentrations. For a more detailed overview of the global carbon cycle, see the recent primer by Archer.⁷

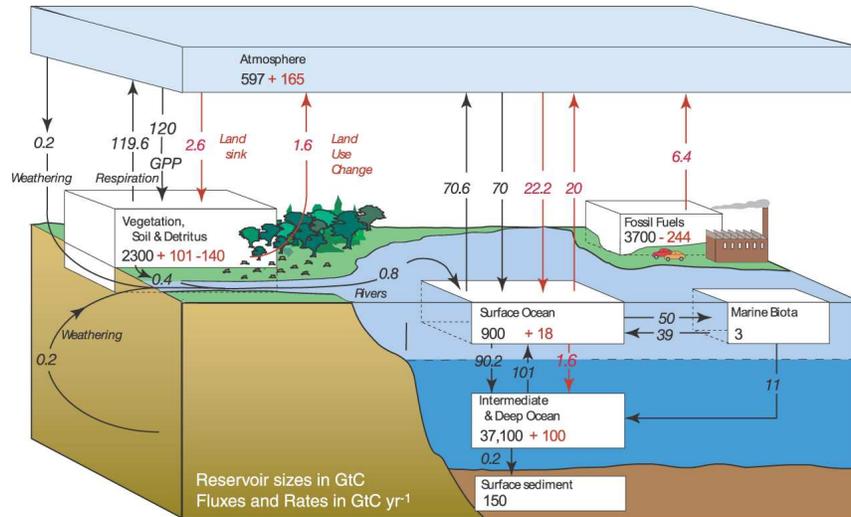


Figure 1: The global carbon cycle for the 1990s, showing the main annual fluxes in $GtC\ yr^{-1}$ and reservoir sizes in GtC :⁸ the pre-industrial ‘natural’ component in black and ‘anthropogenic’ component in red, after AR4, Figure 7.3.⁵

Availability of Data Relating to the Carbon Cycle

Data describing anthropogenic emissions and the growth of atmospheric CO_2 , used in the sequel, are publicly available from the Carbon Dioxide Information Analysis Centre (CDIAC).⁹ Atmospheric CO_2 concentrations are obtained from the Law Dome ice core proxy data-set,^{10, 11} covering the period 1832-1978 and the Mauna Loa observations^{12, 13} covering the period 1958-2008. Data describing anthropogenic emissions comprise of data describing fossil fuel emissions for the period 1751-2006^{14 15} and data describing emissions due to land use changes^{16, 17} covering the period from 1850-2005. All data were converted into GtC, using the conversion factor, gigatons of C = $2.35 \times ppm(v)$, taken from ES09. The availability of these data allow us to easily verify claims made regarding the carbon cycle.

The Recent Rise in Atmospheric CO₂ is Not a Natural Phenomenon

That the rise in atmospheric CO₂ concentrations observed since the turn of the industrial revolution is of anthropogenic, rather than environmental origin, can be inferred from observations of atmospheric CO₂ concentrations and estimates of anthropogenic emissions, using the principle of conservation of mass. These show that the environment has acted as a net sink throughout the industrial era, and hence cannot be responsible for the observed increase. Assuming that the carbon cycle is a closed system, the global atmospheric carbon budget is approximated by

$$\frac{dC}{dt} = F_a + F_i - F_e, \quad (1)$$

(e.g. Raupach *et al.*¹⁸), where $C = v_a[\text{CO}_2]$ is the mass of atmospheric CO₂ (with $[\text{CO}_2]$ being the atmospheric CO₂ mole fraction and following ES09¹ $v_a = 2.35 \text{ PgC ppm}^{-1}$); F_a represents anthropogenic emissions, F_i environmental (oceanic + terrestrial) emissions and F_e environmental uptake (it is assumed that anthropogenic uptake is insignificant); dC/dt is the growth rate of atmospheric CO₂. Rearranging (Eq. (1)) gives,

$$\frac{dC}{dt} - F_a = F_i - F_e, \quad (2)$$

which means that we can infer the net environmental flux $F_i - F_e$, given the observed/estimated data for atmospheric concentrations and anthropogenic emissions, without needing to know the absolute magnitudes of F_i or F_e . It should be noted that a common objection to arguments relating to the anthropogenic effect on the carbon cycle is based on the fact that the best available estimates of the individual fluxes, shown in Figure 1, are highly uncertain, such that the error bars on F_i and F_e are typically larger than the volume of anthropogenic emissions. However, this objection does not apply to the mass balance arguments, as the net environmental flux, $F_i - F_e$, is not calculated from uncertain estimates of F_i and F_e , but from the difference between dC/dt and F_a , both of which are known with far greater certainty.

¹Raupach *et al.*¹⁸ give a lower figure of $v_a = 2.127$, however this lower value does not substantially alter the arguments presented in this paper.

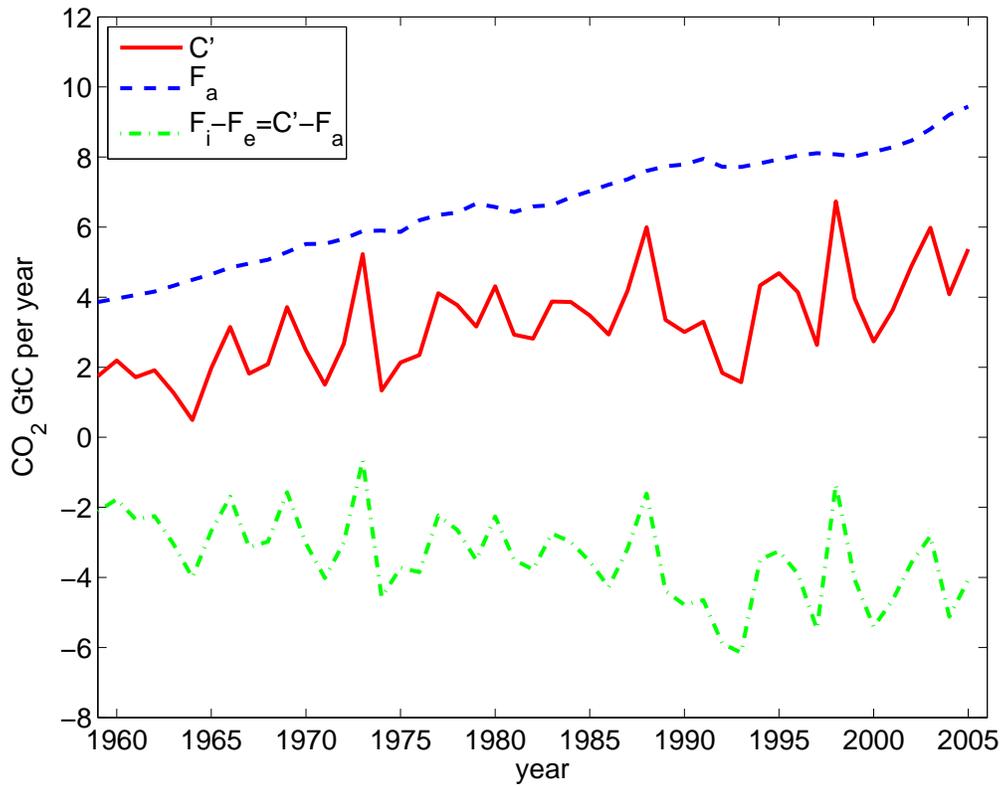


Figure 2: Annual anthropogenic emissions, F_a , annual growth rate C' and inferred environmental flux $F_i - F_e$, in GtC per year (c.f. Raupach *et al.*,¹⁸ Figure 1.d).

Figure 2 shows the results of a mass balance analysis. It can be seen that the net environmental flux, $F_i - F_e$ is always negative throughout the period covered by the Mauna Loa record, and has become more negative as time progressed. This shows beyond reasonable doubt that, at least for the last fifty years, the natural environment has acted as a net sink, rather than a source of atmospheric CO_2 . If the environment were a net source, then the observed rise would exceed anthropogenic emissions (as both the natural environment and anthropogenic emissions would be contributing to the rise), however this is observed not to be the case. As it has been established that the natural environment is a net carbon sink, it cannot be the cause of the observed increase of the last fifty years.

