

Reply to

Comment on "Scrutinizing the carbon cycle and CO₂ residence time in the atmosphere"

by P. Köhler, J. Hauck, C. Völker, D. Wolf-Gladrow, M. Butzin, J. B. Halpern, K. Rice, R. Zeebe

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Abstract

In their Comment on our publication (Harde, 2017) the authors argue that the description of the presented carbon cycle would be too simple, is based on invalid assumptions, and does not address key processes in this cycle that are important on the time scale of relevance. Therefore, they assert, the treatment would lead to an incorrect conclusion on the involvement of anthropogenic emission. In this Reply, we show that these arguments are fallacious, that our treatment is essential to the underlying physics, and that the resulting conclusions conform to actual observations of atmospheric CO₂. These features are contrasted with treatments relied upon by these authors, which are shown to be unphysical: They do not obey the physical laws that govern atmospheric CO₂. It is those key failures which are responsible for the erroneous conclusion that the increase in atmospheric CO₂ over the past 260 years is principally anthropogenic.

Key-words: Carbon cycle, atmospheric CO₂ concentration, CO₂ residence time, anthropogenic emissions, fossil fuel combustion, land use change, climate change

Preliminary Note

In contrast to complex but largely *ad hoc* carbon cycle models used to describe atmospheric CO₂, we considered a fundamental physical constraint that must be obeyed. Atmospheric CO₂ is governed by the balance equation, the conservation law wherein the uptake of CO₂ is proportional to the instantaneous concentration of CO₂. Accounted for in this law was the temperature dependence of natural emission and absorption, dependence that is inherent to physical and chemical processes involved in exchanging CO₂ with the atmosphere. These fundamental considerations alone were shown to be sufficient to account for nearly all of the 20th century change of CO₂ recorded in actual atmospheric observations. They are likewise sufficient to account for nearly all of the paleoclimatic change of CO₂ during the last 150,000 years, inferred from pseudo observations. With the IPCC's own estimates of natural absorption, these fundamental considerations require the following: (1) The average residence time of CO₂ in the atmosphere is only 4 years. Equivalent to the relaxation time following perturbation of CO₂, it is far shorter than the time scale relied upon by the IPCC. (2) The contribution from anthropogenic emission to overall CO₂ is 4.3%. (3) Its contribution to the CO₂ increase over the Industrial Era is only 15%. With other evaluations of absorption, the contribution from anthropogenic emission is even smaller.

In their Comment, Köhler et al. argue that the foregoing description is too simple, is based on invalid

assumptions, and does not address key processes in the global carbon cycle that are important on the time scale of relevance. Therefore, they argue, the treatment leads to an incorrect conclusion on the involvement of anthropogenic emission. Below, we show that these arguments are fallacious, that our treatment is physically sound, indeed essential to the underlying physics, and that the resulting conclusions conform to actual observations of atmospheric CO₂. These features are contrasted with treatments relied upon by Köhler et al., which are shown to be unphysical: They do not obey the physical laws that govern atmospheric CO₂. It is those key failures which are responsible for the IPCC's erroneous conclusion that the increase in atmospheric CO₂ over the past 260 years is principally anthropogenic.

Köhler et al.'s Comment is devoid of concrete analysis. Its tenor is to inundate the reader with citations, a reiteration of the IPCC catalogue.¹ Invoked to support sweeping claims, too many of the citations are of dubious significance to be addressed here. Rather than responding in kind, we focus on key claims of Köhler et al. and the purported evidence upon which they rely. Supporting references that likewise contradict those claims are collected in Appendix A.

1. Claim: Residence Time Differs from Adjustment Time

Köhler et al.'s description of atmospheric changes in response to CO₂ perturbation is misguided. Reiterating IPCC reports, they contend:

"The main reason for the increase [in atmospheric CO₂] is the addition of anthropogenic CO₂."

"The rise in atmospheric CO₂ from a preindustrial value of 278 ppm before 1750 to 390 ppm in 2010 is solely due to anthropogenic emissions."

In making this argument, they introduce the so-called "*adjustment time*", the time for atmospheric CO₂ to re-equilibrate following a perturbation. The adjustment time, they argue, differs from the "*residence time*", the characteristic time that a fixed collection of CO₂ remains in the atmosphere before being absorbed at the Earth's surface. To support this argument, Köhler et al. invoke changes in a wide range of extraneous systems which, because they involve carbon, serve as carbon reservoirs: "*the surface ocean, the intermediate ocean, the deep ocean, marine sediments, and the terrestrial biosphere*". In reality, these reflect countless sinks of atmospheric CO₂, which have an unsteady and complex distribution.²

Different sinks operate with a different time scales. They are represented in the so-called Bern Model of CO₂ absorption (e.g., Joos et al., 1996; Hansen et al., 2007; 2013). A prototype of similar treatments, the Bern Model is actually a model of behavior in other models. Relative to observed absorption, it is largely hypothetical. As Köhler et al. note, the resulting adjustment of atmospheric CO₂ is then "*approximated by a sum of a few exponential [decay] functions with different characteristic time scales*", which reflect absorption of perturbation CO₂ by the different sinks.

In this treatment, the overall adjustment time is dictated by decay on the *longest* time scales - the slowest sinks of atmospheric CO₂. They prevail after faster decay on shorter time scales has collapsed. Gradual decay of perturbation CO₂ then persists over long duration. The slow removal of CO₂ in this treatment enables anthropogenic CO₂ to accumulate in the atmosphere. As a result, more than 30% of an atmospheric perturbation remains after a century, as is seen in Fig. 1 (Red). Even after a thousand

¹ It amounts to validation by consensus, a failure of logic that has been quashed repeatedly in the history of science (see, e.g., Hawking, 1988).

² The difficulty in observing global absorption of CO₂ is underscored by the transient nature of land surfaces, which can operate as a sink of CO₂ under some conditions yet as a source of CO₂ under other conditions (Billings et al., 1982; Lugo and Brown, 1993).

years, almost 20% of the perturbation still remains.

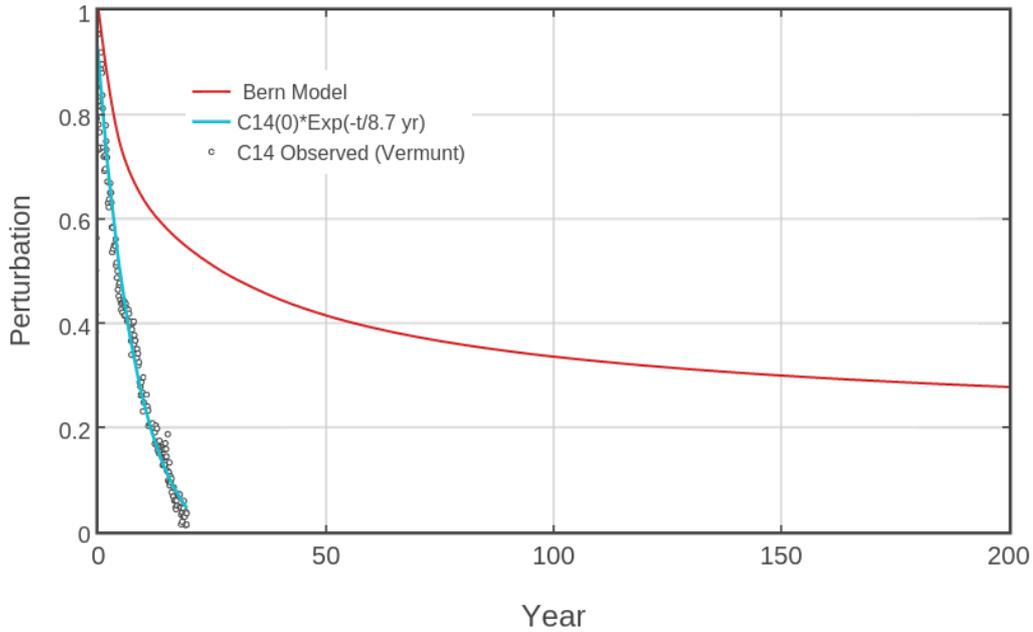


Fig. 1: Decay of perturbation CO₂, predicted by the Bern Model of absorption (Red) compared against that observed in ¹⁴C after elimination of the nuclear source (circles). Superimposed is exponential decay at a constant rate $\alpha = (8.7 \text{ yrs})^{-1}$ (Blue).

The above treatment is the cornerstone of IPCC reports. It forms the basis for emission scenarios and their projected impact on climate properties like global temperature. This treatment, however, suffers from a minor defect. It does not obey the conservation law that governs and therefore controls atmospheric CO₂.

