

# What Controls the Atmospheric $CO_2$ Level?

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## Abstract:

The evolution of nuclear-perturbed  $^{14}CO_2$  is used to determine the removal time of atmospheric  $CO_2$ . The exponential decline of anomalous  $^{14}CO_2$  establishes that absorption of  $CO_2$  is determined, not by extraneous reservoirs of carbon, but autonomously by the atmosphere. Specifically, the rate at which  $CO_2$  is absorbed from the atmosphere is directly proportional to the instantaneous abundance of  $CO_2$  in the atmosphere. It operates with a single time scale, which reflects the collective absorption by all sinks of  $CO_2$  at the Earth's surface. The long-term decline of anomalous  $^{14}CO_2$  reveals an effective absorption time of about 10 years. The accompanying removal of atmospheric  $CO_2$  is much faster than has been presumed to interpret observed changes. Jointly with the Conservation Law governing atmospheric  $CO_2$ , that absorption time is shown to reproduce the observed evolution of  $CO_2$ , inclusive of its annual cycle. The latter treatment provides an upper bound on the absorption time, independent of but consistent with the value revealed by the decline of anomalous  $^{14}CO_2$ . Together, the two determinations of absorption provide an upper bound on the anthropogenic perturbation of atmospheric  $CO_2$ .

**Keywords:** *Carbon cycle;  $CO_2$  residence time; anthropogenic emissions; radiocarbon measurements; seasonal  $CO_2$  variations*

## 1. Introduction

A central question in the climate science of today is: How much does anthropogenic emission of  $CO_2$  contribute to rising atmospheric  $CO_2$  and, thereby, to global warming? The answer to this question requires a quantitative understanding of  $CO_2$  exchange between the atmosphere and the Earth's surface, which removes  $CO_2$  from the atmosphere.

A popular metric of such exchange is the residence time of  $CO_2$ , which characterizes how long  $CO_2$  remains in the atmosphere before being absorbed at the Earth's surface. In its Fifth Assessment Report (AR5-Ch. 6) [1], the UN's Intergovernmental Panel on Climate Change (IPCC) defines multiple residence times, as well as adjustment times. They represent exchanges between extraneous carbon reservoirs at or beneath the Earth's surface. Unlike the atmosphere, those global reservoirs are virtually unobserved, leaving their exchanges largely a matter of speculation. Such time scales are relevant to the storage and sequestration capacity of those reservoirs. However, they are of no direct relevance to  $CO_2$  in the atmosphere - because its abundance is dictated solely by transfers into and out of the atmosphere at the Earth's surface. What transpires to carbon outside of the atmosphere is immaterial.

Residence time is, in fact, incidental to the physics that controls atmospheric  $CO_2$ . Because  $CO_2$  is conserved in the atmosphere, its abundance is determined entirely by emission and absorption of  $CO_2$  at the Earth's surface. Residence time does not determine absorption of  $CO_2$ ; it is determined by it.

Atmospheric  $CO_2$  is governed by the 3D Continuity Equation. It embodies the Conservation Law for atmospheric carbon. Except for miniscule differences (much smaller than the observational uncertainty of global  $CO_2$ ), absorption of atmospheric carbon dioxide does not discriminate between  $CO_2$  of human origin and  $CO_2$  of natural origin. As discussed previously (Harde, 2017 [2] and 2019 [3]), absorption channels at the Earth's surface (e.g., vegetation, soil, and ocean) operate in parallel.

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Their collective impact on atmospheric  $CO_2$  is represented in the total absorptivity:  $\alpha = \alpha_1 + \alpha_2 + \alpha_3 \dots$ . Its inverse is the *direct absorption time* of atmospheric  $CO_2$ ,  $\tau$ , which characterizes its direct removal from the atmosphere. Because  $CO_2$  is virtually conserved in the atmosphere, it is produced and destroyed only at the Earth's surface. The direct absorption time of  $CO_2$  is therefore equal to its *residence time*:

$$\tau = \alpha^{-1} = \tau_R. \quad (1)$$

The direct absorption rate of atmospheric  $CO_2$  is determined by  $\tau$  and its instantaneous concentration,  $C_{CO_2}$ :

$$\mathcal{A} = C_{CO_2} / \tau. \quad (2)$$

Common in physical systems, the dependence of  $CO_2$  removal on  $CO_2$  abundance is an empirical feature of atmospheric carbon dioxide. It is documented in the monotonic decline of nuclear-perturbed carbon 14, an isotopic tracer of atmospheric  $CO_2$  (Salby, 2015 [4]). Following the 1963 *Limited Test Ban Treaty* [5],  $^{14}CO_2$  declined exponentially according to a single absorption time.

Another physical inconsistency in AR5 is its arbitrary division of the carbon budget into a native part, which is presumed to have remained constant before the industrial era, and an anthropogenic part, which is presumed to be solely responsible for increasing  $CO_2$ . The two arbitrarily-defined components are presumed to be independent and, somehow, distinguished by absorption processes. A consequence of the different treatment of these arbitrarily-defined components is that, when recombined, they no longer satisfy the Equivalence Principle of physics and the Conservation Law of atmospheric  $CO_2$  - physical laws that *are* satisfied by  $CO_2$  in the real atmosphere (Salby, 2018 [6]; Harde, 2019 [3], *Sec. 3.4*).

Moreover, models relied upon by the IPCC rest upon an unphysical premise: They assume that absorption of anthropogenic  $CO_2$  is proportional, not to its instantaneous abundance (2), but to its instantaneous emission rate (e.g., Joos et al., 1988 [7]) – irrespective of how much  $CO_2$  is actually in the atmosphere. A consequence of this premise is that  $CO_2$  continuously accumulates in the atmosphere, regardless of its actual abundance. In the presence of real absorption, such behavior is impossible. For constant emission,  $CO_2$  would eventually reach an equilibrium level, at which it is removed through absorption as fast as it is introduced through emission (Salby 2016 [8]; Harde 2017 [2], 2019 [3]; Berry 2019 [9]).

Numerous investigations have sought estimates of the absorption time. Most range between 5 and 15 years. However, some are as short as 1 year (see e.g. compilations by Sundquist 1985 [10] and Segalstad 1996 [11]; Murray  $\tau = 5.4$  yr (1992) [12]; Segalstad  $\tau = 5.4$  yr (1992) [13]; Broecker et al.  $\tau \approx 8$  yr (1979) [14]; Humlum et al.  $\tau \approx 1$  yr (2013) [15]; Salby  $\tau \approx 1$  yr (2013) [16]; Harde  $\tau \leq 4$  yr (2017, 2019) [2, 3]). The residence time has also been alleged to exceed a thousand years (Solomon et al, 2009) [17]. Many of the estimates rest upon the observed decline of the isotopic tracer  $^{14}CO_2$ , which (aside from miniscule differences) experiences the same absorption as overall  $CO_2$  (e.g., Revelle & Suess  $\tau = 7$  yr (1957) [18]; Craig  $\tau = 7$  yr (1957) [19]; Bacastow & Keeling  $\tau = 6.3 - 7$  yr (1973) [20]; Keeling & Bacastow  $\tau = 7.5$  yr (1977) [21]; Siegenthaler  $\tau = 4 - 9$  yr (1989) [22]); Stuiver  $\tau = 6.8$  yr (1980) [23]). Some authors only specify the decay time  $\tau_{\Delta^{14}C}$  for the fractional departure of anomalous  $^{14}C$  from a reference abundance,  $\Delta^{14}C$  (Levin et al.  $\tau_{\Delta^{14}C} = 8.5$  yr (1980, 1994) [24, 25]; Levin et al.  $\tau_{\Delta^{14}C} = 15$  yr (2013) [26]; Hua et al.  $\tau_{\Delta^{14}C} = 16.5$  yr (2013) [27]; Turnbull et al.  $\tau_{\Delta^{14}C} = 16.5$  yr (2017) [28]).

