



# Clear Thinking about Atmospheric CO<sub>2</sub>

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## Abstract

Several articles have been published in this journal purporting to show that the well-documented rise in atmospheric CO<sub>2</sub> is a natural phenomenon rather than human caused. This note reviews the overwhelming case that human activities are the cause. It identifies specific misunderstandings about the carbon cycle and errors in the interpretation of radiocarbon data contained in these papers. Most importantly, misconceptions about the conclusions that can and cannot be drawn from the present composition of the atmosphere are highlighted.

**Keywords:** carbon cycle; net global uptake; radiocarbon; bomb pulse; isoflux

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## 1. Introduction

A cornerstone of the argument that humans are responsible for climate change is the consensus among climate scientists that human activities such as the burning of fossil fuels have caused the rise of atmospheric CO<sub>2</sub> concentration during the Industrial Era, all of it. This is perhaps the most well-established piece of the case for anthropogenic global warming. Yet a few dissenters continue to argue that natural sources are the primary cause of the increase. Papers in *Science of Climate Change* such as (Harde and Salby 2021), (Berry 2021), and (Schroder 2022), and in *Health Physics* (Skrable et al. 2022a; 2022b) conclude that human emissions have made a relatively small contribution. While these papers have no impact on mainstream climate science, they may confuse lay readers who read their conclusions but do not have the tools to critically analyze them. Most active climate scientists ignore such papers if they are even aware of them. But this author believes that it is a mistake to allow misconceptions and errors go unchallenged. The errors need to be clearly spelled out in front of those same lay audiences, to ensure that the development of public policy is based on sound science. The arguments made here are limited to the question of responsibility for atmospheric CO<sub>2</sub> increases and are not original with the author.

A simple and compelling argument that human emissions, not natural sources, have caused the increase will be presented in Section 2. But that will still leave the question of how these authors came to their mistaken conclusions. It will be unnecessary to scrutinize details of their individual calculations because they share a common misconception discussed in Section 3. The quantity that all the papers attempt to calculate, or infer from data, is the fraction of carbon in the present atmosphere that was once contained in a fossil fuel. They call this the “human contribution”, or the “fossil component”, or “the fraction due to fossil fuel burning”. Although at first glance this quantity seems to be a valid measure of human impact, in fact it is not. As we will see, the dynamic atmosphere is more subtle. Finally, in an Appendix, we discuss the misanalysis of radiocarbon data. Errors in earlier papers (Berry 2019) and (Harde 2017; 2019) have been noted before (Andrews 2020). The modified model of (Harde and Salby 2021), an attempt to correct

Harde's earlier mistake, is not credible.

## 2. Human Emissions Cause the Increase

I cite the results of (Ballantyne et al. 2012) who combine all land and sea reservoirs both for simplicity and to avoid uncertainties in the magnitude of changes in individual non-atmospheric reservoirs. Very good measurements are in hand of total atmospheric carbon accumulation between 1960 and 2010. Ballantyne uses CO<sub>2</sub> concentration data from a network of approximately 40 marine boundary sites (the NOAA/ESRL flask network) and converts to pentagrams of carbon in the total atmosphere with the factor 2.124 PgC/ppm. The uncertainty quoted takes into account sampling errors. Good estimates of human emissions are also in hand. Ballantyne uses emission data from the Carbon Dioxide Information Analysis Center, BP, and the Emissions Database for Global Atmospheric Research. Carbon conservation then yields a good estimate of "Net Global Uptake" during that period. Ballantyne's Figure 2 numbers are shown schematically in Figure 1 below.

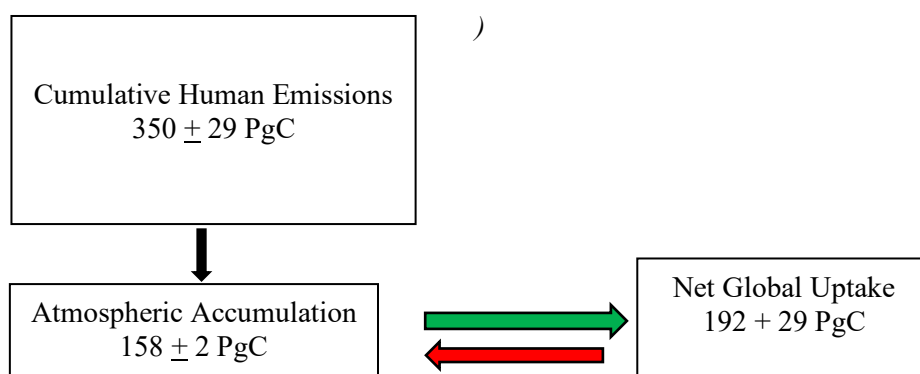


Fig 1: Total carbon changes 1960-2010 from Ballantyne(2012)  
(1 PgC = 1 billion metric tons of carbon)