The balance equation for atmospheric CO₂ is

$$\frac{dC}{dt} = e_T - a, \tag{1}$$

where, because CO₂ is conserved in the atmosphere, its globally averaged concentration C (proportional to the atmospheric mass of CO₂) can change only through its introduction and removal at the Earth's surface: through the upward surface flux of CO₂, equal to globally averaged *total* emission e_T , and the downward surface flux of CO₂, equal to globally averaged absorption a (where, as in Harde (2017), lower case letters denote fluxes with respect to concentration). Note: Exchange between atmospheric CO₂ and extraneous reservoirs, claimed erroneously by Köhler et al. to be absent, is entirely accounted for in the imbalance of surface fluxes e_T and a , which represents the net CO₂ flux in/out of the atmosphere.

In (1), absorption of CO₂ is proportional to the instantaneous concentration of CO₂. The balance equation then becomes

$$\frac{dC}{dt} = e_T - \alpha \cdot C, \tag{2}$$

with absorption rate α equal to the inverse absorption time τ^{-1} . Common to many physical processes, this feature of CO₂ absorption is an empirical fact: It is observed in the decay of carbon 14. ¹⁴C is a tracer of atmospheric CO₂. It was elevated by nuclear bomb testing during the 1950s and 1960s. As seen in Fig. 1 (circles), the decay of ¹⁴C after nuclear testing ended is almost perfectly exponential. The observed decay thus satisfies

$$\frac{dC_{14}}{dt} = -\alpha \cdot C_{14}. \quad (3)$$

It corresponds to absorption that is proportional to instantaneous concentration, with a constant absorption time of only a decade.³

Now, global absorption a is the collective effect of countless sinks that are distributed irregularly across the Earth's surface. Contributions to absorption from those sinks follow as

$$\begin{aligned} a &= \alpha_1 C + \alpha_2 C + \dots + \alpha_N C \\ &= (\alpha_1 + \alpha_2 + \dots + \alpha_N) \cdot C = \alpha \cdot C \end{aligned} \quad (4)$$

Collective absorption thus leads to exponential decay of perturbation CO₂ at a *single* rate

$$\alpha = \alpha_1 + \alpha_2 + \dots + \alpha_N. \quad (5)$$

Note: This decay rate is faster than the rate of any individual sink. Further, the constant decay rate, equal to the rate α of CO₂ absorption, prevails as long its concentration C remains nonzero, i.e., indefinitely.

The above behavior is a consequence of the balance equation. It contrasts sharply with the Bern Model of CO₂ absorption, relied upon by Köhler et al. There, decay proceeds at *multiple* rates. The corresponding sinks operate, not collectively, but independently. After a couple of their decay times, the fastest sinks become dormant. Overall decay then continues only via the slowest sinks, which remove CO₂ gradually. It is for this reason that the treatment relied upon by Köhler et al. leaves atmospheric CO₂ perturbed for longer than a thousand years (Fig. 1). In contrast, the behavior required by the balance equation decays as fast or faster than that of the fastest sink (5). The observed decay of ¹⁴C shows that the corresponding absorption operates on a time scale of only a decade or shorter.

Notice: The treatment of CO₂ relied upon by Köhler obeys the following:

$$\begin{aligned} C &= C_{10} e^{-\alpha_1 t} + C_{20} e^{-\alpha_2 t} + \dots + C_{N0} e^{-\alpha_N t} \\ &= C_1 + C_2 + \dots + C_N \end{aligned} \quad (6)$$

It follows that

$$\begin{aligned} \frac{dC}{dt} &= -\alpha_1 C_{10} e^{-\alpha_1 t} - \alpha_2 C_{20} e^{-\alpha_2 t} \dots - \alpha_N C_{N0} e^{-\alpha_N t} \\ &= -\alpha_1 C_1 - \alpha_2 C_2 \dots - \alpha_N C_N \\ &\neq -(\alpha_1 + \alpha_2 + \dots + \alpha_N) \cdot C \end{aligned} \quad (7)$$

The resulting behavior does not obey the balance equation - the conservation law that *is* obeyed by CO₂ in the atmosphere.

The fallacy of this treatment is illustrated by a mechanical analogue. A bath of water is emptied through multiple drains. Each has different cross-sectional area, which is proportional to the rate at which that drain removes water. In the physical world, all drains operate, irrespective of area, until the bath is empty. The time to empty the bath is therefore close to the removal time of the largest (fastest) drain – much shorter than that of the smallest (slowest) drain.⁴ In the world of Köhler et al., the behavior is reversed. The largest (fastest) drains come to a halt after only a couple of their comparatively short removal times. Thereafter, water is removed only by the smallest (slowest) drains. Under those

³ Apparent absorption in the decay of ¹⁴C is actually an upper bound on true absorption. Accounting for other influences on ¹⁴C shows that the absorption time of atmospheric CO₂ is even shorter than a decade (Appendix B).

⁴ Where, as in (2), removal time of an individual drain is the inverse of its removal rate.

circumstances, the time to empty the bath is close to the removal time of the smallest (slowest) drain. It is much longer than the removal time of the largest (fastest) drain and, hence, much longer than occurs under the balance equation (2).

The latter circumstances could operate only if the largest (fastest) drains were plugged, forcing water to queue for the evening matinee – to wait for removal by the smallest (slowest) drains. For the atmosphere, this would require the fastest sinks of CO₂ to become saturated – and, notably, to do so on a time scale of relevance.⁵ The observed behavior of carbon 14 demonstrates that this is not the case (Fig. 1). Its rapid decay following elimination of the perturbing nuclear source makes it clear that present absorption of CO₂ is 1-2 orders of magnitude faster than that claimed by Köhler et al. Further, the decay of ¹⁴C is almost perfectly exponential, reflecting an absorption time that is constant. Accordingly, the observed record of absorption exhibits no evidence of saturation. Independent analyses reach the same conclusion (Appendix A).

Köhler et al. argue that the adjustment time of atmospheric CO₂, the time for it to re-equilibrate following perturbation, is substantially longer than the residence time of atmospheric CO₂, *citing* “*adjustment times of about 70 years*”. But, according to them, even those are too short. Köhler et al. argue that one-box models “*ignore many relevant processes and consequently under-estimate this [adjustment] time scale (Cawley, 2011)*”. Thus, they claim, the adjustment time is “*well over a hundred years*”.

These claims are contradicted by the conservation law (1) governing atmospheric CO₂. Because CO₂ is conserved in the atmosphere, it can change only through an imbalance of the surface fluxes e_T and a . For this reason, its adjustment to equilibrium must proceed through those influences. They are the same influences that determine the removal time of CO₂ in the atmosphere. If CO₂ is perturbed impulsively (e.g., through a transient spike in emission), its subsequent decay must track the removal of perturbation CO₂, C' , which in turn is proportional to its instantaneous concentration. Determined by the resulting imbalance between e_T and a , that decay is governed by the perturbation form of the balance equation:

$$\frac{dC'}{dt} = -\alpha \cdot C' . \quad (8)$$

This is the same form as observed decay of ¹⁴C following elimination of the perturbing nuclear source (3). From (8), the decay time is seen to be

$$\tau = \frac{1}{\alpha} = -\frac{C'}{dC'/dt} . \quad (9)$$

It is identical to the residence time because α describes the rate at which CO₂ is removed by absorption. This time scale is 1-2 orders of magnitude shorter than the adjustment time claimed by Köhler et al.

Like the Bern Model earlier, treatments that artificially prolong the adjustment time do not obey physical laws that are obeyed by CO₂ in the atmosphere. To distinguish adjustment time from residence time, Cawley (2011) introduces a downward surface flux (equivalent to the absorption rate a) that is a linear function of CO₂ concentration. In his notation:

$$F_e = k_e C + F_e^0 , \quad (10)$$

where F_e and k_e correspond to a and α in (2), and F_e^0 represents a constant downward flux of CO₂, in addition to the downward flux that is proportional to its concentration. From this treatment of absorption, Cawley obtains an adjustment rate (inverse adjustment time τ_{Adj}^{-1}) of

$$k_e = \tau_{Adj}^{-1} = (F_i^0 - F_e^0) / C_{eq} . \quad (11)$$

⁵ The time scale of “relevance” is a couple of decades – the length of observed decay when perturbation CO₂ disappears (Fig. 1). After a century, the issue becomes moot - because fossil fuel reserves will be exhausted.