Absorption of atmospheric  $CO_2$  determines if and how fast  $CO_2$  accumulates in the atmosphere. Current views on this fundamental process rest, in large part, on observations of  $^{14}C$ . Yet, interpretations of those data vary widely, as reflected in the range of estimated absorption times. The uncertainty in this key property is underpinned by the definition of  $^{14}C$  itself, which also varies widely. Depending upon normalization, reference concentration, and units, carbon 14 has been quantified via “a jungle” of different definitions (Stenström et al. 2011 [29]). Relevant to the

absorption time of  $CO_2$  is normalization in some definitions of carbon 14 by carbon 12, which is the principal constituent of  $CO_2$  (Andrews 2020 [30]).

Here, we examine the impact of such normalization and use the observed decline of nuclear-perturbed  $^{14}CO_2$  to determine the actual absorption time of atmospheric  $CO_2$ . Jointly with the Conservation Law governing atmospheric  $CO_2$ , that absorption time is then shown to reproduce the observed evolution of  $CO_2$ , inclusive of its annual cycle. The latter treatment provides an upper bound on the absorption time, independent of but consistent with the value revealed by the decline of anomalous  $^{14}CO_2$ . Thereby, it provides an upper bound on the anthropogenic perturbation of atmospheric  $CO_2$ .

## 2. Perturbation of $^{14}CO_2$

The carbon isotope  $^{14}C$  has a radioactive half-life of 5730 yrs (an e-folding time for radioactive decay of 8267 yrs). On the time scale of the instrumental record of  $CO_2$  (decades),  $^{14}C$  is therefore conserved in the troposphere. Once introduced, it is neither produced nor destroyed. Like all  $CO_2$ ,  $^{14}CO_2$  is removed only through absorption at the Earth's surface.  $^{14}C$ , however, is continuously formed in the upper atmosphere from  $^{14}N$  via interaction with neutrons that are liberated by cosmic radiation, before  $^{14}C$  rapidly oxidizes into  $^{14}CO_2$ .

Nuclear testing during the 1950s and early 1960s dramatically increased free neutrons in the stratosphere. It eventually led to a large perturbation of  $^{14}C$  in the troposphere, where  $^{14}C$  is measured – an increase in  $\Delta^{14}C$  of order 100%. The cessation of atmospheric testing after 1963 [5] witnessed a systematic decline of the nuclear perturbation. The observed decline followed, not from radioactive decay, but from the absorption of  $^{14}CO_2$  at the Earth's surface – the same process that absorbs all  $CO_2$ .

### 2.1 Quantification of Carbon 14

Definitions of  $^{14}C$  follow from measurements of decay activity  $A$ , which is reported in counts. The quantity commonly relied upon in climate studies is *anomalous*  $^{14}C$ , the fractional departure from a reference abundance:

$$\Delta^{14}C = \left( \frac{A_{SN}}{A_{ABS}} - 1 \right) \cdot 1000, \quad (3)$$

where  $A_{SN}$  and  $A_{ABS}$  denote, respectively, the normalized activity of a measured sample and an absolute reference activity. Reported in ‰,  $\Delta^{14}C$  measures the deviation of  $^{14}C$  from the reference concentration. However, even the definition (3) is not universal (Stenström et al. 2011 [29]). In the original definition, the sample activity,  $A_{SN}$ , is normalized for the counting volume, change of mass, and dilution or impurity (see e.g., Stuiver & Polach [31]). But with the invention of accelerator mass spectrometry many groups are now measuring the fractional number density of  $^{14}C$  to  $^{12}C$  atoms; cf. Turnbull et al. (2017) [28]. The increasing concentration of  $^{12}CO_2 \cong CO_2$  must then be accounted for to recover the true concentration of  $^{14}C$ .

The decline of anomalous  $^{14}C$  has been the basis for estimates of  $CO_2$  absorption time (Sec. 1). In many investigations, however, it is unclear whether absorption time was deduced from measurements of  $^{14}C$  based on the original definition or from values normalized by  $^{12}C$ , which increased during the post-testing era with  $CO_2$ .

### 2.2 Observed Decline of Anomalous $^{14}CO_2$

Figure 1 presents the record of tropospheric  $^{14}CO_2$  measured at Vermont, Austria (Blue) (Levin et al. [24, 25], also archived at CDIAC [32]). Extending from 1959 until 1983, the Vermont record of  $\Delta^{14}C$  is recorded in ‰ deviation from the absolute international standard activity  $A_{ABS}$ . Following an abrupt increase in the early 1960s,  $\Delta^{14}C$  underwent a systematic, albeit unsteady, decline. Initial

years are punctuated by half a dozen re-enrichments, which appear annually. Thereafter,  $\Delta^{14}\text{C}$  declined more gradually.

The archived data in Fig 1 are normalized by  $^{12}\text{C}$ , which distorts the actual concentration of  $^{14}\text{C}$  according to the variation of  $\text{CO}_2$ . To quantify the effect of such normalization, particularly over the first 20 years after the test ban treaty, when  $^{14}\text{CO}_2$  was observed at Vermont, those data have been re-normalized to the initial (constant) atmospheric  $\text{CO}_2$  concentration at time  $t_0 = 1959$ ,  $C_0 = C(t_0)$ . In terms of the instantaneous  $\text{CO}_2$  concentration at sample time  $t_s$ ,  $C_s = C(t_s)$ , anomalous  $^{14}\text{C}$  then has the actual or corrected concentration:

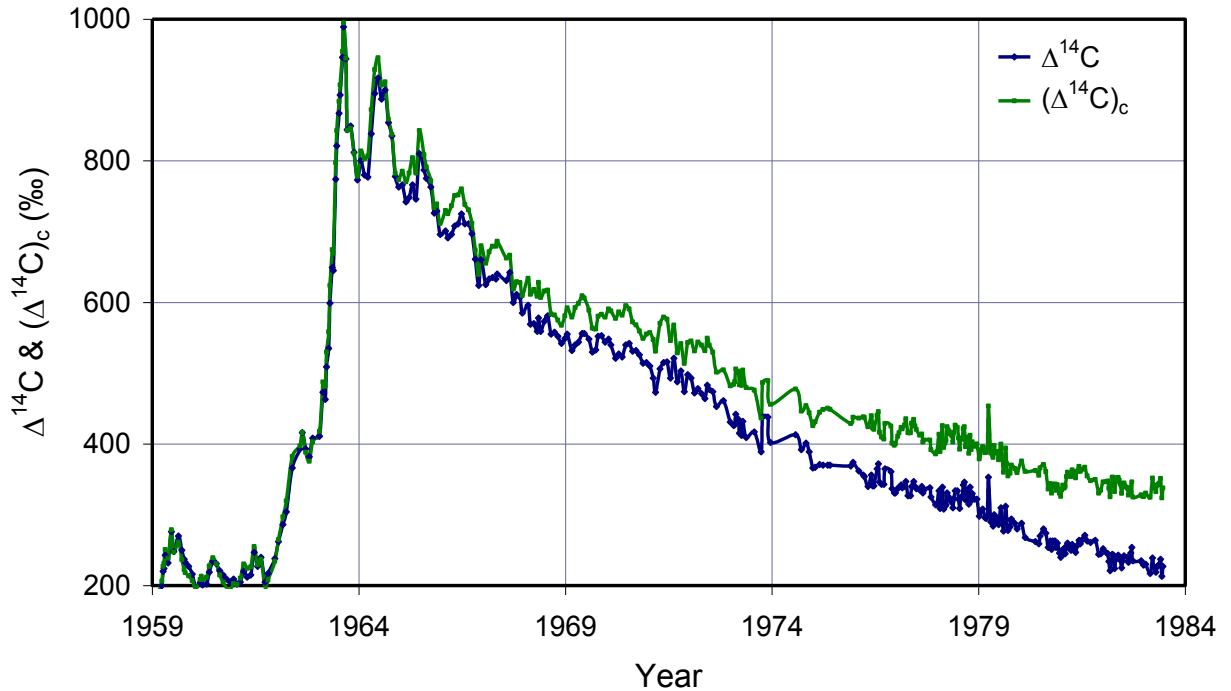
$$(\Delta^{14}\text{C})_c = \left( \frac{A_{SN}}{A_{ABS}} \cdot \frac{C_s}{C_0} - 1 \right) \cdot 1000. \quad (4)$$