It should be noted that the mass balance argument is not the only form of analysis demonstrating that the rise in atmospheric carbon dioxide is due to anthropogenic emissions. Plants exhibit a preference for the lighter ^{12}C isotope of carbon over the heavier ^{13}C and ^{14}C isotopes. As a result, anthropogenic emissions from fossil fuel use and deforestation have a lower $^{12}\text{C}/^{13}\text{C}$ ratio than the atmosphere. Increasing anthropogenic emissions should therefore result in a decrease in the $^{12}\text{C}/^{13}\text{C}$ ratio of the atmosphere, and this is indeed what is observed.¹⁹ Similarly fossil fuels have been excluded from the carbon cycle for many millions of years, far longer than the radioactive half-life of the heavier ^{14}C isotope, and as a result are essentially devoid of ^{14}C . The Suess effect²⁰ describes the observed decrease in atmospheric ^{14}C (prior to nuclear bomb testing) due to fossil fuel emissions. There has been a small decline in atmospheric O_2 that would be expected as a result of fossil fuel combustion.¹⁹ Ice core measurements show that atmospheric CO_2 levels have varied between approximately 180 to 300 ppmv for the last 160,000 years²¹ and prior to the industrial revolution had not exceeded 300 ppmv during the current interglacial. Ocean acidification implies that the oceans are taking in more carbon dioxide than they emit and hence cannot be the cause of the observed increase in atmospheric CO_2 (for an excellent introduction to ocean acidification, see the series of articles by Mackie *et al.*²²). Lastly, the rise in atmospheric carbon dioxide closely parallels the rise in anthropogenic emissions, leading to an approximately constant airborne fraction,¹⁸ which would be somewhat of a coincidence if the rise were essentially

natural in origin! For good non-technical discussions of these issues, see the relevant articles at RealClimate,²³ SkepticalScience²⁴ and Ferdinand Engelbeen's web site:²⁵

(In)Dependence of the Growth of Atmospheric CO₂ on Residence Time

In this section, we demonstrate, by an analogy, that the growth or decline of the atmospheric CO₂ reservoir is essentially independent of the residence time. Consider a wash basin supplied by a constant stream of water flowing from a tap (faucet), but with an open drain at the bottom, so water is also constantly flowing out into a sewer (c.f. EPA²⁶). Clearly if the amount of water flowing in through the tap exactly matches the amount flowing out through the drain then the volume of water in the basin will remain constant. This is true regardless of actual flow; the amount of water flowing out of the basin can be made arbitrarily high, and hence the residence time arbitrarily short, without affecting the level of water in the basin. This is because the rate of change of the volume of a reservoir depends on the difference between total influx and total efflux, rather than on the magnitude of the fluxes. However, the residence time (i.e. the length of time a molecule of water remains in the basin before flowing out through the drain) is given by the ratio of the volume of water in the basin and the rate of flow through the drain. Hence residence time is effectively independent of the rate of growth or decline in the level of water in the basin.

More formally, consider the total environmental fluxes into and out of the atmosphere as being composed of the sum of a steady state equilibrium exchange flux, F_{ss} , plus a component representing the disturbance from that equilibrium, such that

$$F_i = F_{ss} + \Delta F_i \quad \text{and} \quad F_e = F_{ss} + \Delta F_e,$$

then the carbon budget can be written as

$$\frac{dC}{dt} = F_a + (F_{ss} + \Delta F_i) - (F_{ss} + \Delta F_e) = F_a + \Delta F_i - \Delta F_e,$$

while the residence time is

$$RT = \frac{C}{F_{ss} + \Delta F_e}.$$

The changes in the fluxes into the atmosphere are relatively small compared to their pre-industrial equilibrium values (the former are shown in red in Figure 1 and the latter in black, the estimate given there suggest that $F_{ss} = 190.2 \text{ GtC YR}^{-1}$, $\Delta F_i = 20 \text{ GtC yr}^{-1}$ and $\Delta F_e = 24.8 \text{ GtC yr}^{-1}$). This means that the residence time is dominated by the steady state exchange flux, F_{ss} , while the rate of change of the atmospheric reservoir is entirely independent of F_{ss} , which demonstrates that the residence time is of little relevance to a discussion of the cause of the observed rise in atmospheric CO_2 . The IPCC use the term “adjustment time” to characterise the time-scale involved in changes in the mass of the atmospheric reservoir, and draw a sharp distinction between the adjustment time and the residence time.

As an aside, it is often suggested that anthropogenic emissions cannot be the cause of the observed rise in atmospheric CO_2 because anthropogenic emissions are insignificant compared to environmental emissions. While it is certainly true that anthropogenic emissions are small compared to total environmental emissions ($F_a \approx 6.4 \text{ GtC yr}^{-1}$ versus $F_i \approx 112.2 \text{ GtC yr}^{-1}$), the argument is misleading because it is the difference in total emissions and total uptake that determines the rate of increase, and anthropogenic emissions are large compared with the net environmental flux ($F_a \approx 6.4 \text{ GtC yr}^{-1}$ versus $F_i - F_e = -2.8 \text{ GtC yr}^{-1}$). Note that the natural environment is a net carbon sink, and so were it not for ongoing anthropogenic emissions,²⁷ it would seem reasonable to expect atmospheric levels to be currently falling instead of rising.

Definitions of Adjustment Time and Residence Time

ES09 suggests a confusion in the 1990 IPCC WG1 scientific assessment report (FAR) on the definition of adjustment and residence times. However the definitions given in the FAR seem quite clear. For example, in Table 1.1 of the FAR (page 7), providing a “Summary of key greenhouse gasses influenced by human activities”, the row describing “Atmospheric lifetime” is given the

following footnote:

For each gas in the table, except CO₂, the “lifetime” is defined here as the ratio of the atmospheric content to the total rate of removal. This timescale also characterises the rate of adjustment of the atmospheric concentration if the emission rates are changed abruptly. CO₂ is a special case since it has no real sinks, but is merely circulated between various reservoirs (atmosphere, oceans, biota). The “lifetime” of CO₂ given in the table is a rough indication of the time it would take for the CO₂ concentration to adjust to changes in the emissions...

Note that “by no real sinks”, the footnote refers to fluxes into the lithosphere, which permanently remove carbon from the carbon cycle, at least on anything less than a geological timescale. This footnote appears immediately below the table in the same font size used in the main text. Later in section 1.2.1, the distinction between residence (turnover) time and adjustment time is made even more explicitly:

The turnover time of CO₂ in the atmosphere, measured as the ratio of the content to the fluxes through it, is about 4 years. This means that on average it takes only a few years before a CO₂ molecule in the atmosphere is taken up by plants or dissolved in the ocean. This short time scale must not be confused with the time it takes for the atmospheric CO₂ level to adjust to a new equilibrium if sources or sinks change. This adjustment time, corresponding to the lifetime in Table 1.1, is of the order of 50-200 years, determined mainly by the slow exchange of CO₂ between surface waters and the deep ocean. The adjustment time is important for the discussion on global warming potential.

This passage very clearly makes the distinction between “turnover” (residence) time and adjustment time. Note that the last line implicitly indicates that residence/turnover time is not important for the discussion of global warming potential. The distinction between turnover time and adjustment time is also made clearly and explicitly in the glossary of the most recent IPCC WG1 report

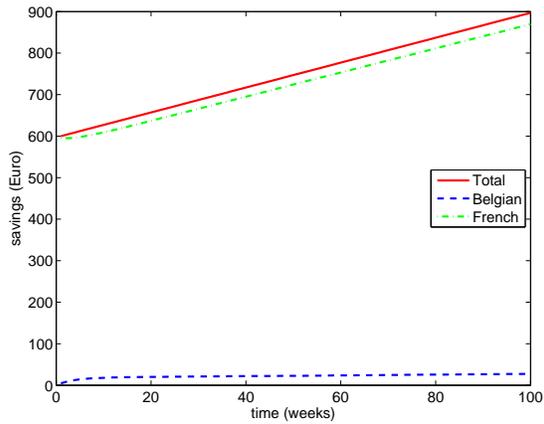
(AR4), in the entry for “Lifetime”.