In (11), F_i^0 represents a constant upward surface flux, which corresponds to the natural component of e_T in (2), and C_{eq} is an equilibrium concentration, which is presumed to have existed before the introduction of anthropogenic emission or an injection of a stronger CO_2 impulse. The consequence of (11) is the emergence of a new time scale, τ_{Adj} , which is distinct from the residence time, τ .

The treatment of absorption (10) is specious. Notice: Absorption of CO_2 is nonzero even if CO_2 concentration vanishes. CO_2 is therefore removed from the atmosphere even if there is no CO_2 in the atmosphere. What world such treatment describes is unclear. What is clear is that it is not the physical world.

This error is fatal. Changes of CO_2 relying on it cannot satisfy the conservation law which *is* satisfied by CO_2 in the atmosphere (Fig. 1). In the physical world, F_e^0 is zero: If atmospheric CO_2 vanishes, so does its absorption. Once this inconsistency is eliminated, the adjustment time (11) reduces to

$$\tau_{Adj} = \tau = C_{eq} / F_i^0 \sim C / e_T. \tag{12}$$

With (2), this is identical to the residence time. The perturbation of CO_2 from anthropogenic emission then reduces to that in our analysis.

The practical consequence of these errors is pivotal. They artificially prolong the time for atmospheric CO_2 to re-equilibrate following impulsive anthropogenic emission. In the presence of steady anthropogenic emission, this feature of the Bern Model enables perturbation CO_2 to grow through accumulation - almost indefinitely. If anthropogenic emission increases as in preceding decades, perturbation CO_2 (which, in 2010, is then already 70% of the observed increase) therefore attains high levels, as seen in Fig. 2 (Red). Under the conservation law (2), the picture differs fundamentally. In the presence of steady anthropogenic emission, perturbation CO_2 reaches equilibrium in a matter of only years. If anthropogenic emission increases as in preceding decades, perturbation CO_2 (which, in 2010, then represents only 15% of the observed increase) increases far slower (Blue). It therefore attains levels that are only a fraction of those attained under the Bern Model, which, by comparison, are grossly exaggerated.

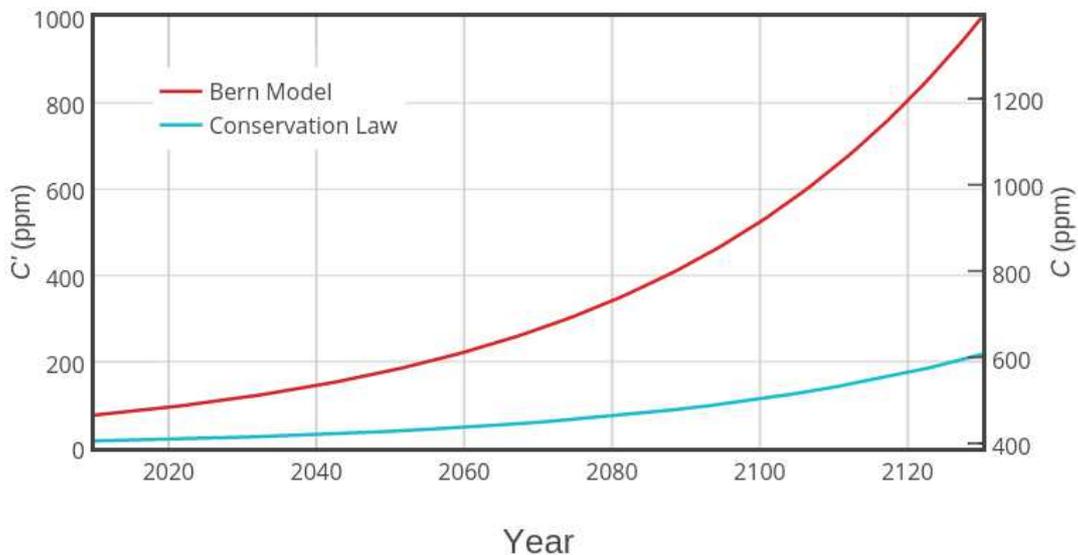


Fig. 2: Growth of CO_2 in the presence of exponentially-increasing anthropogenic emission, extrapolated from the preceding record, under the Bern Model of absorption (Red), compared against that required by the Conservation Law of atmospheric CO_2 (Blue). Left ordinate (C') represents perturbation CO_2 change from 2010, the right (C) total CO_2 .

The carbon cycle is not in steady state, as Köhler et al. recognize. However, the main reason for its transience is not anthropogenic emission, which is a minor contributor to overall emission yet which

Köhler et al. presume is exclusively responsible for changes of CO₂. The main reason for transience of CO₂ is seasonal and decadal changes of radiation, temperature, and other environmental influences. Those changes lead to an imbalance of CO₂ emission and absorption, one that follows through the absorption rate, which in turn is directly related to the residence time.

The role of residence time in controlling changes of CO₂ is illustrated by solving the governing conservation law for the period 1958 - 2017. The balance equation for globally averaged concentration, inclusive of (i) emission and absorption that depend linearly on temperature and (ii) their annual variation, is expressed by (Harde, 2017, Eq. 11):

$$\frac{dC}{dt} = e_A + e_{N0} + \beta_e \cdot \Delta T(t) + a \cdot \sin \omega t - \frac{C}{\tau_0 + \beta_\tau \cdot \Delta T(t)}, \quad (13)$$

where for the anthropogenic emissions e_A the data from the Carbon Dioxide Information Analysis Center (CDIAC, 2017) for fuel emissions were used. The natural emission at pre-industrial times was chosen as $e_{N0} = 80.8$ ppm/yr and the residence time as $\tau_0 = 4$ yr, whereas the temperature coefficient of natural emission is $\beta_e = 12.6$ ppm/yr/°C and that of residence time is $\beta_\tau = 0.47$ yr/°C. The temperature anomaly $\Delta T(t)$ is prescribed from the GISS record of global temperature (GISS, 2017), following a 5 yr moving average. Seasonal variations apparent in the Mauna Loa free-air CO₂ record (NOAA, 2017) are accounted for through the sinusoidal oscillation in natural emission, as appears in ocean observations (Appendix A). Integrating (13) numerically then recovers the evolution of CO₂ at Mauna Loa in Fig. 3 (Green). Its correspondence to the observed evolution of CO₂ (Purple) speaks for itself. Note: This is the same system which shows that, of the apparent CO₂ increase over the industrial era, anthropogenic emission can account for no more than 15%. Contrasting with this integration is one for a removal time of 50 years or longer, as claimed by Köhler et al. It produces CO₂ evolution that bears little resemblance to that observed.

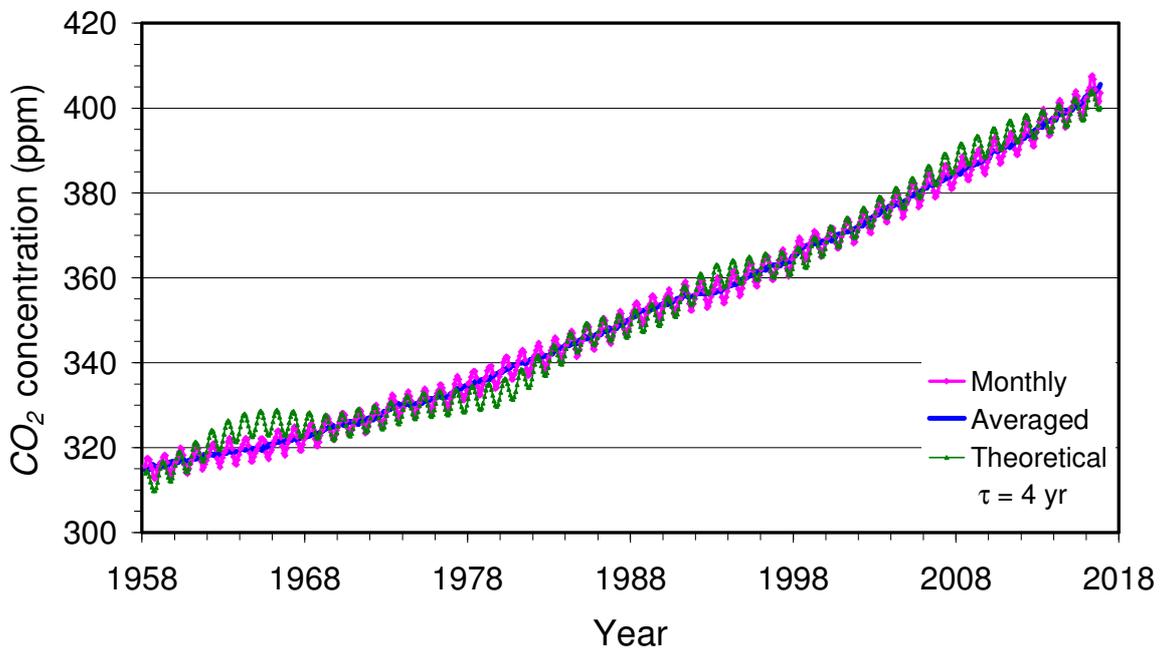


Fig. 3: CO₂ concentration integrated from the balance equation with temperature-dependent emission and absorption and a preindustrial residence time (equal to the adjustment time) of 4 years (Green). Compared against the observed record of CO₂ from Mauna Loa (Purple).