The positive Net Global Uptake for this period is the *net* quantity of carbon that has moved from the atmosphere into land/sea reservoirs. It is the difference between much larger gross exchanges in both directions. Differences between large, similar numbers usually contain large errors, but in this case carbon conservation allows Net Global Uptake to be determined more accurately than either gross exchange. The qualitative observation that humans have put more carbon into the atmosphere than has remained applies not only to the fifty-year span shown, but throughout the Industrial Era, and is seen in the Mauna Loa data sets used by most authors.

Because the carbon from human emissions during this period exceeds the rise in atmospheric *total carbon*, we know immediately that land and sea reservoirs together have been net sinks, not sources, of *total carbon* during this period. We can be sure of this without knowledge of the detailed inventory changes of individual non-atmospheric reservoirs. This is not a model dependent result. It is a simple statement based on carbon conservation, data on emissions and atmospheric levels, and arithmetic. Note that this conclusion contains no assumption whatsoever about the constancy of natural carbon in this period. In fact, the primary conclusion of Ballantyne is that non-atmospheric natural reservoirs have, during the 50-year period studied, not only increased their carbon inventory, they have also increased the rate at which they are doing so in response to the higher atmospheric levels. Nor does the conclusion rely on treating "human" and "natural" carbon differently as (Harde and Salby 2021) and (Berry 2021) both allege. Net Global Uptake is simply what is left over after atmospheric accumulation has been subtracted from total emissions. If more carbon was injected into the atmosphere by human activities than stayed there, it had to have gone somewhere else. It is also a statement that has been made many times before (Cawley 2011); (Richardson 2013); (Denning et al. 2022).

The red arrow in Figure 1 represents processes which transfer carbon from land/sea inventories to the atmosphere: outgassing of oceans, outgassing of freshwater ponds, decay of vegetation, human breathing, even volcanos. (Ballantyne attributes a temporary stabilization of Net Global Uptake in the 1990's to the eruption of Mt Pinatubo in 1991, among other things.) (Schroder 2022), like (Harde and Salby 2021), speculates that outgassing from oceans due to temperature increases is a major factor in atmospheric CO<sub>2</sub> rise. *The data in Figure 1 falsify this hypothesis.* Processes like outgassing are part of the carbon cycle and are accompanied by processes by which carbon is transported from the atmosphere to the oceans, represented by the green arrow. Discussing credits without discussing debits is bad accounting. The green arrow includes the dissolution of CO<sub>2</sub> in the oceans and freshwater ponds, and photosynthesis. If the rates of natural outgassing-like processes (red arrow) were exceeding the rates of natural processes by which carbon is removed from the atmosphere to land/sea reservoirs (green arrow), then they would cause Atmospheric Accumulation to exceed Cumulative Human Emissions, making Net Global Uptake negative. The data for the period 1960-2010 cited by Ballantyne clearly say otherwise. If outgassing-like processes were important sources of new carbon that had not been in the atmosphere in recent centuries, then they would have a radiocarbon signature showing that. They do not.

Ballantyne chose to study a limited 50-year period with the best data in order to understand how the natural sinks were changing with time. During this period, Net Global Uptake was always positive. This has not always been the case. For example, 56 million years ago natural processes caused atmospheric CO<sub>2</sub> to increase by about 10,000 GT over a period of several thousand years. (Voosen 2022). Since human emissions were zero 56 million years ago, Net Global Uptake was then clearly negative. Natural non-atmospheric carbon reservoirs were then net sources, unlike in the present era. The present anthropogenic sources are an order of magnitude larger.