Fraction of Carbon Dioxide of Anthropogenic Origin in the Atmosphere

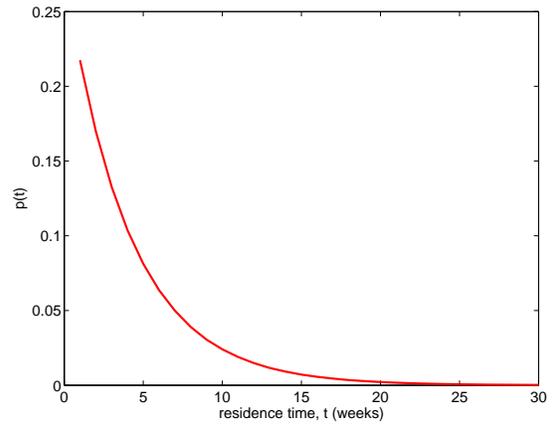
The short residence time of carbon dioxide in the atmosphere means that the mass of CO₂ of directly anthropogenic origin in the atmosphere is only a small fraction of the excess CO₂ that has built up since the industrial revolution. However, while certainly counter-intuitive, this does not support the conclusion that anthropogenic emissions cannot therefore be the cause of the increase in atmospheric CO₂ observed since the industrial revolution. Again, this common misunderstanding of the carbon cycle is a result of the large exchange fluxes, and is most easily explained by an analogy adapted from that of Engelbeen:²⁸

Consider a married couple, who keep their joint savings in a large jar. The husband, who works in Belgium, deposits six euros a week, always in the form of six one-euro coins minted in Belgium, but makes no withdrawals. His partner, who works in France, deposits 190 euros a week, always in the form of 190 one-euro coins, all minted in France. Unlike her husband, however, she also takes out 193 euro per week, drawn at random from the coins in the jar. At the outset of their marriage, the couple’s savings consisted of the 597 French-minted one euro coins comprising her savings. Clearly, if this situation continued for some time, the couple’s savings would steadily rise by 3 euros per week (the net difference between total deposits and withdrawals). It is equally obvious that the increase in their savings was due solely to the relatively small contributions made by the husband, as the wife consistently spent a little more each week than she saved.

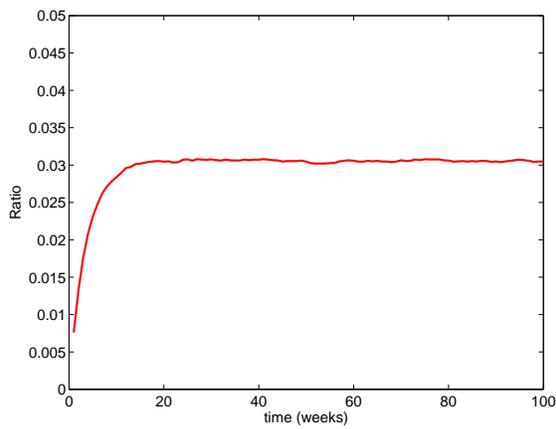
Figure 3 shows the result of a simple Monte-Carlo simulation of the couple’s savings jar. Figure 3 (a) shows the number of Belgian- and French-minted one Euro coins and the couple’s total savings as a function of time; as expected the total steadily rises at a rate of three Euros per week. However, the number of Belgian coins in the jar rises much more slowly, at an average rate of only ≈ 0.27 coins per week; perhaps even more surprisingly, the number of French-minted coins rises at an average rate of ≈ 2.73 coins per week, even though the net result of the wife’s transactions reduces the number of coins in the jar by 3 euros per week. The reason for this is that the volume



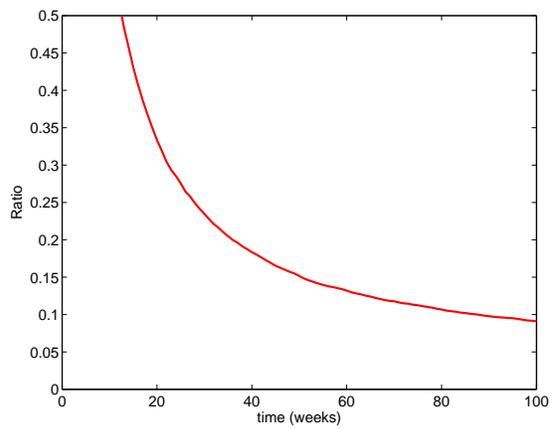
(a)



(b)



(c)



(d)

Figure 3: Results of Monte-Carlo simulation of a married couple's savings: (a) Number of Belgian- and French-minted Euro coins in the jar and total savings. (b) Distribution of residence times for coins withdrawn from the jar. (c) Proportion of Belgian-minted coins comprising the couple's total savings. (d) Proportion of Belgian-minted coins comprising the excess savings over and above the couple's initial savings of 597 Euro.

of the wife's withdrawals means that the mean residence time for coins in the jar is only ≈ 4.6 weeks, as illustrated by the distribution of observed residence times, shown in Figure 3 (b). This means that the coins in the jar, are constantly being exchanged with coins minted in France, due to the wife's transactions, and some of those coins will be the Belgian-minted coins deposited by the husband. As a result, the proportion of Belgian-minted coins in the jar converges to a value of only approximately 3%, as shown in Figure 3 (c). This leads to the rather counter-intuitive, but nevertheless completely correct conclusion, that just because the number of French minted coins in the jar has grown more rapidly than the number of Belgian coins, one cannot infer that the rise in their savings is not due to the husband's deposits. Such an inference ignores the effect of the large exchange flux created by the volume of the wife's deposits and withdrawals.

This analogy provides a very crude model of the global carbon cycle, where anthropogenic and natural fluxes are constant. The jar representing the atmosphere, a Belgian euro representing a Gt of carbon from anthropogenic emissions, a French Euro representing a Gt of carbon from "natural" sources, a week in the couple's finances a year in the global carbon cycle and their initial savings corresponds to the magnitude of the pre-industrial atmospheric reservoir (compare the figures used with those given in Figure 1). Hence for the real-world carbon cycle, it is reasonable to expect the proportion of CO₂ of directly anthropogenic origin to be very small, and for the bulk of the excess CO₂ above pre-industrial equilibrium concentration to be comprised of molecules emitted by the oceans and terrestrial biosphere, *even if the observed rise is of purely anthropogenic origin*. This surprising and counter-intuitive result stems from the effect of the large exchange fluxes in the natural carbon cycle.

Growth of Atmospheric Carbon Dioxide

Before moving on to a more realistic model of the carbon cycle, we would like to make some observations about the relationship between the increase in atmospheric CO₂ and anthropogenic emissions. Firstly, Figure 4 shows the anomalies for atmospheric CO₂ above a baseline of 669GtC for 1850, and the cumulative anthropogenic emissions from both fossil fuel use and land use changes

(deforestation). Clearly cumulative anthropogenic emissions have always been in excess of the increase in atmospheric CO₂, so we can be confident that there have been sufficient anthropogenic emissions to explain *all* of the observed rise in atmospheric concentrations.