2. Claim: Anthropogenic CO₂ is Not Absorbed by Other Reservoirs

Köhler et al. list the production of anthropogenic carbon from 1750 to 2010 as 518 PgC, corresponding to 1,901 Pg of CO₂. Of this, about 45% is assumed to have accumulated in the atmosphere. This value,

the so-called “Airborne Fraction” (AF), is *ad hoc* - an artifact of presuming that increased CO₂ follows exclusively from anthropogenic emission.⁶ During the same period, cumulative natural emission and absorption were 100 times greater: 727.3 Pg/yr x 260 yr = 189,000 Pg. Cumulative anthropogenic CO₂ was therefore less than 0.5% of total emission into the atmosphere.

If the airborne fraction of anthropogenic CO₂ is arbitrarily assumed, absorption of anthropogenic CO₂ follows directly. The result, however, derives from circular reasoning. It is no more reliable than the assumption upon which it is based. Realistically, fractional absorption smaller than 1% of total absorption can hardly be detected, let alone with global coverage that is necessary for a quantitative description. On the other hand, analyses of recent data reveal no significant decrease in the uptake efficiency of extraneous reservoirs (Appendix A). The same conclusion follows from the nearly-steady exponential decay of ¹⁴C (Fig. 1).

Despite huge uncertainties, climate models are invoked to claim that absorption of anthropogenic CO₂ will quickly become saturated, forcing anthropogenic CO₂ to accumulate in the atmosphere: “*Uptake of anthropogenic carbon will become slower if we continue to increase anthropogenic CO₂ emissions*”. Like others, this claim rests upon models that are largely *ad hoc*. It is therefore speculative. What is clear is that the anthropogenic contribution to overall emission during the Industrial Era is minute, less than 1%. Equally clear is that observed absorption, in the record of ¹⁴C, exhibits no evidence of saturation. Climate models are even invoked to claim in which layers of the ocean carbon will accumulate and, thereby, lead to acidification.

Such claims are little more than hypothetical. Observations necessary to substantiate or falsify them are nonexistent. The models upon which the claims rely are themselves grossly under-constrained. Observations are simply too scarce to configure model parameterizations uniquely. For properties necessary to quantify the global abundance of carbon in reservoirs extraneous to the atmosphere, they are limited to a handful of point measurements. The attendant uncertainty leaves climate models free to be tuned, capable of achieving a wide range of results – including those to match existing observations at any given moment. Climate simulations obtained by retro-fitting models to an updated record of preceding changes then reduce to an elaborate exercise in curve fitting.⁷ It is such ambiguity that enables simulated features to vary widely between models, even in properties as basic as global-mean temperature.⁸

Köhler et al. present an inventory of carbon which is purported to quantify changes in the various surface and sub-surface reservoirs, thereby isolating absorption of anthropogenic CO₂. To claim that extraneous systems, like the carbon content of soil, vegetation (canopy and below, as well as decomposing), the sub-surface ocean, and marine sediments, are known with even close to the precision necessary to quantify those properties globally is preposterous. Understated in this description is the huge uncertainty surrounding those estimates, values which Köhler et al. claim are “*supported by observational-based studies*”. The key word is “*based*”. In truth, those estimates follow from a veneer of observations - point

⁶ AF is premised upon an assumption that natural emission and absorption have remained constant over the industrial era, with natural sinks saturated. This assumption makes anthropogenic emission exclusively responsible for increasing CO₂ - a self-fulfilling prophecy. AF can then be defined from the comparative growth rates of anthropogenic emission and CO₂. In treatments cited by Köhler et al, absorption of anomalous CO₂ is therefore proportional to only the contribution from anthropogenic emission. The conservation law (2), however, requires absorption to be proportional to anomalous CO₂ from *all* sources. The arbitrarily-defined quantity AF is therefore not physically meaningful.

⁷ As was noted by von Neumann, who, during the 1940s and 1950s, helped develop the predecessor of the climate model, “With 4 arbitrary parameters, I can fit an *elephant*. With 5, I can make him wiggle his trunk.” (Dyson, 2004).

⁸ Simulations by several dozen climate models, although following a common drift, are entirely uncorrelated (AR5, Fig 11.25a). On decadal time scales, changes in one model thus have no relationship to changes in the others.

measurements that are scarce to nonexistent. The observational vacuum must be compensated for by models, which are intertwined with the scarce observations. Without global observations necessary to quantify those properties, the purported inventory of changes that could be associated with absorption of anthropogenic CO₂ is fanciful.⁹

The rapid absorption revealed by ¹⁴C (Fig. 1) makes efficient removal of anthropogenic CO₂ clear. Where, specifically, removed CO₂ ends up is of secondary importance. Transferred to the Earth's surface and below, that CO₂ is removed from the atmosphere along with other CO₂ - much faster than is claimed by Köhler et al. (Section 1).

3. Claim: Treatment is Too Simple

Köhler et al. argue that changes of CO₂ in the atmosphere cannot be understood without understanding changes in extraneous systems. In particular, they refer to carbonate chemistry in the ocean, where CO₂ is mostly converted to bicarbonate ions. As only about 1% remains in the form of dissolved CO₂, they argue that only this small fraction could be exchanged with the atmosphere. Due to this so-called Revelle effect, carbonate chemistry would sharply limit oceanic uptake of anthropogenic CO₂.

Köhler et al.'s claim is confused. In regard to understanding changes of CO₂ in the atmosphere, changes in extraneous systems are unnecessary. Note: The governing law of CO₂ in the atmosphere (2) is self contained. With the inclusion of e_T and $a = \alpha \cdot C$, which account for influences on atmospheric CO₂, details of "*the surface ocean, the intermediate ocean, the deep ocean, marine sediments, the terrestrial biosphere*", and countless other extraneous reservoirs of carbon are entirely irrelevant. This feature of the governing physics is not only powerful, but fortunate. Global observations of those surface and sub-surface properties, which are necessary to describe them quantitatively, do not exist.

Concerning carbonate chemistry, it is noteworthy that, in the Earth's distant past, CO₂ is thought to have been almost 2000% as great as its present concentration (e.g., Royer et al., 2004). Most of that was absorbed by the oceans, in which carbon today vastly exceeds that in the atmosphere. According to the IPCC, even in modern times the oceans account for 40% of overall absorption of CO₂ (AR5, 2013, Fig.6.1). In relation to other sinks, their absorption of CO₂ is clearly not limited. Of that 40%, anthropogenic CO₂ represents less than 1%. Contrasting with that minor perturbation in absorption is oceanic emission of CO₂. Through upwelling of carbon-enriched water, the oceans significantly enhance natural emission of CO₂ (Zhang et al., 2017).

It should be noted that the heading of Köhler et al.'s Section 3 is misleading. We did not claim to model carbon in the complete Earth-atmosphere system. That would require a wider analysis, accounting for processes within extraneous systems and exchanges between them. Our analysis focuses upon CO₂ in the atmosphere, which is controlled by the governing conservation law. Köhler et al. characterize this physical law as a flawed 1-box description - because, they claim, a single balance equation does not account for details in other reservoirs, systems that are extraneous to the atmosphere. Köhler et al.'s interpretation is confused. With the inclusion of surface fluxes e_T and a , which account for influences on the atmosphere, the balance equation (1) entirely determines the evolution of CO₂. Details of extraneous systems, which are largely unobservable, are then irrelevant.