We note that (Beck 2022) presents a record of atmospheric CO<sub>2</sub> levels in the first half of the 20th century that differs markedly from the US Energy Information Administration data set used by most authors (EIA 2022.) Beck assembled this record from observations at multiple locations, by multiple researchers using multiple techniques. This author has no insight into reasons for the discrepancy between the two records. The Beck record shows a substantial rise in atmospheric CO<sub>2</sub> between about 1920 and 1940, a rise which exceeds anthropogenic emissions in that period. Therefore, Net Global Uptake was negative in those years according to the Beck record, though it was positive according to the EIA record. In the Beck record, CO<sub>2</sub> levels then fall rapidly from 1940 to 1950. No reason is given for the change in the sign of Net Global Uptake, i.e, for the sudden switch around 1940 of natural reservoirs from sources to sinks. Both data sets agree that averaging over the entire period from 1900 to 1960, anthropogenic emissions exceeded atmospheric CO<sub>2</sub> rise and natural reservoirs were net sinks. It is worth remarking that over 70% of the CO<sub>2</sub> rise in the Industrial Era has occurred since 1960, a period in which Beck does not challenge EIA data, and a period in which natural reservoirs have clearly been sinks. This is the period which is depicted in Figure 1.

In their quest to determine the fraction of anthropogenic carbon in the present atmosphere Harde and Salby, Berry, and Schroder focus on anthropogenic and natural carbon separately. This - complicates their analysis and they miss the simple conclusions made here. What they do find is that natural carbon is accumulating in the atmosphere faster than human carbon, *and indeed it is!* Nothing in the analysis presented here conflicts with this fact. We will see in Section 3 why this happens. Anthropogenic carbon can be the cause of the entire Industrial Age increase without being a large part of the present atmospheric composition. Of course, it is the *total* atmospheric carbon that impacts climate, and the above analysis leaves no doubt where the increase in the total is coming from. Tracking anthropogenic carbon separately from natural carbon is a distraction, which is why few papers in the serious peer-reviewed literature do so.

As a final comment for this section, note that a positive Net Global Uptake is not only measured, but also completely expected. The higher partial pressure of CO<sub>2</sub> in the atmosphere in 2010 compared to 1960 leads, by Henry's Law, to higher carbonate concentrations in the oceans. Ocean acidification has been observed and confirms what Net Global Uptake tells us: the primary

reservoir, the oceans, have on balance globally been taking in net carbon, even while outgassing net carbon at some locations. Natural processes mitigate anthropogenic caused CO<sub>2</sub> rise; they do not enhance it.

### 3. Why “Natural Carbon” accumulates in the atmosphere over “Human Carbon”

#### 3.1 The concept of isofluxes

The two-way carbon exchanges between the atmosphere and other reservoirs shown in red and green in Figure 1 are each much larger than their difference which Net Global Uptake measures. In other words, the flows in either direction are almost balanced, though a small net excess flows out of the atmosphere. What some do not necessarily appreciate is the consequence of these balanced two-way exchanges. Balanced exchanges are equivalent to simple mixing. Carbon in the atmosphere mixes with carbon in the oceans in the course of dissolving in one place and outgassing in another.

Because of the mixing, composition differences between the carbon inventories of different reservoirs tend to decrease. This is no different than say, pouring 30% of an alcohol and water mixture back and forth between two containers. If Container A started with a higher alcohol concentration than Container B, after several transfers the two concentrations will have become more equal. Even with no net exchange of liquid, a net exchange of alcohol between containers will have occurred. In the same way, even with little or no net exchange of carbon between the atmosphere and oceans, a substantial net exchange of, for example <sup>14</sup>C can occur. The phenomenon of isotope flow between reservoirs by this process has a name among specialists: isotope disequilibrium fluxes or “isofluxes” (Levin et al. 2010). The direction of the isoflux flow always tends to reduce the composition difference between the two reservoirs. It is simply about mixing.

#### 3.2 Understanding the bomb pulse with isofluxes

We can illustrate the workings of isofluxes by looking at some atmospheric <sup>14</sup>C data. Figure 2 shows two measures of atmospheric radiocarbon from 1920 to 2015. The line in green is of “Δ<sup>14</sup>C” (left axis), the standard variable by which the <sup>14</sup>C community presents data, from 1915 to 2015. Only northern hemisphere data from (Graven et al. 2017) is shown, to align with the data used by (Harde and Salby 2021). The most prominent feature of this graph is the “bomb pulse”, the dramatic increase in atmospheric <sup>14</sup>C from atmospheric nuclear testing in the 1950’s and early 1960’s, before the 1963 Test Ban Treaty. Clearly this perturbation presents an opportunity to learn about exchange processes.