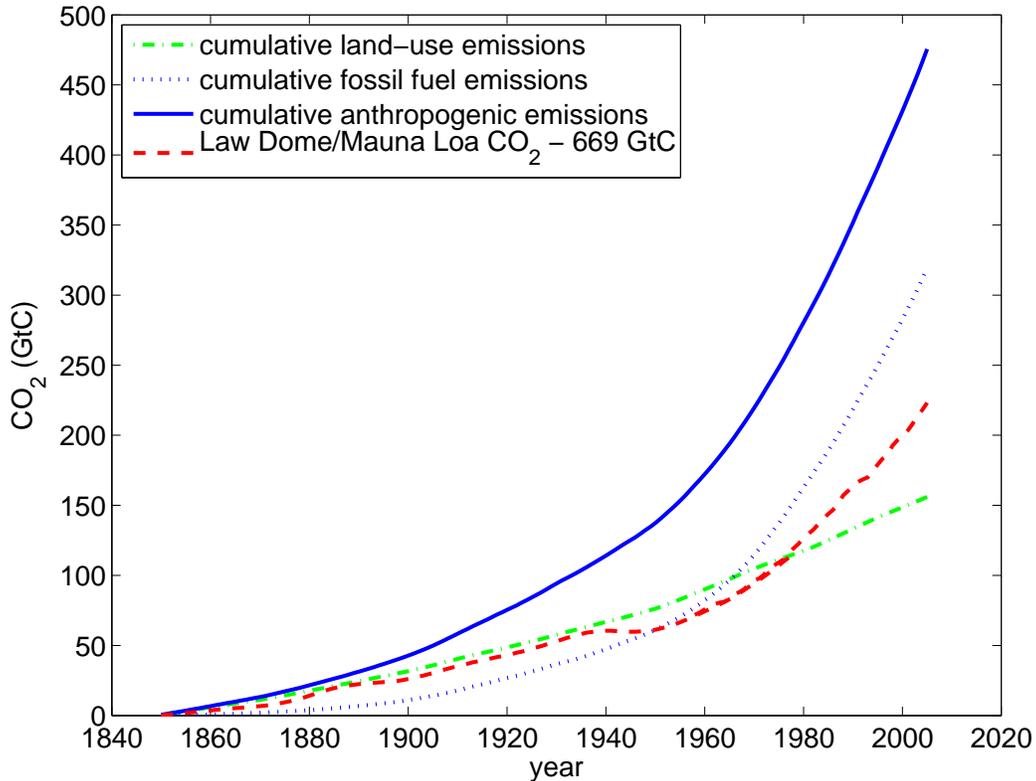


Figure 4: Atmospheric CO₂ anomalies and cumulative anthropogenic emissions (fossil fuel and land use), expressed in GtC, 1850-2005.

Figure 5 demonstrates that the rise in atmospheric CO₂ is closely approximated by a constant fraction of cumulative anthropogenic emissions. This fraction, known as the “airborne fraction”, is about 45%. If the observed rise in atmospheric concentrations were the result of environmental emissions, then the close correspondence between cumulative anthropogenic emissions and the observed increase would seem a rather inexplicable coincidence. While this is not in itself strong evidence of an anthropogenic origin of the observed rise, the fact that the simple one-box model, under some quite reasonable assumptions, gives rise to a constant airborne fraction is rather more convincing.

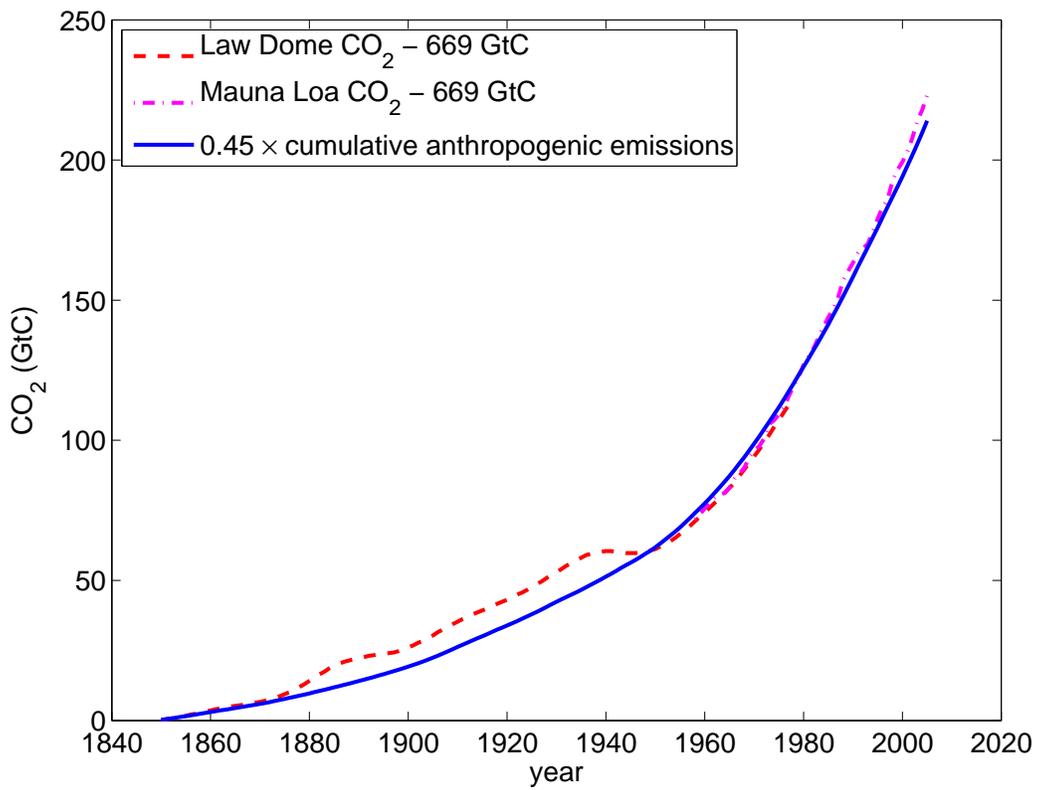


Figure 5: Atmospheric CO₂ anomalies and scaled cumulative anthropogenic emissions (fossil fuel and land use), expressed in GtC, 1850-2005.

A One-Box Model of the Carbon Cycle

In this section, we demonstrate that a single well-mixed box model, equivalent to the perfectly stirred reactor model of ES09, can be used to estimate both a short residence time, of about 4 years, and a longer adjustment time of approximately 74 years. Both of those figures are in line with the estimates given by the IPCC FAR. The model also demonstrates that a constant airborne fraction and a relatively small mass of atmospheric carbon dioxide of directly anthropogenic origin are also completely consistent with the commonly accepted purely anthropogenic cause of the observed atmospheric increase.

Modelling the Adjustment Time

We begin by setting up a simple one-box model of the carbon cycle, equivalent to the perfectly stirred reactor model of ES09, except that, for the sake of simplicity, we will deal directly with the mass of atmospheric carbon, C , rather than the concentration. The change in atmospheric carbon is given by the difference between the influx from environmental emissions, F_i , and the out-flux due to environmental uptake, F_e , i.e.

$$\frac{dC}{dt} = F_i - F_e$$

In the model described in ES09, environmental emissions are assumed to be approximately constant, and the return flux, F_i , is assumed to be a first order process, proportional to atmospheric carbon content, C , such that

$$F_i \approx \text{constant} = F_i^0$$

$$F_e = k_e C$$

Neither of these assumptions are satisfactory, however with a slight extension they can be used as the basis of a simple first order local linear approximation, for the period covered by the Mauna Loa record. Instead of assuming simple proportionality, the return flux is approximated by a linear

function of atmospheric CO₂,

$$F_e = k_e C + F_e^0. \quad (3)$$

Substituting, we get

$$\frac{dC}{dt} = F_i^0 - k_e C - F_e^0 \quad (4)$$

At equilibrium,

$$\left. \frac{dC}{dt} \right|_{C=C_{eq}} = 0 \quad \implies \quad F_i^0 = k_e C_{eq} + F_e^0,$$

from which we deduce that

$$k_e = \frac{1}{\tau} = \frac{F_i^0 - F_e^0}{C_{eq}}, \quad (5)$$

where τ is the *adjustment time*. Eq. (4) is a first-order inhomogeneous differential equation, that can easily be solved using the method of integrating factors, to give

$$C = C_{eq} + \gamma e^{-k_e t}, \quad (6)$$

where γ is a constant of integration, describing an initial perturbation from equilibrium conditions. The glossary of the AR4 defines the adjustment time as “... the time scale characterising the decay of an instantaneous pulse input into the reservoir.”, and so can be estimated using this simple one-box model. If we consider the case of a disturbance from equilibrium conditions by an instantaneous injection of an amount, C_A^0 , of anthropogenic carbon at time $t = 0$, then we obtain

$$C = C_{eq} + C_A^0 e^{-k_e t}.$$

This implies that total atmospheric carbon will decay exponentially from $C_{eq} + C_A^0$ at time $t = 0$ back down to C_{eq} , at a rate determined by the adjustment time, $\tau = k_e^{-1}$. Note that for the adjustment time, the rate of change of atmospheric carbon content is proportional to the difference between environmental emissions and environmental uptake, i.e. the net environmental flux, as suggested by our earlier informal arguments.