Atmospheric CO₂ is fully described by this single equation for a reason. It follows from the 3-dimensional continuity equation, the physical law that governs the global distribution of atmospheric CO₂. In flux form, the continuity equation is given by

⁹ It can be shown, for instance, that even a minor change in the oceanic gradient of dissolved inorganic carbon, one small enough to be unobservable, is sufficient to completely alter the budget of carbon in this inventory.

$$\frac{\partial c}{\partial t} + \nabla \cdot (\mathbf{v}c) = c \nabla \cdot \mathbf{v}, \quad (14)$$

where the local CO₂ concentration, c , is transported with velocity \mathbf{v} . When integrated over the volume of the atmosphere and subjected to the divergence theorem, (14) reduces to the governing balance equation (1) for globally averaged CO₂. If, in the world of Köhler et al., (1) is flawed, then so is the fundamental physical law from which it follows.

Köhler et al. also argue that the analogy to radiocarbon is incorrect - because, they claim, changes in the bulk inventory of CO₂ would be confused with changes in tracers at minute concentration. What is confused is Köhler et al.'s interpretation. Carbon 14 is a tracer of overall carbon, which is dominated by carbon 12. ¹⁴C is therefore a tracer of atmospheric CO₂. Exponential decay of ¹⁴C following elimination of the nuclear source (Fig. 1) is then a direct measure of overall absorption of CO₂ - because, with the elimination of that perturbing source, the conservation law for ¹⁴C reduces to (3). It, in turn, has identical form to the conservation law controlling a perturbation in overall CO₂ (8). The absorption time apparent in ¹⁴C is, in fact, comparable to the residence time of only 4 years.

Köhler et al. argue that the signature of absorption in ¹⁴C is corrupted by dilution via fossil fuel emission, which is mostly free of ¹⁴C (the so-called Suess effect). The claim is specious. Dilution by fossil-fuel emission that is ¹⁴C-free has negligible influence on the decay time of ¹⁴C (Appendix B).¹⁰ Far more influential is re-emission of ¹⁴C from the Earth's surface: ¹⁴C that was recently absorbed from the atmosphere, for example, by vegetation that subsequently decomposes and re-emits that ¹⁴C along with other CO₂. By counteracting absorption, such re-emission significantly prolongs the decay time of ¹⁴C from what would be observed in the presence of absorption alone (Salby, 2016). Consequently, the decade-long decay time of ¹⁴C (Fig. 1) provides an upper bound on the actual absorption time, which can be only shorter. Both are vastly shorter than the purported adjustment time invoked by Köhler et al.

4. Claim: Application of Paleoclimate Record is Incorrect

We showed that linear temperature dependence of CO₂ emission and absorption is a good approximation to their observed interdependence in the modern record (Harde, 2017; see also: Humlum et al., 2013; Salby, 2013, 2016). In fact, a numerical integration of the resulting balance equation, based on the records of global temperature from GISS and anthropogenic emission from CDIAC, recovers nearly all of the observed change of CO₂ during 1958 – 2017 (Fig. 3). Contrary to Köhler et al.'s narrative, those natural factors alone are thus clearly sufficient to account for the preponderance of the observed increase of CO₂ in the record of actual atmospheric observations.

Next, Köhler et al. claim that treatment of the paleoclimate record is a product of erroneous conclusions. Changes of CO₂ in the ice core record, even with the wide uncertainty surrounding those pseudo observations of atmospheric CO₂, exhibit a strong correlation with changes of temperature (e.g., Petit et al., 1999, Fig. 3). The interdependence of ancient CO₂ and temperature is a counterpart of their interdependence in the modern record (Humlum et al., 2013; Salby, 2013). And, as seen above, the same interdependence reproduces observed changes in the modern record of actual atmospheric measurements (our Fig. 3). Köhler et al. then complain that the paleoclimate records of Vostok and EPICA, in which temperature changes by 8 °C, are not global but local. By that standard, the entire industry of paleoclimate records can be dismissed. So too for the IPCC reports which rely heavily, if not uncritically, on those records. Paleoclimate records are, by necessity, point measurements (see, for example, Lisiecki & Raymo, 2005). Global observations are, for many properties, not available for the

¹⁰ This result is anticipated by the observed decay of ¹⁴C (Fig. 1). Despite a substantial increase in anthropogenic emission over successive decades, the decay of ¹⁴C remained almost perfectly exponential, corresponding to a constant absorption time of about a decade.

modern period. They are certainly not available for prehistoric periods.

The complaint surrounding which paleoclimate records to believe misses the point. In likelihood, no paleoclimate record gives a description of global-mean properties that is quantitatively accurate. What our analysis shows is that, with account of temperature dependence, changes of natural emission and absorption are sufficient to account for ancient changes of CO₂ inferred from any plausible record of paleoclimate. This fundamental mechanism is the same one which was shown to be sufficient to account for nearly all of the observed change in the modern record (Fig. 3).

Uncertainties surrounding the ice core record are numerous. Among them is low-pass filtering. Smoothing of CO₂ changes follows from the century-long compaction time of snow in the firn - the uppermost 100 m, where air is exchanged freely with the overlying atmosphere. Similarly, diffusion significantly influences the amplitude and phase CO₂ variations in air bubbles and secondary cavities, which thereby interact with surrounding ice (Jaworowski, 1992, 2004; Jouzel, 2013).

Cores extracted from different sites exhibit similar relative changes in CO₂ and temperature, including their interdependence. However, those changes differ in absolute value (e.g., Vostok vs EPICA). Among the sources of uncertainty is dating of gas bubbles and secondary cavities with respect to the surrounding ice (Petit et al., 1999; Jouzel, 2013). Discrepancies between the age of trapped air and the age of surrounding ice are as large as 2500 - 7000 years at Vostok and 1000 - 5000 years in the Fuji Dome ice core (Kawamura et al., 2003).

Warm periods can render information in ice sheets useless. When upper layers are sunlit, melt water from one level percolates into another, contaminating whatever properties were present. Much the same occurs when ice at higher temperature melts, destroying the layered structure relied upon for dating individual depths. For this reason, ice core data for temperatures even 2-3 °C warmer than during the 20th century were excluded from our analysis.

Of fundamental importance to interpreting the ice core record is the transformation of carbon dioxide to CO₂ clathrate, a hybrid solid wherein CO₂ actually joins H₂O in the molecular lattice. This transformation occurs for all gases at sufficiently great pressure. For CO₂, however, clathrate forms readily - at pressures of only 5 bars (Jaworowski et al., 1992). Corresponding to core depths of only ~100 m, this feature of carbon dioxide means that, over almost the entire ice core record, much of the CO₂ actually resided, not in gas phase, but in the form of clathrate.

The formation of clathrate imposes a serious limitation on relating CO₂ that has been extracted from ice cores to CO₂ that was in the atmosphere when that water substance was laid down. Once CO₂ has passed into clathrate phase, it cannot be recovered conservatively. When the core is extracted, it is necessarily unloaded of the overlying weight, relieving the pressure that binds CO₂ in clathrate. The sharp reduction of pressure then leads to an abrupt reverse transformation, CO₂ bound in clathrate being immediately transformed back into gas phase. Unavoidable, the abrupt formation of CO₂ gas then increases pressure within the compacted ice - explosively.

Extracted cores are proliferated with microscopic fractures (ibid; Jaworowski, 2004). Formed by unbalanced pressure inside the ice, those ruptures serve as veins through which CO₂ gas escapes. This process is inevitable because it is inherent to the compaction of ice at pressure and subsequent extraction of the ice, which relieves the overpressure. It is also irreversible because, once the core is extracted, CO₂ that was initially bound in clathrate is lost. For the same reason, the resulting error in relating CO₂ in the extracted ice to CO₂ that was in the ancient atmosphere is impossible to quantify.

Köhler et al. claim that the ice core record of CO₂ perfectly matches the modern record of actual atmospheric measurements. With respect, this claim is preposterous. The only reason that the two records agree is the introduction of a kludge. As shown in Fig. 4, actual CO₂ data recovered from ice disagree with contemporaneous measurements of CO₂ in the atmosphere. The two were artificially

brought into alignment by arbitrarily re-calibrating the ice core data – to force it to match actual measurements of CO₂ in the atmosphere. Air in uppermost layers of the Siple Dome ice core (less than 68 m depth and, hence, largely free of clathrate) was arbitrarily declared to be 83 years younger than the ice in which that air was trapped (Neftel et al., 1985). This arbitrary adjustment shifted CO₂ concentration of 328 ppm in ice that was dated to the year 1890 to the year 1973. Thereby, CO₂ retrieved from ice matched CO₂ in the record of actual atmospheric observations – another self-fulfilling prophecy.