Δ<sup>14</sup>C measures the fractional deviation of the *specific activity* of a sample from a standard, in parts per thousand (‰):

$$\Delta^{14}C = 1000 \left[ \frac{A_{measured}}{A_{standard}} - 1 \right] \quad (1)$$

That is, it describes what *fraction of the carbon* in a sample is <sup>14</sup>C. It does not, in general, measure the concentration of <sup>14</sup>C i.e., the *fraction of the sample* that is <sup>14</sup>C. It can be used as a proxy for concentration only when the total carbon in a sample is fixed, as it is in a liter of oxalic acid. The <sup>14</sup>C/<sup>12</sup>C ratio (or <sup>14</sup>C/C<sub>total</sub>) is not only what is most directly measured, but also what is useful for <sup>14</sup>C dating. The standard used depends on the measurement technique. Common A<sub>standards</sub> are 226 Bq/kgC or 1.176x10<sup>-12</sup> mole<sup>14</sup>C/moleC. [See (Stuiver and Polach 1977), (Stenstrom et al. 2011), (Andrews 2020).]

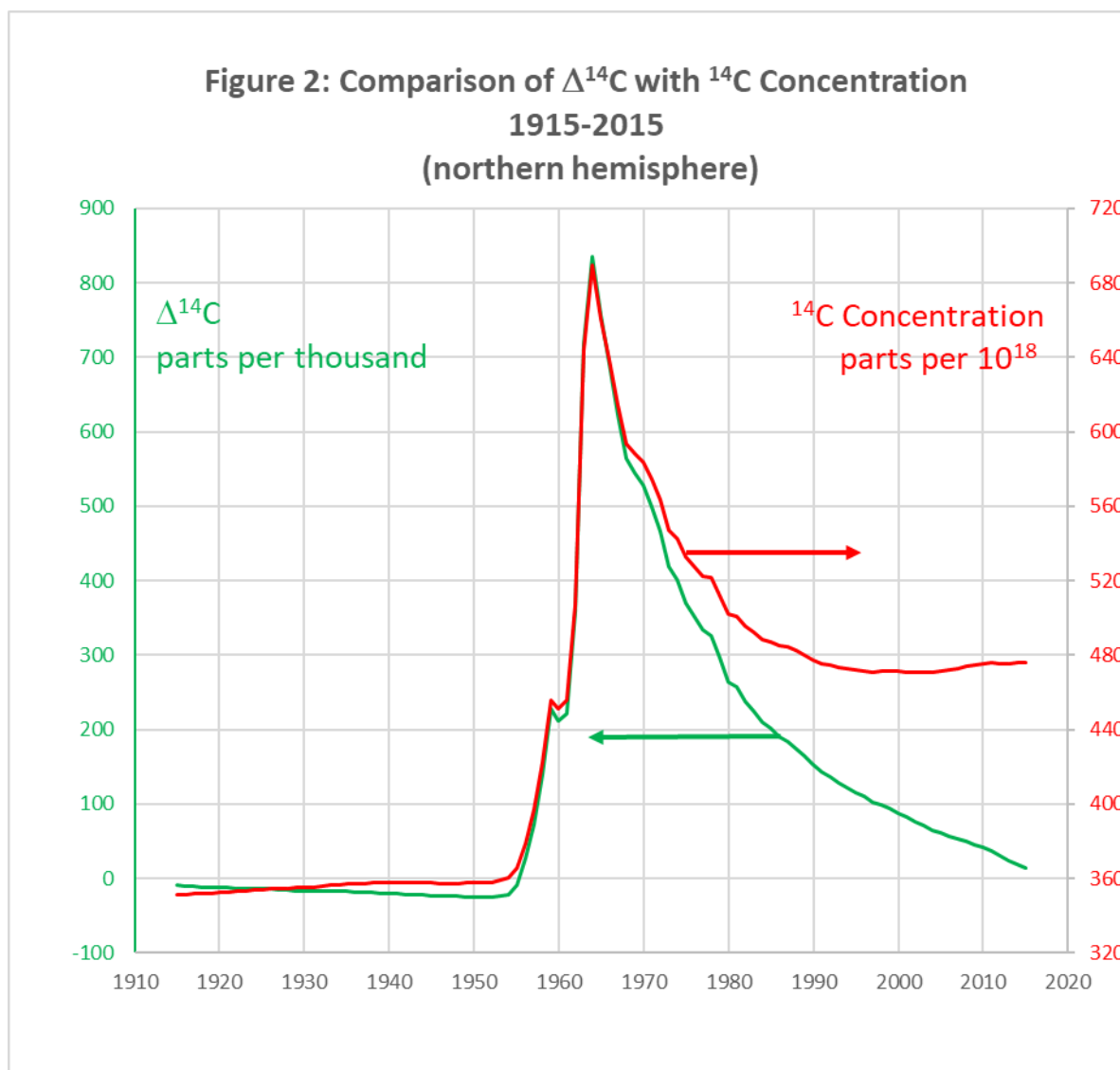


Fig 2. Northern Hemisphere Atmospheric Radiocarbon 1915-201.  $\Delta^{14}\text{C}$  measures the fraction of carbon in a sample that is  $^{14}\text{C}$  compared to a standard.  $^{14}\text{C}$  Concentration is the molar fraction of the atmosphere which is  $^{14}\text{C}$ .

Figure 2 also shows in red the atmospheric  $^{14}\text{C}$  concentration (right axis) during the same period, calculated from the same  $\Delta^{14}\text{C}$  and data on total atmospheric carbon concentration from the US Energy Information Administration (EIA 2022). After the “bomb pulse”, the  $^{14}\text{C}$  concentration did not return to its 1950 value. It remained about 30% higher.

While nuclear testing was increasing the specific activity of the atmosphere, the “Suess effect” was decreasing it.  $^{14}\text{C}$  is naturally produced in the upper atmosphere from cosmic rays at a more or less steady rate, and carbon cycle processes distribute it to the oceans and biosphere. (See the Appendix for a discussion of small variations in the natural production rate of  $^{14}\text{C}$ .) Because the half-life of  $^{14}\text{C}$  is about 5730 years, fossil fuel carbon which has been isolated from the atmosphere for much longer times is essentially devoid of  $^{14}\text{C}$ ; it is “cold”. When released into the atmosphere by fossil fuel burning, this cold carbon reduces the