Figure 6 shows scatter plot of the difference between environmental emissions and environmental uptake (inferred via the mass balance argument from observations of atmospheric carbon dioxide at the Mauna Loa observatory and estimates of anthropogenic emissions from land use change and fossil fuel use) against the size of the atmospheric reservoir. A linear regression analysis of these data can be used to determine values for F_e^0 , k_e and thus τ . The regression implies that the best least-squares estimate of the relationship between environmental emissions and atmospheric CO_2 is given by

$$F_i^0 - F_e = \beta_1 C + \beta_0 \quad \implies \quad F_e = F_i^0 - \beta_1 C - \beta_0,$$

where $\beta_1 = -0.0135$ and $\beta_0 = 7.470$ are the regression coefficients. Comparison with Eq. (3) shows that

$$\begin{aligned} F_e^0 &= F_i^0 - \beta_0 = 190.2 - 7.470 = 182.7 \text{ GtCyr}^{-1}. \\ k_e &= -\beta_1 = 0.0135 \\ \tau &= \frac{1}{k_e} = 74.2 \text{ years}, \end{aligned}$$

where the total pre-industrial carbon dioxide emissions from natural sources is taken to be 190.2 GtCyr^{-1} (c.f. Figure 1). So this box model gives an estimate of 74.2 years for the adjustment time (95% confidence interval 72.1 – 76.4 years), which lies comfortably within the range of values given by the 1990 IPCC WG1 report (50-200 years).

Modelling the Residence Time

The next step is to obtain an estimate of the residence time from this same model. To do so, we note that atmospheric CO_2 is composed of an anthropogenic component and a “natural” component, such that

$$C = C_A + C_N,$$

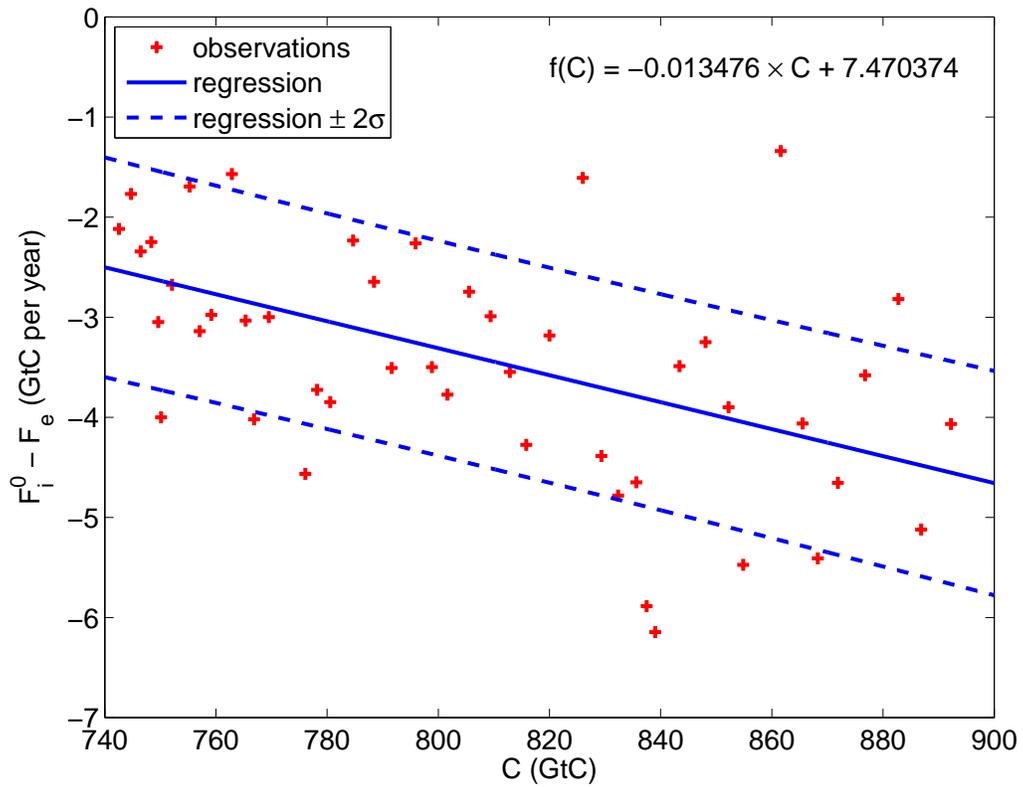


Figure 6: Regression analysis of the net environmental flux as a function of atmospheric carbon dioxide concentration.

where C_A is the atmospheric mass of carbon of anthropogenic origin and C_N is the mass of atmospheric carbon from natural sources. Clearly the environmental emissions are composed entirely of “natural” carbon; the uptake however is composed of both anthropogenic and natural carbon. Assuming the natural environment has no physical mechanism capable of distinguishing between carbon molecules according to their source, and that the CO_2 from both sources is well mixed, it seems reasonable to assert that the uptake is proportional to their mixing ratio, such that

$$\frac{dC_A}{dt} = -\frac{C_A F_e}{C} \quad (7)$$

and

$$\frac{dC_N}{dt} = F_i^0 - \frac{C_N F_e}{C} \quad (8)$$

Note that the rate of change of anthropogenically sourced carbon is proportional to environmental uptake. As this is much larger than the difference between environmental emissions and environmental uptake, we should expect the residence time to be much shorter than the adjustment time. Eq. (7) is a linear homogeneous differential equation, which can be solved using the method of separation of variables, however we can obtain an illuminating approximation by assuming that C and F_e are approximately constant, in which case (by inspection), the solution is given by

$$C_A \approx \xi e^{-k_r t}, \quad \text{where} \quad k_r = \frac{1}{\tau_r} = \frac{F_e}{C},$$

where τ_r is the *residence time*. Using the boundary condition that $C_A = C_A^0$ at time $t = 0$, we obtain

$$C_A \approx C_A^0 e^{-t/\tau_r},$$

such that the anthropogenic CO_2 decays from its initial value to zero at a rate governed by the residence time, which is approximately four years. Assume $C_A^0 = 165 \text{GtC}$ of anthropogenic CO_2

is injected at time $t = 0$, then at $t = 0$,

$$C = C_{eq} + C_A^0 = 579 + 165 = 744 \text{ GtC} \quad \text{and} \quad F_e = k_e C + F_e^0 = 0.0013476 * 774 + 182.7 \approx 192.7$$

and so

$$\tau_r = \frac{1}{k_r} = \frac{C}{F_e} \approx 3.86 \text{ years.}$$

Let us now return to the Eq. (7) and solve it exactly; it is a first order homogeneous linear differential equation, and hence can be solved by the method of separation of variables, giving,

$$C_A = C_A^0 \exp \left\{ k_e t - k_r t + \frac{k_r}{k_e} \log \left[\frac{C_{eq} + C_A^0}{C} \right] \right\}.$$

This gives an exponentially decreasing level of anthropogenic CO_2 , however the rate of this decrease declines as C and F_e decline as excess carbon is removed from the atmosphere.

Results of First Simulation

Figure 7 shows the results of the first simulation, where a pulse of 165 GtC anthropogenic carbon dioxide is introduced at time $t = 0$, and then allowed to decay back to the equilibrium state. Figure 7 (a) shows that the total carbon in the atmospheric reservoir declines slowly back to the equilibrium level over the course of a few hundred years, at a rate determined by the adjustment time of ≈ 74 years. However, the anthropogenic carbon content of the atmosphere declines very rapidly, and is essentially eliminated after only about a decade, due to the short atmospheric residence time of only about 4 years. This demonstrates that the single box model can simultaneously represent a long adjustment time, and a short residence time, with clearly distinct interpretations. The reason that the anthropogenic component decays at a rate being governed by the residence is that while it is *removed* from the atmosphere at a rate governed by net difference in total emissions and total uptake, it is also *replaced* by “natural” CO_2 at a rate governed by environmental uptake. It should be noted that the amount of carbon from natural sources increases as the amount of anthropogenic

carbon decreases. However, this is caused by the exchange of natural for anthropogenic carbon, due to the large exchange fluxes characterising the carbon cycle. This is highlighted in Figure 7 (b), which shows that at all times natural uptake, F_e , exceeds natural emissions, $F_i = F_i^0$, and so the natural environment is a net sink throughout. Finally, Figure 7 (c) illustrates the difference between the approximate and exact solutions to the differential equation (Eq. (8)). Clearly anthropogenic carbon decreases slightly more slowly than would be expected from the estimate of residence time given by the approximate solution, because the size of the atmospheric reservoir and environmental uptake are both decreasing with time.