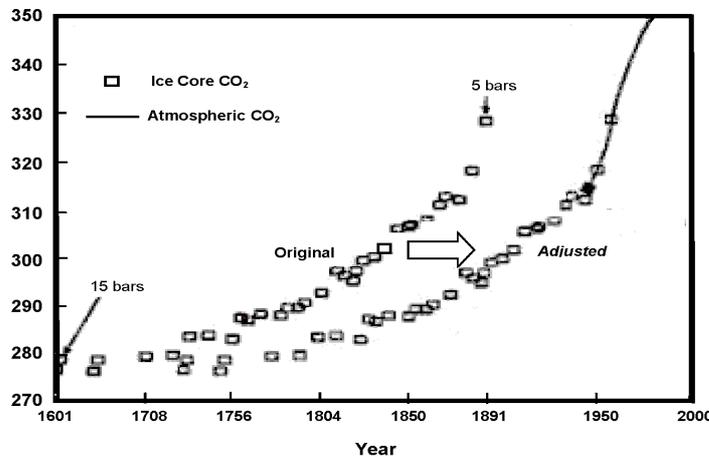


Fig. 4: CO₂ recovered from Siple Dome ice core, before and after adjustment (squares). Compared against atmospheric CO₂ (solid). Adapted from Jaworowski et al. (1992).

It is to be underscored that the period of overlap in the two records involves *only the firn*, the uppermost layer of ice core. Those shallow depths are sufficiently porous to exchange air freely with the overlying atmosphere. Young and at low pressure, the firn layer is free of the most confounding influences that plague deeper layers - the preponderance of the ice core, which is much older and was subjected to far greater pressure.

Fundamental limitations are inherent in all records of paleo climate, which, by necessity, involve pseudo observations of atmospheric behavior. An alternative to the ice core record of CO₂ is CO₂ inferred from plant stomata. Although discounted by Köhler et al. (if not provincially), the record of fossilized stomata evidences significantly greater changes of CO₂. Nevertheless, for relating modern changes to ancient changes, it too is limited. Figure 5 plots the sensitivity of stomata index to changes of atmospheric CO₂.

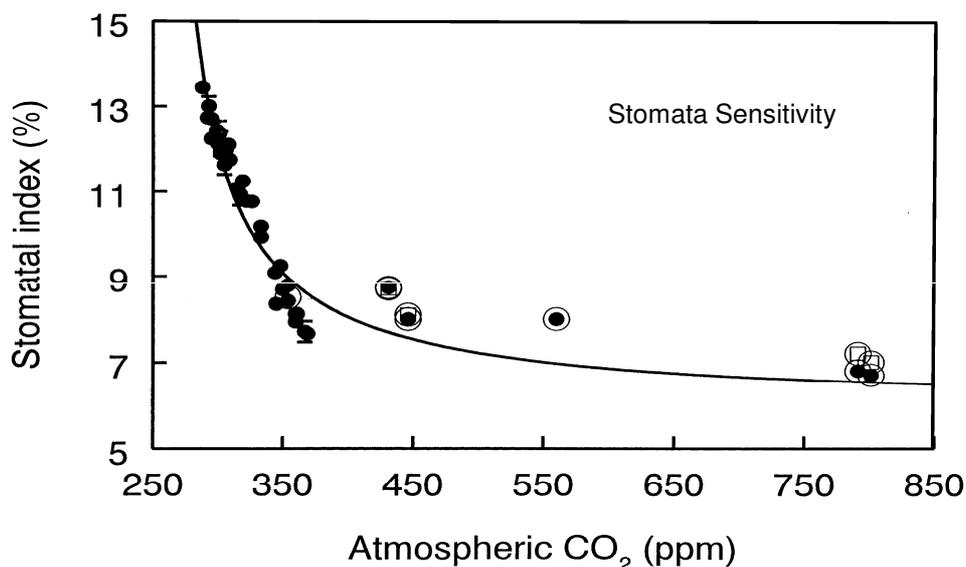


Fig. 5: Stomata index, as a function of atmospheric CO₂. After Beerling et al. (2002).

At modern concentrations, stomata index varies sharply with changes of CO₂. However, at CO₂ exceeding 450 ppm, the sensitivity of stomata all but vanishes. Consequently, in the stomata record of CO₂, 450 ppm cannot be distinguished practically from 1,450 ppm.

These and analogous considerations leave paleoclimate records of CO₂ clouded by uncertainty. They render claims of ancient changes in relation to modern changes largely speculative. Our paper explicitly recognizes uncertainties in the paleo record, noting that the ice core record is reliable only for the general trend of CO₂ in relation to temperature.

Figure 3 in Harde (2017) plots CO₂ concentration versus temperature at coincident times (core depths). From nearly 300 values in the Vostok record, CO₂ concentration was evaluated by interpolating between neighboring data. A plot of the original data (Fig. 6) shows that CO₂ concentrations corresponding to individual temperatures are scattered by as much as 80 ppm, or 30% in range. This variability reflects natural fluctuations in emission and absorption, as well as numerous uncertainties that cloud the ice core record of CO₂ in relation to atmospheric CO₂. Despite such variability, ice core CO₂ exhibits a clear and systematic increase with increasing temperature, one manifest over 150 kyr of the last glacial/interglacial period. Earlier periods exhibit similar dependence, albeit with greater variability. It is this interdependence of CO₂ and temperature, universal in the ice core record (e.g., Alley, 2010; Jouzel, 2013) that is accounted for in our nonlinear treatment of paleoclimate changes.

As shown in Harde (2017), the gradual transition from lower to present temperatures is well represented by an exponential variation, which approximates the long-term evolution present in the data. That long-term variation of temperature is, in fact, a generalization of the short-term variation that was adopted for the modern record. Over periods of order a century, it reduces to a linear variation of temperature, which in turn reproduces observed changes of CO₂ in the modern record (Fig. 3 above).

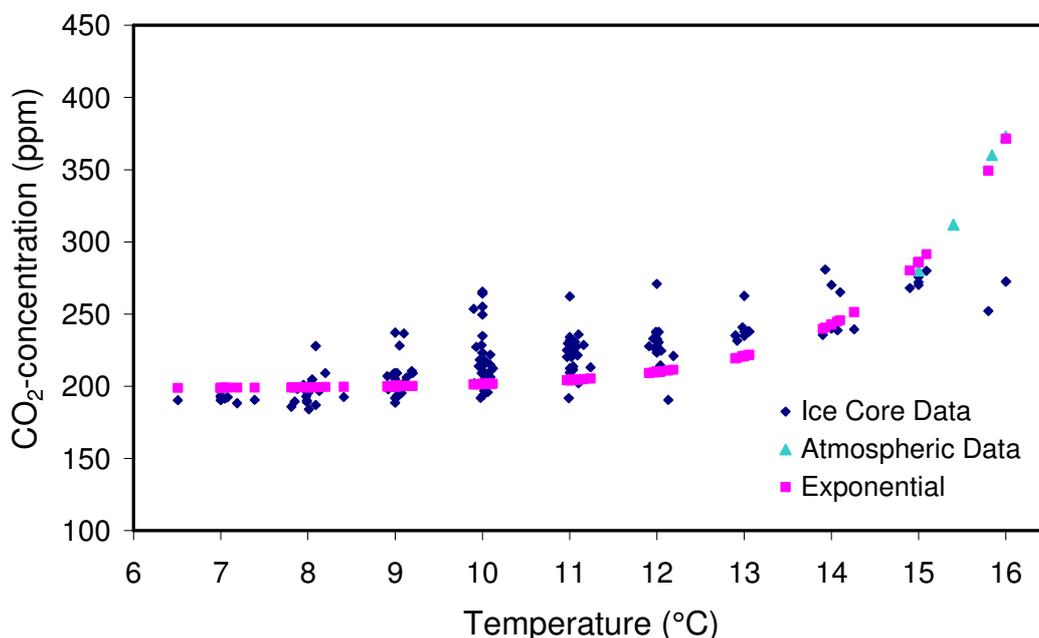


Fig. 6: Scatter plot of Vostok ice core data (diamonds) for the CO₂ concentration over temperature. Data derived from ice core data over the last 150 kyr. Also shown are atmospheric observations (triangles) and exponential dependence on temperature (squares).

5. Claim: Reference of Motivational Material is Invalid

Lastly, Köhler et al. claim that references to material which inspired our investigation but which has either been criticized provincially, on dubious merits, or has not appeared in a journal are invalid. Among the

treatments invoked by Köhler et al. to discount contradictory evidence are treatments which were shown in Section 1 to be unphysical. Köhler et al.'s complaint over source is ironic, contradicting their own position. To challenge our demonstration of fundamental physics, they cite material, even casual opinion, that was published on the internet. Accordingly, Köhler et al. expect one standard for others, but another for themselves.