Expressing the normalized sample activity  $A_{SN}$  in terms of the original  $\Delta^{14}\text{C}$  data, with  $A_{SN} = (\Delta^{14}\text{C}/1000 + 1) \cdot A_{ABS}$ , gives:

$$(\Delta^{14}\text{C})_c = \left( \left( \frac{\Delta^{14}\text{C}}{1000} + 1 \right) \cdot \frac{C_s}{C_0} - 1 \right) \cdot 1000. \quad (5)$$

For the instantaneous  $\text{CO}_2$  concentration  $C_s$ , we use the monthly Mauna Loa record, also archived at CDIAC [32], which was adapted to the slightly different sample times of the  $^{14}\text{C}$  measurements. Superimposed in Figure 1 is the corrected concentration,  $(\Delta^{14}\text{C})_c$  (Green). It undergoes the same fast decline over the first half dozen years after the test ban treaty, followed by more gradual decline thereafter. Noteworthy is the almost-pure exponential form of the long-term decline. The rate of  $^{14}\text{CO}_2$  removal is then directly proportional to its instantaneous abundance, in confirmation of (2). With e-folding time of about 10 yrs, the decline is nearly the same in both records. It differs only in a somewhat different equilibrium level to which the two metrics of  $^{14}\text{CO}_2$  approach.

To understand the observed decline, we evaluate  $^{14}\text{CO}_2$  through the Conservation Law governing it. Relative to its initial concentration in 1959, anomalous  $^{14}\text{CO}_2$  is represented in (4) by the relative concentration  $C'_{14} = (A_{SN}/A_{ABS}) \cdot (C_s/C_0)$ .



**Figure 1:** Observed record of  $\Delta^{14}\text{C}$  data (Blue) sampled at Vermont and corrected concentration  $(\Delta^{14}\text{C})_c$  (Green).

$C'_{14}$  can change through changes of natural production in the stratosphere, described by the relative rate  $e'_{N,14}$ . More important is its absorption at the Earth's surface, which is proportional to its instantaneous abundance  $C'_{14}$  (2). Of  $^{14}CO_2$  that is removed from the atmosphere with the direct absorption time  $\tau$ , a fraction  $\beta$  is returned to the atmosphere through re-emission from the Earth's surface (e.g., via outgassing and decomposition of vegetation). Re-emission represents an additional source

$$e'_{R,14} = \beta \cdot C'_{14} / \tau, \quad (6)$$

which offsets direct absorption. Collecting these opposing influences gives the net absorption, which, with (2), operates with the *effective absorption time*

$$\tau_{eff} = \tau / (1 - \beta). \quad (7)$$

In terms of these influences, the Conservation Law governing the anomalous concentration  $C'_{14}$  becomes (cf. Harde 2019, Appendix B [3]):

$$\frac{dC'_{14}}{dt} = e'_{N,14} - \frac{C'_{14}}{\tau_{eff}}. \quad (8)$$

Integrating (8) reproduces the long-term decline of anomalous  $^{14}C$  introduced by nuclear testing. However, it does not describe oscillations that are apparent during the initial half dozen years (Figure 1). Those oscillations reflect annual re-enrichments of tropospheric  $^{14}CO_2$  from the stratosphere. Such transport occurs through the Brewer-Dobson circulation (BDC) of the stratosphere, an equator-to-pole overturning that intensifies each year during late winter and spring; see, e.g., Holton (2004) [33], Salby (2012) [34]. The intensified BDC transports  $^{14}C$ -enriched stratospheric air downward into the Arctic troposphere.

To account for such enhancement, (8) is augmented by an oscillatory source with radial frequency  $\omega$  that decays with the time scale  $\tau_E$ , which reflects the gradual exhaustion of anomalous  $^{14}CO_2$  in the stratosphere. The Conservation Law then becomes:

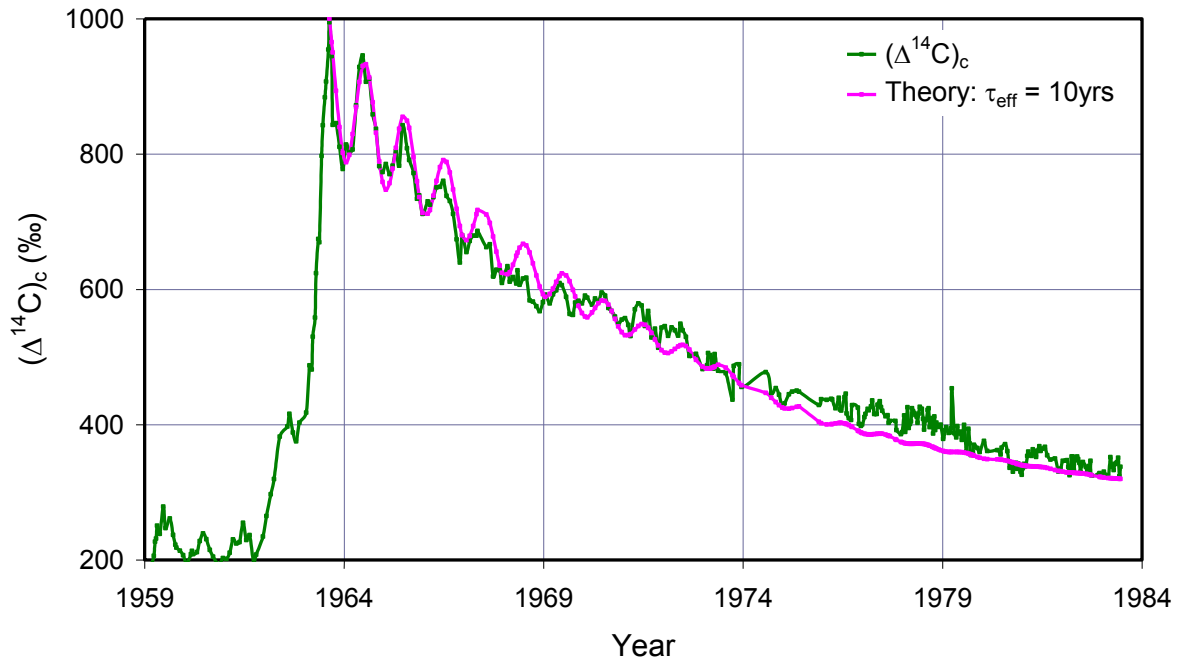
$$\frac{dC'_{14}}{dt} = e'_{N,14} + m' \cdot \cos(\omega \cdot t + \varphi) \cdot e^{-t/\tau_E} - \frac{C'_{14}}{\tau_{eff}}. \quad (9)$$

Equation (9) is integrated numerically, with an effective absorption time  $\tau_{eff} = 10$  yrs, natural  $^{14}CO_2$  production that accounts for a change from the pre-test era  $e'_{N,14} = 123$  ‰/yr, with  $m' = 0.65$  yrs<sup>-1</sup> and  $\tau_E = 4$  yrs. The calculated  $C'_{14}$  is then transformed back to anomalous carbon 14 that is referenced against the standard activity (5).