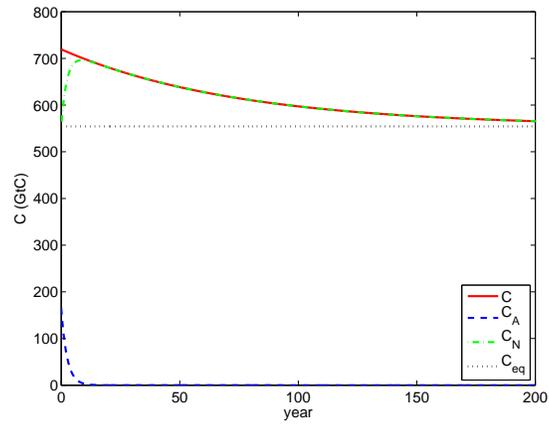
Modelling the Airborne Fraction

The *airborne fraction* is defined as the ratio of the increase in atmospheric CO_2 and CO_2 emissions from anthropogenic sources. A comparison of cumulative anthropogenic emissions from both fossil fuel consumption and land use changes and the growth in atmospheric CO_2 , shown in Figure 5, shows that the airborne fraction has remained fairly constant throughout the industrial era, with annual growth of approximately 45% of anthropogenic emissions. This is suggestive of a causal link between anthropogenic emissions and the observed rise in atmospheric CO_2 . However, a constant airborne fraction arises naturally from the single box model, as a consequence of exponential growth in anthropogenic emissions. If instead of a single slug of anthropogenic CO_2 injected into the atmosphere at time $t = 0$, we consider a scenario with exponential growth of anthropogenic emissions, F_a , the differential equation describing the single box model becomes

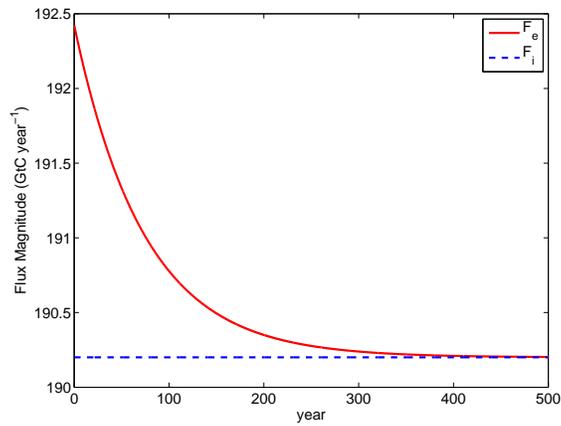
$$\frac{dC}{dt} + k_e C = F_i^0 - F_e^0 + F_a, \quad \text{where} \quad F_a = \lambda \exp\{\rho t\}.$$

Again this is a first order inhomogeneous differential equation, and can be solved by the method of integrating factors, with integrating factor

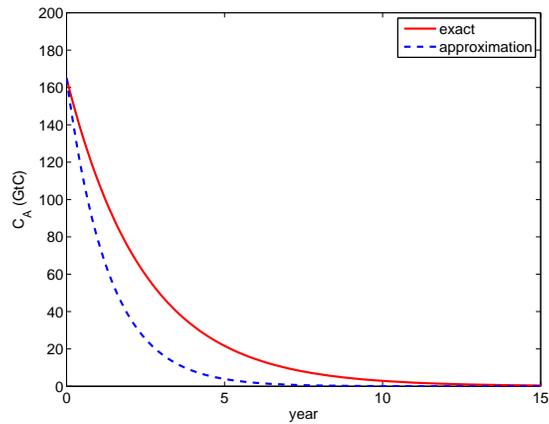
$$\mu(t) = \exp\left\{\int k_e dt\right\} = \exp\{k_e t\},$$



(a)



(b)



(c)

Figure 7: One-box simulation of a pulse of anthropogenic carbon injected into the atmosphere at time $t = 0$: (a) levels of natural, anthropogenic and total atmospheric carbon, (b) magnitudes of environmental uptake and emissions and (c) exact and approximate solutions to the differential equation (Eq. (8)).

such that

$$\begin{aligned}\exp\{k_e t\} C &= \int \exp\{k_e t\} [F_i^0 - F_e^0 + \lambda \exp\{\rho t\}] dt \\ &= \left[\frac{F_i^0 - F_e^0}{k_e} + \frac{\lambda}{k_e + \rho} \exp\{\rho t\} \right] \exp\{k_e t\} + K,\end{aligned}$$

and so, noting that $(F_i^0 - F_e^0)/k_e = C_{eq}$,

$$C = C_{eq} + \frac{\lambda}{k_e + \rho} \exp\{\rho t\} + K \exp\{-k_e t\}$$

The first term is simply the equilibrium mass of CO₂, the second term is the system response to anthropogenic emissions and the final term represents the response to an initial deviation from equilibrium. In this case, the system begins at equilibrium, so we take $K = 0$, and neglect the final term, giving

$$C = C_{eq} + \frac{\lambda}{k_e + \rho} \exp\{\rho t\} \quad (9)$$

Note that the response of the one-box model to exponential growth in anthropogenic emissions is an exponential growth in atmospheric CO₂, that is a constant fraction of anthropogenic emissions, the scaling factor depending on k_e and ρ , which is essentially what we observe in reality.

The next step is to estimate the constants, λ and ρ , describing the exponential growth of anthropogenic emissions. This is achieved by linear regression of $\log F_a$ as a function of time, i.e.,

$$\log F_a = \rho t + \log \lambda + \varepsilon, \quad \text{where} \quad \varepsilon \sim \mathcal{N}(0, \sigma^2).$$

Fitting this model to the available estimates of fossil fuel and land use emissions gives $\rho = 0.0188$ and $\log \lambda = -35.5432$. Figure 8 (a) shows total annual anthropogenic emissions from land use changes and fossil fuel consumption, along with the fitted exponential model, which appears to provide a subjectively adequate representation of estimated emissions. The airborne fraction is

then given by the ratio of cumulative emissions and atmospheric growth,

$$\frac{\int \lambda \exp\{\rho t\} dt}{\lambda \exp\{\rho t\}/(k_e + \rho)} = \frac{\lambda \exp\{\rho t\}/\rho}{\lambda \exp\{\rho t\}/(k_e + \rho)} = \frac{\rho}{k_e + \rho} \approx 0.5825.$$

Figure 8 (b) shows cumulative anthropogenic emissions and the change in atmospheric CO₂ from the pre-industrial equilibrium, and the values predicted by the single box model. The predicted value for the airborne fraction, at $\approx 58\%$ of anthropogenic emissions is rather higher than the observed value of $\approx 45\%$, however given that the single box model adapted from ES09 provides only the most crude representation of the carbon cycle, this result provides good support for the assertion of a constant airborne fraction and for the causal relationship between anthropogenic emissions and the observed rise in atmospheric CO₂.