Likewise invalid then are scholarly books, invited lectures, and summaries of conference presentations. Although not appearing in a journal, all are routinely cited in research literature. In fact, by the declared standard of Köhler et al, the IPCC reports themselves, which are relied upon heavily in this Comment, are invalid.

6. Closing

The considerations discussed above, supported in Appendix A by auxiliary considerations of various data, point to changes of temperature being a major driver of changes in atmospheric CO₂. Together, the modern record of actual atmospheric observations and the paleo record of pseudo observations demonstrate the following: The same physical mechanism which is sufficient to account for nearly all of the modern change of CO₂, without anthropogenic influence, is also sufficient to account for nearly all of its change from ancient times – whatever that change may actually be.

In regard to public policy, whether the time scale for removal of atmospheric CO₂ is 10 years, or 4 years, or 1 year matters not. Far shorter than relied upon by the IPCC, all of those removal times have the same implication: Most, if not the preponderance, of increasing CO₂ follows from mechanisms that are not under human control. Restricting the anthropogenic component of emission therefore amounts to an exercise in postponing the inevitable.

Acknowledgement

The author would like to thank Prof. Murry Salby, formerly Macquarie University Sydney, for his continuous interest in this so important field of climate science, and in particular for many extremely helpful and stimulating discussions concerning the physical principles with their consequences for an adequate description of the atmospheric carbon cycle. The author also thanks W. Soon from Harvard-Smithsonian Center for Astrophysics for his constructive suggestions when preparing this Reply.

Appendix A

3. Claim: Treatment is Too Simple

The absorption efficiency of extraneous reservoirs has been claimed to have decreased, based on changes in the arbitrarily-defined airborne fraction (e.g., Canadell et al., 2007; Le Quéré et al., 2009). Such claims are dubious because they rely on the presumption that changes of CO₂ are exclusively of anthropogenic origin. Nor are the claims supported by recent atmospheric CO₂ data. Gloor et al. (2010) found that decadal changes of AF followed from changes in the growth of anthropogenic emissions - not from changes in absorption efficiency, which were comparatively small. Further, uncertainties in emission and absorption exceeded any changes in AF. Ballantyne et al. (2012) arrived at a similar conclusion. They used global atmospheric CO₂ measurements and CO₂ emission inventories to evaluate changes in global CO₂ sources and sinks during the past 50 years. Their mass balance analysis indicates that net CO₂ uptake significantly increased, by about 0.18 Pg/yr (0.05 GtC/yr) and, between 1960 and 2010, that global uptake actually doubled, from 8.8 to 18.4 Pg/yr. It follows that, without quantitative knowledge of changes in natural emission, interpretations based on AF are little more than speculative.

The uptake and outgassing of atmospheric CO₂ by oceans is simulated with complex marine models. How much CO₂ enters or leaves the ocean surface is calculated from the difference between atmospheric and surface concentrations of CO₂, modified by the Revelle factor. However, most of these models involve dubious assumptions which are not in agreement with observed behavior (see, e.g., Steele, 2017). They assume that the surface layer absorbs CO₂ through equilibrium with atmospheric concentration. On this premise, they calculate how much Dissolved Inorganic Carbon (DIC) will be added to the ocean based on increased atmospheric CO₂ since pre-industrial times. In reality, the surface layer is not at equilibrium with the atmosphere. A difference in concentration results from conversion of CO₂ into organic carbon by photosynthesis. Organic carbon produced then sinks into the deep ocean, where it is sequestered. This downward transport to the deep ocean is known as the biological pump. Only in those regions where strong upwelling of DIC from the deep ocean exceeds sequestration of carbon via photosynthesis can CO₂ be outgassed to the atmosphere. The latter is found primarily in the tropical oceans (Takahashi et al., 2009; Zhang et al., 2017). Several models estimate that, without the biological pump, atmospheric CO₂ would be 200 to 300 ppm higher than current levels (see also Evans, 2011).

With increasing primary production, carbon export to depth also grows. Arrigo et al. (2015) reported that, since 1998, annual primary production in the Arctic has increased by 30%. Steinberg et al. (2012) observed a 61% increase in meso-plankton between 1994 and 2006 in the Sargasso Sea. The North Atlantic coccolithophores have increased by 37% between 1990 and 2012 (Krumhardt et al., 2016). And Chavez et al. (2011) found a dramatic increase in primary production in the Peru Current since the end of the Little Ice Age (LIA). Together, the increase in primary production and downward transport of organic carbon is sufficient to account for anthropogenic CO₂ that was absorbed from the atmosphere (Steele, 2017).

Further, seasonal changes in surface CO₂ illustrate that absorption of CO₂ by the oceans and accumulation of DIC near the surface are determined, not by the Revelle factor, but by the biological pump. Evans et al. (2011) found from buoy data off the coast of Newport, Oregon that each spring photosynthesis lowers ocean surface CO₂ to 200 ppm - far below current atmospheric concentrations and much lower than what would be expected from equilibrium with a pre-industrial atmosphere. Anthropogenic CO₂ in surface water is then quickly removed. It is also well known that higher concentrations of CO₂ magnify photosynthesis. At increased atmospheric CO₂, the plankton community consumed 39% more DIC (Riebesell et al., 2007). During summer and autumn, surface CO₂ can rapidly

increase to 1000 ppm - more than twice the concentration of CO₂ in the atmosphere. Surface water then significantly enhances natural emission to the atmosphere. Conversely, during winter, surface CO₂ remains at about 340 ppm. Despite reduced photosynthesis, CO₂ in surface water then remains below equilibrium with the atmosphere, reflecting efficient removal through downward transport by the biological pump. It is noteworthy that these strong seasonal variations of CO₂ in surface water are manifest in the record of atmospheric CO₂.

Under steady state conditions, diffusion of CO₂ into the ocean is believed to require about 1 year to equilibrate with an atmospheric perturbation. But, when increased sunlight enhances photosynthesis, such equilibration is no longer achieved. Perturbation CO₂ is then simply transported to depth, where it is sequestered from surface waters (MacDonnell et al., 2010). Under such conditions uptake of CO₂ is not restricted by the Revelle factor but by the biological pump.

The foregoing processes are controlled essentially by sunlight and temperature. There is no reason to believe that net primary production, the biological pump, and sequestration of CO₂ below surface waters would be the same today as 260 years ago, when temperature and atmospheric CO₂ were likely lower.

In simulating transport of carbon in the ocean, complex models assume behavior that is found in tracers like chlorofluorocarbons (CFCs). Because those species accumulate near the ocean surface, models assume DIC does as well. But unlike CFCs, which are inert, CO₂ entering sunlit waters is quickly converted to organic matter by photosynthesis (Steele, 2017). Although dissolved CFCs and dissolved carbon are passively transported in the same manner, particulate organic carbon (alive or dead) behaves very differently. It rapidly sinks, removing carbon from surface water through mechanisms which do not operate on CFCs.

The removal of carbon from surface water depends on the sinking velocity and also on how rapidly organic matter is decomposed. After descending below the pycnocline (depths of 500-1000 meters), carbon is effectively sequestered - because water at those depths does not return to the surface for centuries (Weber et al., 2016). For the atmosphere, this long-term sequestration translates into removal that is effectively permanent. Before such carbon can return to the atmosphere, fossil fuel reserves will have long since been exhausted (Section 1).

The combination of sinking velocities and sequestration depth suggests that a significant fraction of primary production is sequestered in a matter of days to weeks (Steele, 2017). Therefore, increasing primary production leads to a proportionate increase and rapid export of carbon to depth. If marine productivity has increased since pre-industrial times, it will have also sequestered the respective anthropogenic carbon into the deeper ocean. Observations from ocean basins suggest that, since the Little Ice Age, marine productivity and carbon export have indeed increased as the oceans warmed (Chavez et al., 2011; Abrantes et al., 2016).

The various mechanisms, along with their dependence on temperature and other environmental properties, could not have remained constant during the pre-industrial era. This inconsistency invalidates the fundamental IPCC assumption, that natural emission and absorption during the pre-industrial period did remain constant. Even less this is valid over the Industrial Era, a period which is characterized by the IPCC as the fastest rise in temperature over the Holocene or even the last interglacial. Equally inconsistent is the presumption that additional uptake of anthropogenic CO₂, which represents less than 0.5% of the total, has, somehow, exceeded the storage capacity of oceans and other surface and sub-surface reservoirs, capacity which is orders of magnitude greater. As seen in Section 1, the observed record of absorption exhibits no evidence of saturation (Fig. 1).