Plotted in Fig 2 is calculated  $(\Delta^{14}C)_C$  (Magenta). It tracks the evolution of observed  $(\Delta^{14}C)_C$ , which is superimposed (Green). Reproduced by the perturbed Conservation Law (9) is the long-term decline of  $(\Delta^{14}C)_C$ , as well as its repeated enhancement during the initial half dozen years following the test ban treaty.

With a single effective absorption time of  $\tau_{eff} = 10$  yrs, calculated  $(\Delta^{14}C)_C$  reproduces the salient features of the observed evolution. Direct absorption, however, is considerably faster. For a re-emission fraction  $\beta = 0.6$ , the time scale of direct absorption (7) is only  $\tau = 4.0$  yrs. That time scale is consistent with absorption evaluated from the global balance between total emission and absorption of  $CO_2$  (Harde [2, 3]), as well as the observed decline of annual oscillations. Both reflect absorption that is an order of magnitude faster than the adjustment times used by the IPCC to interpret changes of  $CO_2$ .

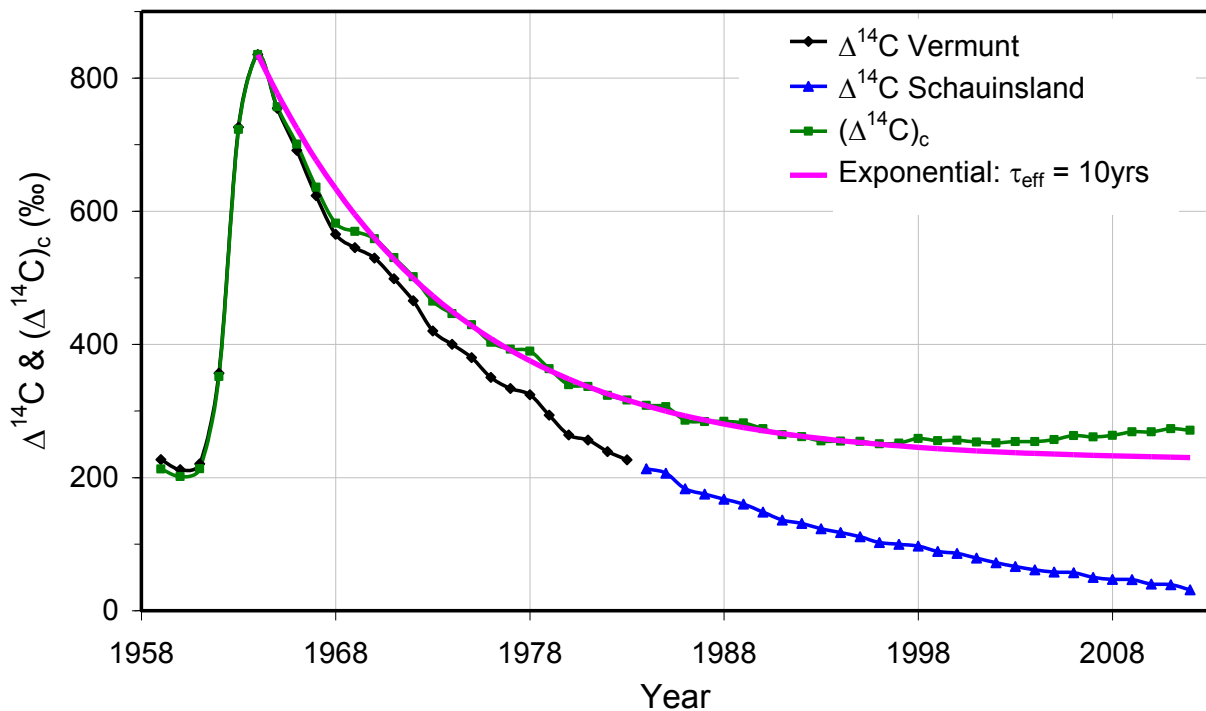
The observed evolution is recovered by absorption that operates on a single time scale,  $\tau_{eff} = 10$  years, and is proportional to the instantaneous abundance of  $^{14}CO_2$ . The calculated evolution of  $^{14}CO_2$  demonstrates that multiple adjustment times, invented by the IPCC, are superfluous and, more importantly, are incongruous with the physics that actually controls atmospheric  $CO_2$ .



**Figure 2:** Anomalous  $^{14}\text{C}$  measured at Vermont,  $(\Delta^{14}\text{C})_c$  (Green), compared against calculated  $(\Delta^{14}\text{C})_c$  with an effective absorption time of  $\tau_{\text{eff}} = 10$  yrs (Magenta).

### 2.3 Extended Decline

Observations of  $\Delta^{14}\text{C}$  at Vermont end in 1983. Observations at Schauinsland Germany (Levin 2013 [26]) enable them to be extended almost continuously to 2012. Over durations longer than 20 years, normalization by increasing  $^{12}\text{C}$  leads to greater distortion of  $\Delta^{14}\text{C}$ . The prolonged decline also brings carbon 14 closer to its unperturbed equilibrium level.



**Figure 3:** Annual-mean  $\Delta^{14}\text{C}$  observed at Vermont (Black) and Schauinsland (Blue), along with the  $^{12}\text{C}$ -corrected data,  $(\Delta^{14}\text{C})_c$  (Green). Superimposed is pure exponential decline with an effective absorption time  $\tau_{\text{eff}} = 10$  yrs (Magenta).

Figure 3 displays the collective record of annual-mean  $\Delta^{14}\text{C}$  measured at Vermont (Black) and Schauinsland (Blue). Superimposed is corrected anomalous carbon 14,  $(\Delta^{14}\text{C})_c$ , with normalization by  $^{12}\text{C}$  (5) removed (Green). The long-term decline of perturbation  $^{14}\text{C}$  apparent during the initial 20 years continues to the end of the concatenated record. The record of  $(\Delta^{14}\text{C})_c$ , however, approaches an equilibrium level. It reflects carbon 14 in the early 1960s, somewhat higher than the equilibrium level inferred a decade earlier, when  $^{14}\text{C}$  measurements began. Also superimposed in Figure 3 is pure exponential decline with an effective absorption time  $\tau_{\text{eff}} = 10$  yrs (Magenta). Having time scale equal to that of effective absorption in Figures 1 and 2, it tracks the observed record of  $(\Delta^{14}\text{C})_c$ .

The equilibrium level of  $^{14}\text{C}$  is determined by total production in (9). Most of this production is natural. Despite the *Limited Test Ban Treaty*, however, some nuclear testing continued after 1963. Involved were more than a thousand detonations (see Arms Control Association, 2020 [35]), the last ones in 2017 by North Korea. Albeit chiefly under ground and in the ocean, some of the  $^{14}\text{C}$  produced in those tests emerged at the Earth's surface, where it was emitted into the atmosphere along with other  $^{14}\text{C}$ . Contemporaneous is the release of  $^{14}\text{C}$  from nuclear power generation, which increased sharply after 1963 (Runte, 2013 [36]). Both extraneous sources of  $^{14}\text{C}$  act to elevate its equilibrium level.