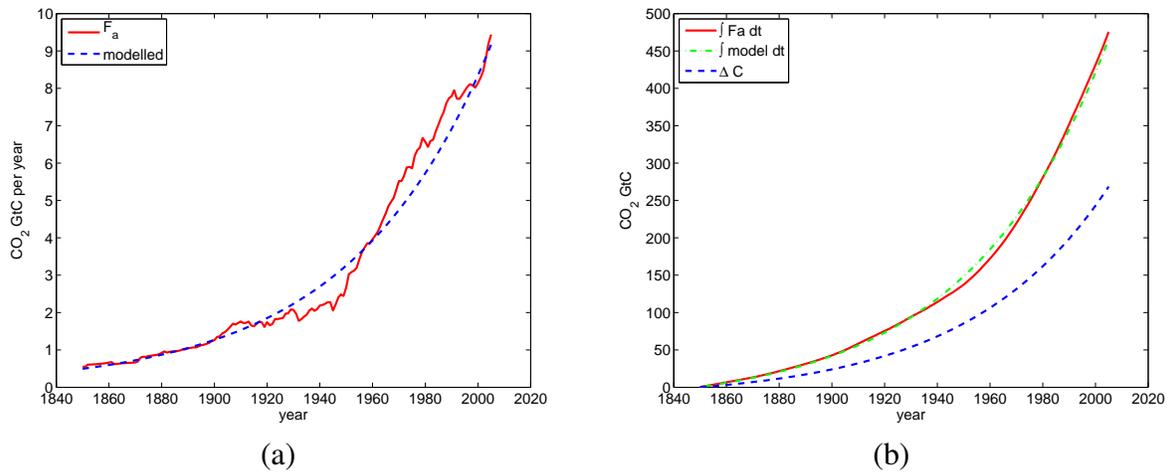


Figure 8: Regression analysis of the net environmental flux as a function of atmospheric carbon dioxide concentration.

Proportion of Anthropogenic Emissions Remaining in the Atmosphere

The airborne fraction is often described as representing the proportion of anthropogenically sourced CO₂ that remains in the atmosphere, however that may be somewhat misleading. The very large exchange fluxes mean that the residence time of CO₂ in the atmosphere is short, and hence anthropogenically sourced CO₂ is soon exchanged for environmentally sourced CO₂. As a result

the proportion of molecules of CO₂ comprising the excess that is of anthropogenic origin is much smaller than the airborne fraction. The purpose of the airborne fraction is to describe the effect of anthropogenic emissions on the excess over equilibrium levels, rather than the composition of the excess. The proportion of anthropogenic emissions that remains in the atmosphere can also be estimated using the single box model (although again the estimate will be crude as the model is too simple to adequately capture the dynamic behaviour of the carbon cycle). Under exponentially increasing anthropogenic emissions, the amount of CO₂ of anthropogenic origin remaining in the atmosphere is given by the following differential equation,

$$\frac{dC_A}{dt} = \lambda \exp\{\rho t\} - \frac{C_A F_e}{C} \quad (10)$$

As an engineer's "back of an envelope" approximation, we could assume that f_e and C are approximately constant at their equilibrium values, such that,

$$\frac{dC_A}{dt} = \lambda \exp\{\rho t\} - \frac{C_A F_e^0}{C_{eq}} \quad (11)$$

This is a simple inhomogeneous linear differential equation and can be solved by the method of integrating factors, giving the solution,

$$C_A \approx \frac{\lambda}{k_r + \rho} \exp\{\rho t\}$$

The fraction of the excess of CO₂ above its equilibrium value is then

$$\frac{C_A}{C - C_{eq}} \approx \frac{\frac{\lambda}{k_r + \rho} \exp\{\rho t\}}{\frac{\lambda}{k_e + \rho} \exp\{\rho t\}} = \frac{k_r + \rho}{k_e + \rho} \approx 0.0928,$$

likewise, the proportion of atmospheric CO₂ of directly anthropogenic origin is given by

$$\frac{C_A}{C} \approx \frac{\frac{\lambda}{k_r + \rho} \exp\{\rho t\}}{C_{eq} + \frac{\lambda}{k_e + \rho} \exp\{\rho t\}}, \quad \text{such that} \quad \left. \frac{C_A}{C} \right|_{t=2010} = 0.0334.$$

Thus the simple one-box model predicts that, if the observed rise is of purely anthropogenic origin, we should expect only $\approx 9\%$ of the increase to be composed of molecules of carbon dioxide of anthropogenic origin, and only $\approx 3.34\%$ of total atmospheric CO_2 . Eq. (10) is not easily solved analytically, however the amount of anthropogenic CO_2 in the atmosphere can still be estimated using numerical ordinary differential equation solvers, such as the `ode23` routine in MATLAB. Figure 9 shows the results obtained using approximate and numerical solutions; the numeric solution suggests a slightly higher proportion of anthropogenic CO_2 remains in the atmosphere.

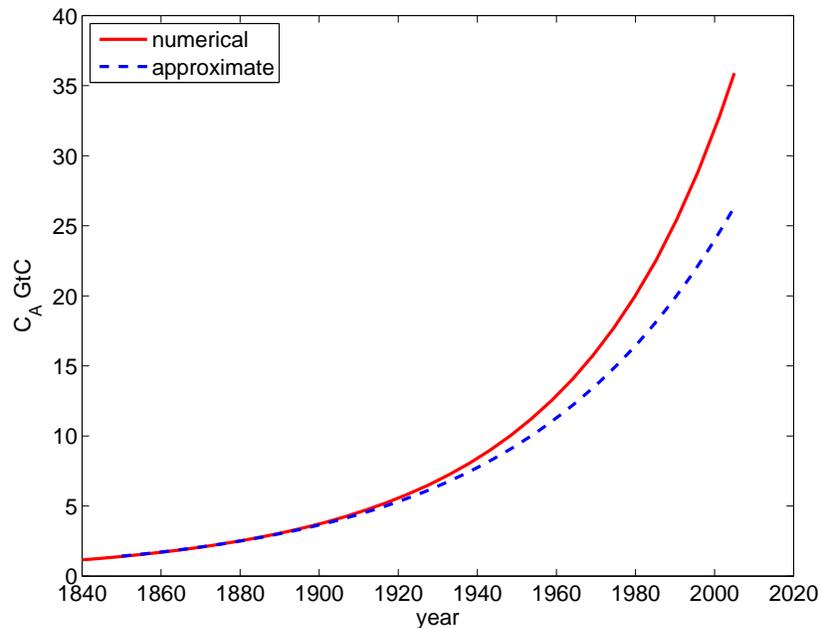


Figure 9: Carbon dioxide of anthropogenic origin as a function of time, estimated using approximate and numeric solutions to Eq. (10).

Limitations of the One Box Model

The simple one box model of the carbon cycle used in this paper is sufficient to demonstrate that a short residence time of atmospheric carbon dioxide is fully consistent with the generally accepted anthropogenic origin of the observed post industrial increase in the mass of the atmospheric reservoir. The one box model is however inadequate for accurate quantitative analysis of the carbon cycle as key components of the natural carbon cycle are excluded from the model.

The one box model treats all of the carbon released into the atmosphere by natural sources as being of purely “natural” origin, however in reality this is not actually the case. The annual uptake by the oceanic and terrestrial reservoirs will include some anthropogenic carbon, and so as time progresses those reservoirs will become, like the atmospheric reservoir, increasingly contaminated with anthropogenic CO₂, and hence emissions from natural sources will be a mixture of “natural” and anthropogenic carbon. As a result the model is likely to underestimate the proportion of anthropogenic carbon in the atmosphere, c.f. Figure 7 (a). A second box, representing the combined oceanic and terrestrial reservoirs would result in a more accurate quantitative analysis, however this is not required to establish the conclusions drawn in this paper.