specific activity (measured by Bq/kgC or by the  $^{14}\text{C}/\text{C}_{\text{tot}}$  ratio) by simple dilution. This effect was noticed in (Suess 1955). Wood produced in 1950 had lower specific activity than wood produced in 1900 because of the presence of cold anthropogenic carbon in the atmosphere, diluting the cosmic-ray-produced component and potentially complicating  $^{14}\text{C}$  dating.

Using the concept of isofluxes caused by two-way exchanges (mixing), we can understand fine details of both  $\Delta^{14}\text{C}$  and concentration shown in Figure 2:

- From 1915 until the early 1950's, the addition of anthropogenic cold carbon into the atmosphere by fossil fuel burning lowered  $\Delta^{14}\text{C}$  via the Suess effect, as shown by the slightly decreasing green curve. With  $^{14}\text{C}/\text{C}_{\text{tot}}$  now lower in the atmosphere than in the oceans, an isoflux of  $^{14}\text{C}$  moved from the oceans to the atmosphere, causing a small but clear rise in the red curve during the same period. This is perhaps a first hint that exchange processes cause subtle effects: *adding cold carbon to the atmosphere ended up increasing its  $^{14}\text{C}$  concentration!*
- The atmosphere was left with a much higher  $^{14}\text{C}/\text{C}_{\text{tot}}$  than the oceans after the bomb testing ceased. Mixing between the atmosphere and other reservoirs by two-way exchanges brought both measures of atmospheric radiocarbon down between 1965 and 1990. The characteristic time for the mixing can be seen to be about one decade.
- While two-way exchanges were diminishing the “bomb pulse”, anthropogenic cold carbon was being added to the atmosphere. Eventually, around 2000,  $^{14}\text{C}/\text{C}_{\text{tot}}$  in the atmosphere was again less than it was in the oceans for the first time since the early 1950's. The isotope gradient had reversed sign. This again caused a net flow, an isoflux, of  $^{14}\text{C}$  from the oceans to the atmosphere. The red curve began increasing again, just as it had between 1915 and 1950.

The narrative here is qualitative, but quantitative analyses have been performed by (Caldiera et al. 1998), and more recently by (Graven et al. 2020). The Caldeira analysis preceded and anticipated the rise in  $^{14}\text{C}$  concentration beginning about 2000. Successful predictions are the hallmark of good science.