More central to the equilibrium level, however, is natural production of  $^{14}\text{C}$  in the stratosphere. Embodied in  $e'_{N,14}$  (9), natural production occurs through absorption of free neutrons that are librated by cosmic radiation. The latter is modulated by solar wind. The departure from constant natural emission,  $e'_{N,14}$ , must evolve likewise (see also: Damon & Peristykh 2004 [37]; Connolly et al. 2021 [38]).

Neutron flux has been monitored for the last 56 years at the Cosmic Ray Station of the University of Oulu Finland [39]. Plotted in Fig 4, it reveals a systematic increase over the last 30 years - of 5-10%. The observed increase tracks decreasing sun spot number and weakening solar magnetic field.

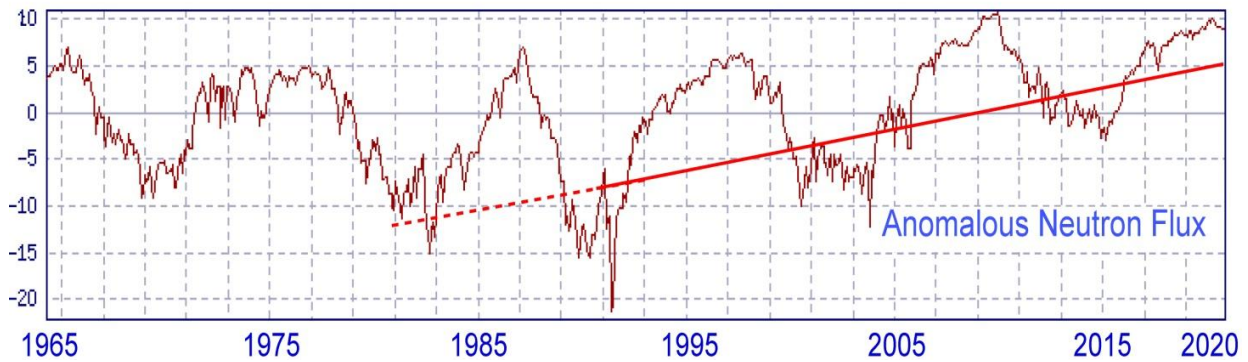


Figure 4: Anomalous neutron flux (%) observed at Oulu Finland.

Inclusive of time-varying natural production (but exclusive of oscillatory reinforcement), the Conservation Law becomes:

$$\frac{dC'_{14}}{dt} = e'_{NB,14}(t) - \frac{C'_{14}}{\tau_{\text{eff}}}, \quad (10)$$

with  $e'_{NB,14}$  accounting for time-varying natural emission as well as nuclear power generation and nuclear testing after the test ban treaty.

Equation (10) has been integrated for an effective absorption time of  $\tau_{\text{eff}} = 10$  yrs (with a re-emission fraction of  $\beta = 0.6$ , it corresponds to a direct absorption time of 4 yrs) and with total emission of  $e'_{NB,14} = 123$  %o/yr that now increases since 1990 at  $0.3$  %o/yr<sup>2</sup>, characteristic of observed neutron flux in Figure 4. The resulting anomaly,  $C'_{14}$ , is then transformed back to anomalous carbon

14 that is referenced against the standard activity (5). Plotted in Figure 5 is calculated  $(\Delta^{14}C)_c$  (Magenta). The calculated evolution tracks the observed evolution of  $(\Delta^{14}C)_c$ , which is superimposed (Green). Faithfully represented is its approach to a new equilibrium level. About 250‰ higher than the equilibrium level during the pre-test era, its elevation is well accounted for by the observed enhancement of neutron flux.

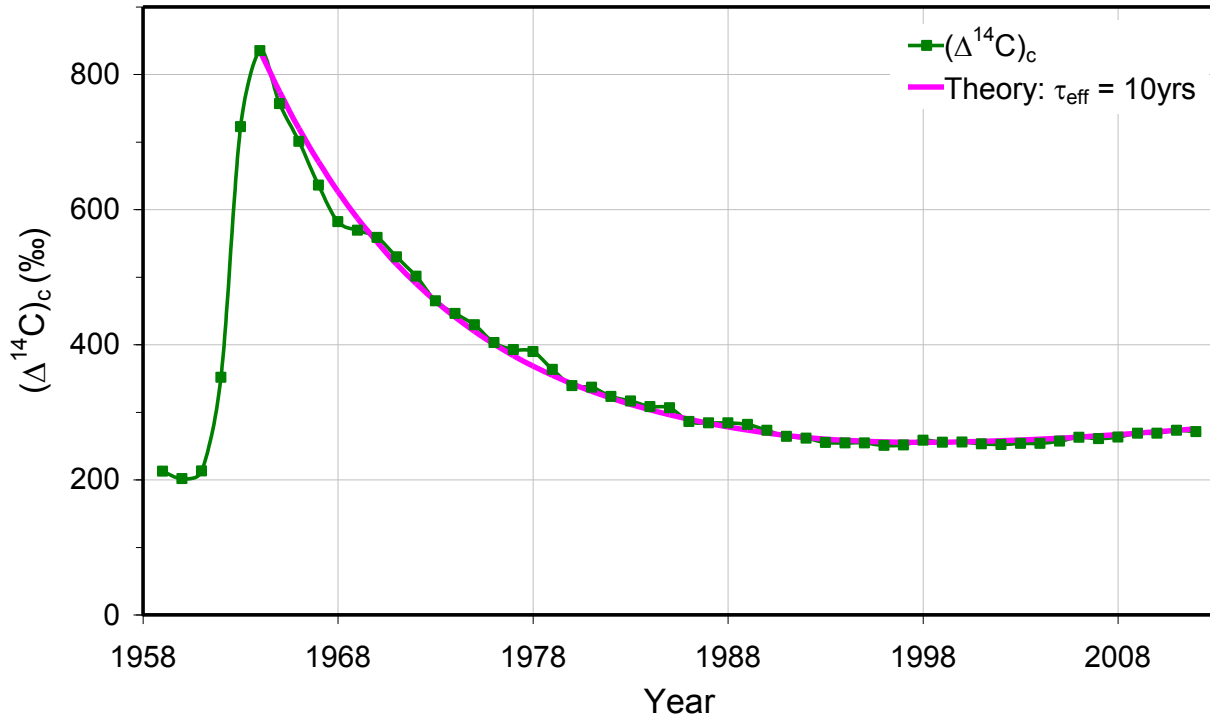


Figure 5: Evolution of calculated  $(\Delta^{14}C)_c$  with effective absorption time of  $\tau_{\text{eff}} = 10$  years (Magenta), compared against observed  $(\Delta^{14}C)_c$  (Green).

Integrations of the Conservation Law that governs atmospheric  $CO_2$  (Figures 3 and 5) illustrate the operation of  $CO_2$  absorption with a single absorption time of only 10 yrs. Thereby, they eviscerate unqualified claims that were advanced by Andrews 2020 [30]):

*Unconventional models motivated by a misinterpretation of the isotope ratio variable “ $\Delta^{14}C$ ” are excluded when the error is corrected... Harde and Berry erroneously concluded that after atmospheric nuclear testing ceased, the “pulse” of extra  $^{14}C$  introduced by the tests exponentially disappeared from the atmosphere with a time constant of approximately 16 years.*

The foregoing integrations demonstrate that these claims are incorrect. It is noteworthy that, had the removal of perturbation  $^{14}C$  and, hence, of  $CO_2$  come to a halt after 1990, as implied by Andrews, continued emission (which is of order 100 ppmv/yr; see Sec 3.1) would have increased  $CO_2$  since then by several hundred ppmv. The observed increase was nothing of the sort, proceeding at about the same rate as during earlier years.