The one box model presented here is essentially a first order linear approximation to the action of the carbon cycle, fitted to current conditions, giving an exponential recovery from the injection of a pulse of carbon dioxide with a single, fixed time constant. However the ability of the surface ocean to absorb carbon dioxide from the atmosphere will decrease as the concentration of dissolved carbon increases, rather than remain constant, and so the one-box model substantially underestimates the actual adjustment time. Essentially, the ocean can be thought of as being composed of a number of different components, for example a mixed layer (approximately the upper 100m), a thermocline (approximately the upper 500 – 1000m) and the deep ocean. As a result, the processes governing the uptake of carbon from the atmosphere operate on a number of different timescales. The initial uptake of carbon dioxide from the atmosphere by the mixed layer is rapid, and might be modelled by an exponential decay on an approximately annual timescale. However as the dissolved carbon concentration in the mixed layer rises, the absorption rate depends more strongly on the rate of transport of carbon from the mixed layer to the remainder of the thermocline, with a decadal exponential timescale. On longer timescales, the uptake of carbon depends on the transport of carbon from the thermocline to the deep ocean, which takes place on a centennial timescale. The one-box model used here, being calibrated on only ≈ 50 years of observations represents only the uptake of carbon from the atmosphere into the thermocline. A more realistic estimate of adjustment time requires the use of a box-diffusion model,²⁹ that accounts for

the transport of carbon into the deep ocean. The response of the carbon cycle to the injection of a pulse of CO₂ can also be reasonably be approximated by a sum of exponentials, with different time-constants reflecting the range of timescale on which the physical mechanisms involved operate.³⁰ Even uptake of CO₂ by the deep ocean will not fully restore atmospheric carbon dioxide levels back to their pre-industrial equilibrium; anthropogenic emissions have increased the total amount of carbon in the active carbon cycle and a fraction of that additional carbon will remain in the atmosphere after the atmospheric, oceanic and terrestrial reservoirs have fully equilibrated. A full return to pre-industrial levels will require the removal of the carbon from the active carbon cycle via chemical weathering, which permanently sequesters the carbon in the lithosphere. This process takes place on a timescale of tens of thousands of years.⁴

Conclusions

In this paper we have explained why the short residence time of CO₂ in the atmosphere has little bearing on the origin of the observed rise in atmospheric carbon dioxide since the start of the industrial revolution. As a result, the conclusion drawn in ES09, that the short residence time means this increase cannot be due to anthropogenic emissions is a *non-sequitur*, and furthermore, the mass balance argument demonstrates it to be false. We do not question the estimates of residence time of 5-15 years given in ES09, as this is essentially uncontroversial, indeed similar figures are given in the IPCC scientific assessment reports, and so can be considered widely accepted. However, any conclusion that controls on carbon emissions are unnecessary, or would be unsuccessful, on the basis of the short residence time of atmospheric carbon dioxide are ill-founded and deeply misleading and thus potentially dangerous if used as the basis for policy.

Acknowledgements

I thank the anonymous reviewers, Robert Essenhigh, Philip Goodwin, Riccardo Reitano, Nigel Franks, Andy Skuce, and John Cook for their helpful discussion and comments on previous drafts

of this manuscript.

Notes and References

- (1) Essenhight, R. H. *Energy and Fuels* **2009**, *23*, 2773–2784.
- (2) Houghton, J. T., Jenkins, G. J., Ephraïm, J. J., Eds. *Climate Change - The IPCC Scientific Assessment*; Cambridge University Press: Cambridge, U.K., 1990.
- (3) Solomon, S., Qin, D., Manning, M., Marquis, M., Averyt, K., Tignor, M. M. B., Miller Jr., H. L., Chen, Z., Eds. *Climate Change 2007 : The Physical Science Basis - Working Group I Contribution to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press: Cambridge, U.K., 2007.
- (4) Archer, D.; Eby, M.; Brovkin, V.; Ridgwell, A.; Cao, L.; Mikolajewicz, U.; Caldeira, K.; Matsumoto, K.; Munhoven, G.; Montenegro, A.; Tokos, K. *Annual Review of Earth and Planetary Sciences* **2009**, *37*, 117–134.
- (5) Denman, K. L.; Brasseur, G.; Chidthaisong, A.; Ciais, P.; Cox, P. M.; Dickinson, R. E.; Hauglustaine, D.; Heinze, C.; Holland, E.; Jacob, D.; Lohmann, U.; Ramachandran, S.; da Silva Dias, P. L.; Wofsy, S. C.; Zhang, X. In *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., Miller, H. L., Eds.; Cambridge University Press: Cambridge, United Kingdom and New York, NY, USA., 2007.
- (6) Unless indicated otherwise, a positive flux implies the flow of carbon into the atmospheric reservoir.
- (7) Archer, D. *The Global Carbon Cycle*; Princeton Primers in Climate; Princeton University Press, 2010.

- (8) In this paper, quantities of carbon will be expressed in terms of *GtC* (gigatons of carbon), rather than *GtCO₂* (gigatons of carbon dioxide), as carbon does not exist solely in the form of carbon dioxide in any of the reservoirs, although it is the dominant form in the atmosphere.
- (9) <http://cdiac.ornl.gov/>.
- (10) Etheridge, D. M.; Steele, L. P.; Langenfelds, R. L.; Francey, R. J.; J.-M., B.; Morgan, V. I. Historical CO₂ records from the Law Dome DE08, DE08-2, and DSS ice cores. In Trends: A Compendium of Data on Global Change, Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tennessee, U.S.A., 1998.
- (11) <http://cdiac.ornl.gov/ftp/trends/co2/lawdome.smoothed.yr20>
(downloaded April 2, 2010).
- (12) Keeling, R. F.; Piper, S. C.; Bollenbacher, A. F.; Walker, J. S. Atmospheric CO₂ records from sites in the SIO air sampling network. In Trends: A Compendium of Data on Global Change, Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tennessee, U.S.A., 2009.
- (13) <http://cdiac.ornl.gov/ftp/trends/co2/maunaloa.co2> (downloaded April 2, 2010).
- (14) Boden, T. A.; Marland, G.; Andres, R. J. Global, Regional, and National Fossil-Fuel CO₂ Emissions. Carbon Dioxide Information Analysis Center, Oak Ridge Laboratory U.S. Department of Energy, Oak Ridge, Tennessee, U.S.A., 2009.
- (15) http://cdiac.ornl.gov/ftp/ndp030/global.1751_2008.ems (downloaded April 2, 2010).
- (16) Houghton, R. A. Carbon flux into the atmosphere from land-use changes: 1850-2005. In TRENDS: A Compendium of Data on Global Change, Carbon Dioxide Information Analysis

Center, Oak Ridge Laboratory U.S. Department of Energy, Oak Ridge, Tennessee, U.S.A., 2008.

- (17) <http://cdiac.ornl.gov/trends/landuse/houghton/1850-2005.txt>
(downloaded April 2, 2010).
- (18) Raupach, M. R.; Canadell, J. G.; Le Quéré, C. *Biogeosciences* **2008**, *5*, 1603–1613.
- (19) Battle, M.; Bender, M. L.; Tans, P. P.; White, J. W. C.; Conway, T.; Francey, R. J. *Science* **2000**, *287*, 2467–2470.
- (20) Tans, P. P.; de Jong, A. F. M.; Mook, W. G. *Nature* **1979**, *280*, 826–828.
- (21) Barnola, J. M.; Raynaud, D.; Korotkevich, Y. S.; Lorius, C. *Nature* **1987**, *329*, 408–414.
- (22) http://www.skepticalscience.com/Mackie_OA_not_OK_post_1.html.
- (23) <http://www.realclimate.org/index.php/archives/2004/12/how-do-we-know-that-recent-cosub2sub-increases-are-due-to-human-activities-updated/>.
- (24) <http://www.skepticalscience.com/co2-residence-time.htm>.
- (25) http://www.ferdinand-engelbeen.be/klimaat/co2_measurements.html.
- (26) United States Environmental Protection Agency, Endangerment and Cause or Contribute Findings for Greenhouse Gases Under Section 202(a) of the Clean Air Act: EPA's Response to Public Comments, Volume 2: Validity of Observed and Measured Data. 2009.
- (27) The net environmental sink is thought to be a response to all previous and current anthropogenic emissions; in the absence of significant anthropogenic emissions the carbon cycle, and thus atmospheric carbon dioxide concentrations, would have remained close to their pre-industrial equilibrium states. We would only expect CO₂ levels to be falling in the case where previous anthropogenic emissions had perturbed the carbon cycle from its equilibrium state,

but where current and ongoing anthropogenic emissions had ceased. In that scenario, the various feedback mechanisms comprising the carbon cycle would act to bring the atmospheric carbon dioxide concentration back towards its equilibrium level.

(28) http://www.ferdinand-engelbeen.be/klimaat/co2_measurements.html.

(29) Siegenthaler, U. *Journal of Geophysical Research* **1983**, 88, 3599–3608.

(30) Caldiera, K.; Kasting, J. F. *Nature* **1993**, 366, 251–253.