4. Claim: Application of Paleo climate Record is Incorrect

Temperature-dependence of natural emission and absorption is confirmed by recent analysis of

Antarctic ice from the Law Dome (MacFarling et al., 2006). Ice from the firn, which (unlike deeper ice) communicates freely with the overlying atmosphere, spans the past 65 years. It indicates that CO₂ growth temporarily halted during the 1940s and 1950s, maintaining values of 310 - 312 ppm. During the mid 1940s, extracted CO₂ actually decreased. The reversal in CO₂ growth coincided with global temperature decreasing (Moberg et al., 2005). It corresponds to an additional sink of ~ 10 Pg/yr (Trudinger et al., 2002). The record of $\delta^{13}\text{C}\text{O}_2$ suggests that the additional sink was caused, not by lower fossil emissions or a change in the terrestrial biosphere, but by a change in oceanic absorption.

Also examined by MacFarling et al. (2006) was ice that was deeper and older. Extracted CO₂ manifests a conspicuous decrease of ~10 ppm, one that is accompanied by a simultaneous decrease in CH₄ of 40 ppb. Reflecting the strong correlation between changes of carbon dioxide and methane in the ice core record (e.g., Petit et al., 1999), both appeared during the 16th and 17th centuries - a period of decreasing temperature known as the Little Ice Age (LIA). From this they follow that the 10-ppm decrease in CO₂ likely resulted from reduced temperature during LIA and not from vegetation re-growth.

Appendix B

Carbon 14 obeys the balance equation

$$\frac{dC_{14}}{dt} = E_{14} - \alpha C_{14}. \quad (\text{B1})$$

E_{14} , which represents emission of relative concentration, follows from background emission of ^{14}C as well as anthropogenic emission. Because fossil fuel is devoid of ^{14}C , the latter dilutes net emission of ^{14}C - the so-called Suess effect.

Anthropogenic emission of CO_2 is a small fraction of overall CO_2 emission (in 1960, about 1%).

If the fraction from anthropogenic emission is

$$\varepsilon = \frac{E_A}{E} \ll 1, \quad (\text{B2})$$

then the diluted net emission of ^{14}C is given by

$$E_{14} = E_{14}^0 (1 - \varepsilon), \quad (\text{B3})$$

where E_{14}^0 is the background emission of ^{14}C , emission that would result in the absence of dilution. By reducing E_{14} , dilution by emission that is devoid of ^{14}C ($-\varepsilon E_{14}$) acts to reinforce absorption ($-\alpha C_{14}$) in (B1). Thereby, it accelerates decay of the concentration C_{14} over what it would be in the presence of absorption alone. Exclusive of other influences, the apparent absorption time in ^{14}C is therefore shorter than the actual absorption time. But how strong is this effect?

Following elimination of the nuclear source, anomalous ^{14}C is governed by the perturbation balance equation

$$\frac{dC'_{14}}{dt} = E'_{14} - \alpha C'_{14}, \quad (\text{B4})$$

where primed quantities are referenced against unperturbed values before introduction of the nuclear source. For the sake of illustration, we consider background emission

$$E_{14}^0 = \text{const}. \quad (\text{B5})$$

Then, from (B3) the perturbation in dilution is

$$\begin{aligned} E'_{14} &= -\varepsilon' E_{14}^0 \\ &\cong -\varepsilon' E_{14}, \end{aligned} \quad (\text{B6})$$

where ε' is the change in fractional anthropogenic emission (B2) during the interval of decay. E'_{14} represents the additional dilution that is introduced following elimination of the nuclear source. At the end of the ^{14}C record, anthropogenic emission was $\sim 75\%$ greater than at the beginning, when anthropogenic emission was approximately 1% of the total emission. It follows that, throughout the record of ^{14}C decay,

$$\varepsilon' \leq 0.0075. \quad (\text{B7})$$

Therefore,

$$E'_{14} \geq -0.0075 E_{14}. \quad (\text{B8})$$

The perturbation balance equation (B4) then becomes

$$\frac{dC'_{14}}{dt} \geq -0.0075 E_{14} - \alpha C'_{14}. \quad (\text{B9})$$

Now the unperturbed balance (i.e., exclusive of the nuclear source) is, under the conditions of (B5),

$$E_{14} = \alpha C_{14}. \quad (\text{B10})$$

Hence, (B9) becomes

$$\begin{aligned} \frac{dC'_{14}}{dt} &\geq -0.0075\alpha C_{14} - \alpha C'_{14} \\ &\geq -\alpha C'_{14} \left(1 + 0.0075 \frac{C_{14}}{C'_{14}} \right). \end{aligned} \quad (\text{B11})$$

The apparent absorption rate is thus

$$\begin{aligned} \alpha_{App} &= -\frac{1}{C'_{14}} \frac{dC'_{14}}{dt} \\ &\geq \left(1 + 0.0075 \frac{C_{14}}{C'_{14}} \right) \alpha. \end{aligned} \quad (\text{B12})$$

Relative to the actual absorption rate, α_{App} is weakly accelerated:

$$\frac{\alpha_{App}}{\alpha} \geq 1 + 0.0075 \frac{C_{14}}{C'_{14}}. \quad (\text{B13})$$

Now, ^{14}C decayed following removal of the perturbing nuclear source, which left absorption unbalanced (B1). As seen in Fig. B1, anomalous ^{14}C then was

$$\frac{C'_{14}}{C_{14}} \cong 3. \quad (\text{B14})$$

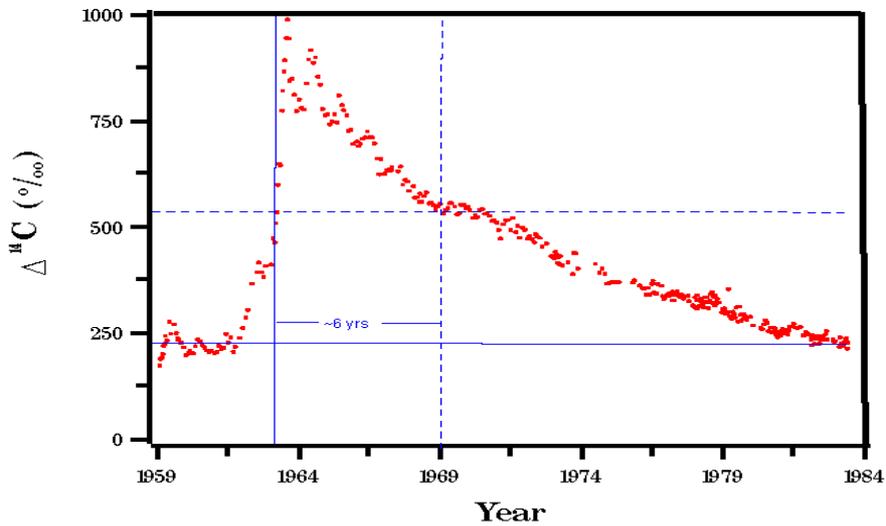


Fig. B1: Evolution of ^{14}C observed at Vermunt Austria. Source: CDIAC

(B13) then becomes

$$\frac{\alpha_{App}}{\alpha} \geq 1 + 0.0025 \quad (\text{B15.1})$$

or, in terms of the absorption time,

$$\frac{\tau_{App}}{\tau} \leq 1 - 0.0025 \quad (\text{B15.2})$$

Relative to actual absorption, the apparent absorption time is *shorter* by less than 1%. Even after two e-foldings, the apparent absorption time is shorter than that of actual absorption by less than 2%.

The dilution effect of fossil fuel emission is inconsequential. Far more influential is re-emission of ^{14}C that was absorbed from the atmosphere. On the timescale of observed absorption, little of that ^{14}C is sequestered beneath the Earth's surface. A substantial fraction is therefore returned to the atmosphere through re-emission (e.g., through decomposition of vegetation which has absorbed that ^{14}C).¹¹ In the perturbation balance (B4), re-emitted ^{14}C opposes absorption of ^{14}C . Unlike the dilution effect, which is minor, it therefore slows decay over what it would be in the presence of absorption alone. The apparent absorption time is therefore *longer* than the actual absorption time. Consequently, the actual absorption time must be even shorter than a decade.

¹¹ Because it is proportional to absorption, re-emission simply modifies the effective absorption rate. The resulting decay therefore remains exponential (Fig. B1).



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