### 3. Perturbation of $CO_2$

Independent of its isotopic tracer  $^{14}CO_2$ ,  $CO_2$  itself reveals its absorption - through its seasonal perturbation. In its monthly deviation from annual-mean conditions,  $CO_2$  increases by  $\sim 6$  ppmv, after which it declines by  $\sim 4$  ppmv. Like perturbation  $^{14}CO_2$ , this seasonal perturbation depends on the absorption time of  $CO_2$ .



### 3.1 Simulation of CO<sub>2</sub>

Inclusive of anthropogenic emission,  $e_A$ , the Conservation Law governing atmospheric CO<sub>2</sub> is

$$\frac{dC_{CO_2}}{dt} = e_N(T, t) + e_A(t) - \frac{C_{CO_2}}{\tau_{eff}}, \quad (11)$$

where natural emission,  $e_N$ , is generally temperature and time dependent. Seasonality can enter CO<sub>2</sub> through emission, or absorption, or through a combination of the two. Its influence on CO<sub>2</sub>, however, depends only on the seasonality of *net* emission: the residual between emission and absorption. Therefore, it is sufficient to introduce seasonality through emission; similar behavior can be obtained through equivalent seasonality in the other preliminary avenues.

Natural emission is allowed to vary with anomalous temperature,  $\Delta T$ , and month:

$$e_N(T, t) = e_{N0} + e_T(\Delta T, t) + \frac{e_{s0}}{2} \cdot \{1 + \cos(\omega(t - t_0) + \varphi_e + m \cdot \sin \omega(t - t_0))\}, \quad (12)$$

where  $e_{N0}$  is the undisturbed emission rate,  $e_{s0}$  is the amplitude of its seasonal modulation,  $\varphi_e$  is its constant background phase, and  $m \cdot \sin \omega(t - t_0)$  is a modulation of its phase that recovers the asymmetric shape of the observed seasonality. Note: Because the seasonal modulation in (12) is nonnegative, it has non-vanishing annual mean.<sup>A</sup>

Background emission, unperturbed by temperature and anthropogenic emission, is prescribed as  $e_{N0} = 3$  ppmv/yr, with remaining mean emission represented in the nonnegative seasonal emission. For anomalous temperature,  $\Delta T(t)$ , we rely on the record of annual-mean tropical temperature observed at Hawaii (NOAA [40]), which underwent systematic warming (trend) during the Mauna Loa era of 0.13°C/decade. Distinguished from other latitudes, the same warming was observed across the tropics by the Microwave Sounding Unit suite of satellite instruments (Spencer et al. 2017 [41]). It is also evident in the record of Tropical Sea Surface Temperature (Kennedy et al. 2019 [42]). The dependence of emission on temperature is defined to be slightly nonlinear (see, e.g., Harde 2019 [3]):

$$e_T(\Delta T, t) = \beta_e \cdot \Delta T(t)^{1.3}, \quad (13)$$

where  $\beta_e = 10$  ppmv/yr/°C<sup>1.3</sup> is the coefficient of temperature dependence. During the Mauna Loa era, this temperature dependence increases emission by 6.9 ppmv/yr, with an average of  $\langle e_T \rangle = 3$  ppmv/yr.  $\tau_{eff}$  can also be temperature dependent. But, to deduce an upper limit on absorption time, it is sufficient to consider changes in  $e_N$ .

Seasonal modulation of emission is assigned an amplitude  $e_{s0} = 40$  ppmv/yr, a background phase  $\varphi_e = \pi$ , and a modulation amplitude of  $m = 0.8$ . The asymmetric form of seasonal modulation intensifies emission during 8 months (September - April) but weakens it over only 4 months (May - August). Together with its non-vanishing mean, the asymmetry leads to annual-mean emission of  $\langle e_s(t) \rangle = 27.4$  ppmv/yr.

Anthropogenic emission,  $e_A(t)$ , is prescribed from the time-varying record Fossil Fuel Emissions, which are well documented (CDIAC [32]). Although poorly documented, we also include the record of Land Use Change (e.g., Le Quéré et al. [43]; CICERO [44]). Together, those contributions reach 5.5 ppmv/yr in 2018, about 5% of total emission.

The effective absorption time,  $\tau_{eff}$ , is defined analogous to (7). Therefore, the absorption rate  $\mathcal{A} = C_{CO_2}/\tau_{eff}$  in (11) includes re-emission,  $e_R = \beta \cdot C_{CO_2}/\tau$ ; see (6) – (7). It is noteworthy that  $\tau_{eff}$ , here is determined independently of its determination in Sec. 2: by requiring the observed evolution, which is recovered from (11), to satisfy the Conservation Law.

<sup>A</sup> Retaining the annual mean of seasonal modulation in  $e_s(t)$  is arbitrary. It could equivalently be isolated in the constant background emission,  $e_{N0}$ .

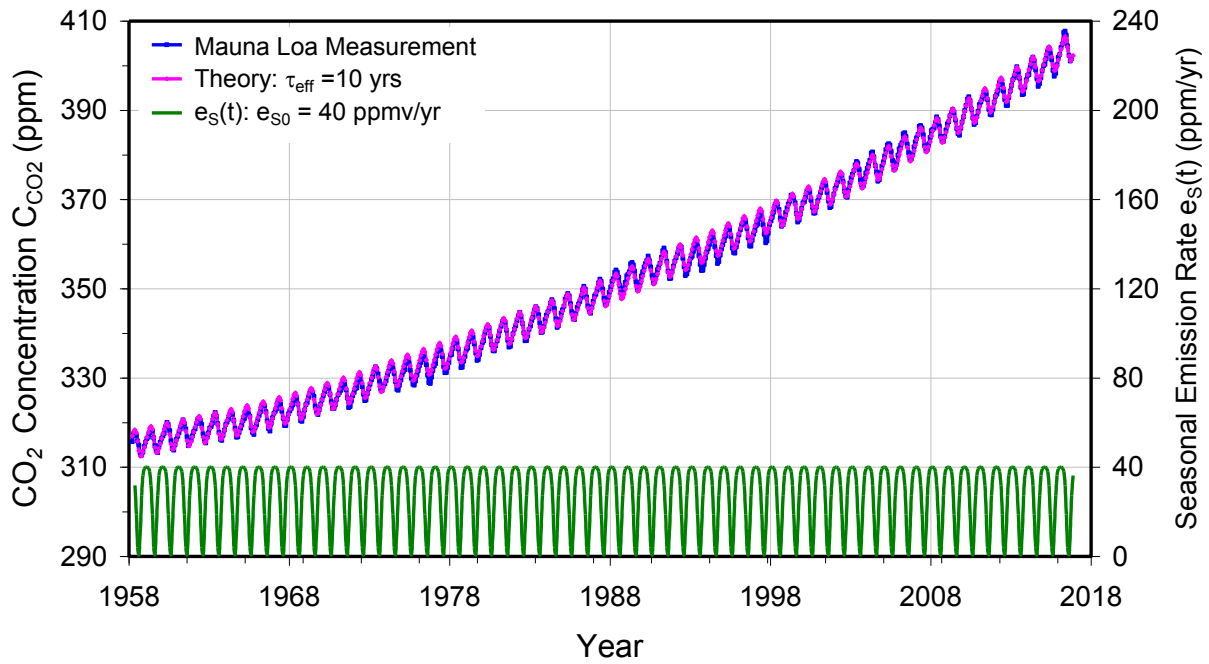


Figure 6: Calculated evolution of  $CO_2$  (Magenta), inclusive of temperature-dependent natural emission, its seasonal modulation, and anthropogenic emission, all for an effective absorption time  $\tau_{eff} = 10$  yrs (see text). Superimposed is the observed evolution of  $CO_2$  (Blue). Also shown is the seasonal modulation of emission (Green).

With an effective absorption time of  $\tau_{eff} = 10$  yrs, numerical integration of (11) yields the evolution of  $CO_2$  in Figure 6 (Magenta). It tracks the observed evolution of  $CO_2$  which is superimposed (Blue). Also shown in Figure 6 is the seasonal modulation of emission  $e_s(t)$  (Green).

Noteworthy is the fraction of total emission responsible for the observed evolution that follows from natural emission. Implicit in the absorption term in (11) is re-emission of  $CO_2$ :

$$-\frac{C_{CO_2}}{\tau_{eff}} = -(1 - \beta) \frac{C_{CO_2}}{\tau}, \quad (14)$$

where  $\beta$  is the fractional re-emission of  $CO_2$  that was removed through direct absorption, which operates with the time scale  $\tau$ . The term  $+\beta \cdot C_{CO_2} / \tau$  in (14) represents re-emission of absorbed  $CO_2$  (6), most of which was emitted by natural sources. With  $\beta = 0.6$  ( $\tau = 4.0$  yrs) and  $\langle C_{CO_2} \rangle \approx 395$  ppm over the last decade, mean re-emission is then  $\langle e_R \rangle = \beta \cdot \langle C_{CO_2} \rangle / \tau = 59.3$  ppmv/yr. Collective emission from natural sources is thus:  $\langle e_{NO} \rangle + \langle e_S \rangle + \langle e_T \rangle + \langle e_R \rangle \cong 3 + 27.4 + 3 + 59.3 \cong 92.7$  ppmv/yr. It corresponds to natural emission of 93 ppmv/yr that was estimated by the IPCC who, in contrast to temperature dependence accounted for in (13), presumed that natural emission remained constant.

Figure 7 presents the same record as Figure 6, but on a magnified time scale. The asymmetric form of seasonality is well captured by the prescribed phase modulation (12). Accordingly, the calculated monthly variation (Magenta) tracks the observed variation of  $CO_2$  (Blue). Seasonal modulation, (Green), intensifies emission during intervals of  $CO_2$  growth, leaving it weaker during intervals of  $CO_2$  decline.

The calculated seasonal variation of  $CO_2$  is determined largely by the seasonality of emission. This dependence follows from absorption at  $\tau_{eff} = 10$  yrs being slow compared to seasonal transience in emission, which therefore alters anomalous  $CO_2$  before it can be influenced substantially by absorption.

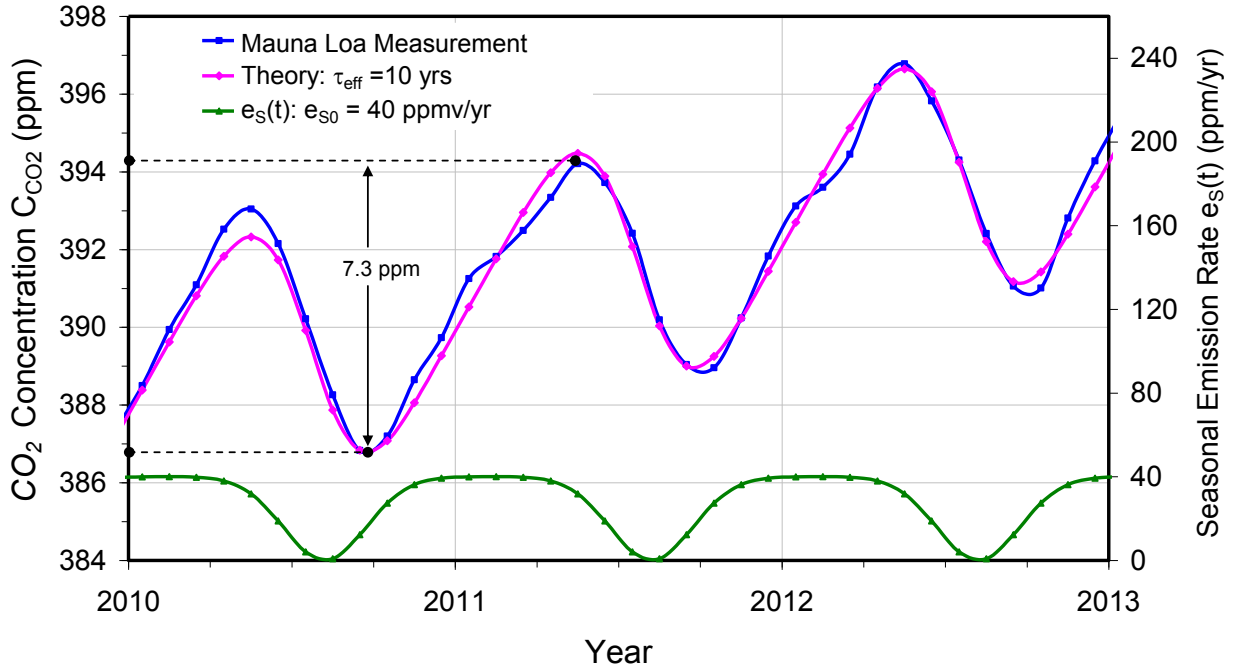


Figure 7: As in Figure 6, but on an expanded time scale.

The observed evolution (Figures 6 and 7) can, within bounds, be recovered for other values of emission. This is a consequence of the local mean of  $CO_2$  being determined by the product of total emission and the absorption time (see Eq. (11)). A change in one can therefore be compensated by a change in the other. However, the observed evolution of  $CO_2$  is recovered only for  $\tau_{eff}$  shorter than 11 yrs - e.g., in previous simulations that reproduce observed features (Harde 2017 [2], 2019 [3]; Berry 2019 [9], 2021 [45]). Regardless of temperature dependence and seasonality in natural emission (12), slower absorption ( $\tau_{eff} > 11$  yrs) does not recover the long-term increase and the seasonality of observed  $CO_2$ .

This upper bound on  $\tau_{eff}$  applies equally to pre-industrial conditions, for example, with  $CO_2$  concentration of 280 ppmv and without anthropogenic or thermally-induced emission.<sup>A</sup> With seasonal perturbation as exists today, mean emission  $\langle e_s(t) \rangle = 27.4$  ppmv/yr, and under quasi-equilibrium conditions, (11) then gives an effective absorption time of

$$\tau_{eff} = \frac{C_{CO_2}}{\langle e_s(t) \rangle} = 10.2 \text{ yrs}. \quad (15)$$

Integrations with seasonality present in both emission and absorption (not shown) reveal much the same upper bound:  $\tau_{eff} < 12$  yrs. Following from the Conservation Law and the observed perturbation of  $CO_2$ , both upper bounds are consistent with the effective absorption time that was determined independently from the observed perturbation of  $^{14}CO_2$  (Sec. 2).

All of the permissible absorption times represent removal of atmospheric  $CO_2$  that is an order of magnitude faster than is assumed by the IPCC. Absorption times in (11) longer than 100 yrs, which are relied upon by the IPCC to interpret observed changes, lead to  $CO_2$  evolution that diverges conspicuously from its observed evolution.

<sup>A</sup> The often-cited value of 280 ppmv, which is claimed to have been invariant before the industrial era, is merely a representative constant. That value follows, not by actual measurements of atmospheric  $CO_2$ , but from ice core analyses, which are shrouded in uncertainty (see, e.g., Jaworowski et. al, 1992) [46].

### 3.2 Anthropogenic Perturbation

The effective absorption time provides an upper bound on the anthropogenic perturbation of  $CO_2$ . In the presence of constant or increasing anthropogenic emission, the anthropogenic perturbation of  $CO_2$  can never exceed its equilibrium level: that level at which  $CO_2$  is removed by absorption,  $C_{CO_2}^A / \tau_{eff}$ , as fast as it is introduced by emission,  $e_A$ . The equilibrium level, thus, places a hard cap on accumulation of  $CO_2$  in the atmosphere.

Contrasting fundamentally is the position adopted by the IPCC: that, even for constant emission, anthropogenic  $CO_2$  would continue to accumulate in the atmosphere. The forgoing considerations, which rest on immutable physical constraints, show that actual  $CO_2$  would do nothing of the sort.

Upon reaching its equilibrium level, the growth rate of anomalous  $CO_2$  vanishes. The conservation law governing the anthropogenic perturbation (e.g., Eq. (11) with emission restricted to  $e_A$ ) then reduces to

$$e_A = C_{CO_2}^A / \tau_{eff}. \quad (16)$$

Rearrangement gives the equilibrium concentration of anthropogenic  $CO_2$ :

$$C_{CO_2}^{A,Eq} = \tau_{eff} \cdot e_A. \quad (17)$$

With  $\tau_{eff} = 10$  yrs and anthropogenic emission equal to its mean over the Mauna Loa Era,  $\langle e_A \rangle \cong 3.4$  ppmv/yr (e.g., CDIAC 2017) [32], (17) yields an upper bound on the net perturbation of  $CO_2$ :

$$\langle C_{CO_2}^A \rangle \leq 34 \text{ppmv}. \quad (18)$$

The anthropogenic perturbation (which the IPCC claims was entirely responsible for increasing  $CO_2$  during the Mauna Loa era) could actually have contributed no more than about a third of the observed increase ( $\sim 100$  ppmv). Much the same follows directly from the Conservation Law, without explicit reference to the equilibrium level (Appendix).<sup>A</sup>

Even if, throughout the last half century, anthropogenic emission had been as large as its recent maximum, 5.0 ppmv/yr, the anthropogenic perturbation of  $CO_2$  could have contributed no more than half of the observed increase.

Whereas the perturbation introduced by anthropogenic emission cannot exceed this upper bound, it can be smaller. If anthropogenic  $CO_2$  experiences absorption faster than the limiting absorption that was determined in Sec 2 and independently in Sec 3.1 (i.e., if  $\tau_{eff}$  is shorter than  $\sim 10$  yrs), the anthropogenic perturbation will be reduced accordingly. For example, at  $\tau_{eff} = 4$  yrs, anthropogenic emission, even as great as its recent maximum, could perturb  $CO_2$  by no more than 20 ppmv. This perturbation of  $CO_2$  represents less than 20% of its observed increase to date, mirroring the fractional increase of total emission (Appendix). Continued emission at this rate, even indefinitely, would not increase the anthropogenic fraction further. On the contrary, continued growth of observed  $CO_2$ , which has existed since at least the onset of uninterrupted monitoring, would render the anthropogenic fraction of increased  $CO_2$  even smaller.

### 4. Conclusions

<sup>14</sup> $CO_2$  is an isotopic tracer of all  $CO_2$ . The exponential decline of its nuclear perturbation establishes that absorption of  $CO_2$  is determined, not by extraneous reservoirs of carbon, but autonomously by the atmosphere. Specifically, the rate at which  $CO_2$  is removed from the atmosphere is directly proportional to the instantaneous abundance of  $CO_2$  in the atmosphere. Removal of  $CO_2$  operates

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<sup>A</sup> If considered over the entire industrial era, the anthropogenic fraction of increased  $CO_2$  must be even smaller. With emission equal to its mean since 1850, 1.8 ppmv/yr, (17) gives an upper bound on the anthropogenic perturbation of only 18 ppmv. The anthropogenic fraction of increased  $CO_2$ , allegedly from 280 ppmv in 1850, would then have been smaller than 14%.

with a single time scale, which reflects the collective absorption by all sinks of  $CO_2$  at the Earth's surface.

The long-term decline of anomalous  $^{14}CO_2$  reveals an effective absorption time of about 10 years. It represents removal of atmospheric  $CO_2$  that is much faster than has been presumed to interpret observed changes. The absorption time establishes an upper bound on perturbations of  $CO_2$ . Included is anomalous  $CO_2$  introduced by anthropogenic emission, a perturbation to natural emission that constitutes less than 5% of total emission.

The equilibrium level of anthropogenic  $CO_2$  represents a hard cap on its accumulation in the atmosphere. Determined by anthropogenic emission and the effective absorption time, it is too small for anthropogenic emission to be responsible for the observed increase of atmospheric  $CO_2$ . At the existing level of anthropogenic emission, or even with foreseeable increases, the anthropogenic fraction of increased  $CO_2$  will remain small.

The title of this paper poses the question: What controls atmospheric  $CO_2$ ? - a question central to understanding its observed evolution. The preceding analysis of its removal follows independently from perturbations of  $CO_2$  and of its isotopic tracer,  $^{14}CO_2$ . Through a diagnosis of exclusion, it provides a clear and unambiguous answer: The controlling influence is not anthropogenic emission.

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## Appendix Anthropogenic Contribution to Increased $CO_2$

During its long-term increase, atmospheric  $CO_2$  remains in a state of quasi equilibrium, wherein emission of  $CO_2$  is approximately balanced by its absorption. The Conservation Law for total  $CO_2$  (11) then reduces to

$$e \cong C_{CO_2} / \tau_{eff}. \quad (A1.1)$$

Differencing (A1.1) between initial and final states obtains the Conservation Law for the change of  $CO_2$ :

$$\Delta e \cong \Delta C_{CO_2} / \tau_{eff}. \quad (A1.2)$$

As anthropogenic emission also changes gradually, anthropogenic  $CO_2$  likewise remains in a state of quasi equilibrium. The Conservation Law governing the anthropogenic component of  $CO_2$ , which follows from (11) with  $e$  restricted to  $e_A$ , therefore assumes the same form as (A1.1):

$$e_A \cong C_{CO_2}^A / \tau_{eff}. \quad (A2)$$

Dividing (A2) by (A1.2) obtains

$$\frac{C_{CO_2}^A}{\Delta C_{CO_2}} \cong \frac{e_A}{\Delta e}. \quad (A3)$$

Anthropogenic emission introduces a fractional increase of  $CO_2$  that is equal to its fractional increase of emission.

Under quasi equilibrium, the change of emission must be approximately equal to the change of absorption. Incorporating the upper bound on absorption time,  $\tau_{eff} \leq \tau_{eff}^{\max}$ , then gives

$$\frac{C_{CO_2}^A}{\Delta C_{CO_2}} \cong \frac{e_A \cdot \tau_{eff}^{\max}}{\Delta C_{CO_2}}. \quad (A4)$$

For mean anthropogenic emission of 3.4 ppmv/yr, an upper bound on absorption time of  $\tau_{eff}^{max} = 10$  yrs, and a  $CO_2$  increase over the Mauna Loa era of  $\sim 100$  ppmv, (A4) yields the following upper bound on the anthropogenic contribution to increased  $CO_2$ :

$$\frac{C_{CO_2}^A}{\Delta C_{CO_2}} \leq 34\% . \quad (A5)$$

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