### 3.3 Applying the isoflux concept to “human” and “natural” carbon

If we think of human (anthropogenic) and natural (all other) carbon as two types of carbon whose fraction can be different in different environmental reservoirs, not unlike the situation with isotopes, then the concept of mixing via two-way balanced exchanges can be applied. The net result of exchanges will again be a tendency to reduce differences between the human fraction in the atmosphere and the human fraction in the other reservoirs. Fossil fuel burning creates an excess of anthropogenic carbon in the atmosphere's composition, compared to land and sea. The “isoflux-like” flows will therefore move natural carbon into the atmosphere and human carbon out of it.

We can make a rough quantitative estimate of what these flows do to the atmospheric composition. From the bomb pulse data, we know the mixing time is about one decade. Were human emissions to abruptly stop, isoflux-like flows would cause the atmospheric human fraction to approach the land/sea human fraction, a quite small number, in a couple of decades. Therefore, it is the last decade's worth of human emissions, which have yet to be diluted from exchange processes, which dominate the present value of the atmosphere's “human component.”

### 3.4 Measuring and interpreting the human component of atmospheric CO<sub>2</sub>

This rough estimate of the present human component can be corroborated by data on the present specific activity of the atmosphere. (Skrable et al. 2022a)'s initial analysis of this was compromised by wrong input data for the specific activity and a lack of appreciation of the lingering effects of bomb carbon. In a follow-up paper, (Skrable et al. 2022b) used for the specific activity record a “no bomb scenario” model developed by (Graven et al. 2020). Although this means that the input data used is itself model dependent, that is of no concern here. The Skrable papers developed a simple formula to measure the human carbon fraction of the atmosphere in any year, based on the observed magnitude of the Suess effect. They take the 2018 “no bomb scenario” atmosphere to contain a mixture of cold carbon ( $\Delta^{14}\text{C} = -1000$ ) and carbon with  $\Delta^{14}\text{C} = 0$ . (Had

no nuclear testing occurred, the 2018 value of  $\Delta^{14}\text{C}$  would be  $-.130$  in Graven et al.'s "no bomb scenario".) This leads to Skrabble et al.'s inferred measurement via isotope analysis of what they call the "fossil fraction". This quantity is what Berry, Harde and Salby, and Schroder attempt to calculate. This is the quantity that (Segalstad 1992) inferred from  $^{13}\text{C}$  data. These authors all equate this fraction to the fraction of human responsibility for CO<sub>2</sub> increase.

The number (Skrabble et al. 2022b) get for the fossil component in 2018 is 321 GT CO<sub>2</sub>, (87 GT C). This is 32% of the atmospheric carbon increase since 1750, motivating them to declare it to be "much too low to be the cause of global warming". *But they ignored mixing from two-way, nearly balanced exchanges in jumping to this conclusion* (Schwartz et al. 2022), (Andrews 2022) It is noteworthy that the 321 GT CO<sub>2</sub> number they get is quite comparable to the 346 GT CO<sub>2</sub> of human emissions in the years 2009-2018, the last decade of their data set. All that they have really measured is the anthropogenic carbon emitted in roughly the past decade. That is about all the information that can be extracted from the present atmospheric composition. Mixing has obscured the earlier history. But that is no problem. We learned in Section 2 why atmospheric CO<sub>2</sub> is increasing.

The Skrabble analysis illustrates that the measured Suess effect is substantially smaller than it would have been without isofluxes. This was noted by Suess himself: "The decrease [in specific activity] can be attributed to the introduction of a certain amount of C<sup>14</sup>-free CO<sub>2</sub> into the atmosphere by artificial coal and oil combustion *and to the rate of isotopic exchange between atmospheric CO<sub>2</sub> and bicarbonate dissolved in the oceans*" [italics added]. In the same paper Suess began the rich history of using radiocarbon to learn about atmospheric processes when noting "the rate by which this CO<sub>2</sub> exchanges must be greater than previously assumed" (Suess 1955).

### 3.5 A helpful analogy

We have seen that the statement "Human carbon in the present atmosphere is only 30% of the Industrial Age increase" is not the same as "Human emissions caused only 30% of the increase." As the disconnect between current inventories and fundamental causes is subtle, an analogy may be helpful for understanding it. (Cawley 2011) proposed a good one, worth repeating verbatim here:

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

Cawley goes to the trouble of showing with a Monte Carlo simulation that, after some time, Belgian coins make up only 3% of the inventory, even though they accounted completely for the savings increase. In a like manner human carbon, while a relatively small percentage of carbon in the present atmosphere, is completely responsible for the increase. A net sink of total carbon can raise natural carbon levels elsewhere, but it cannot raise total carbon levels elsewhere.

## 4. Summary

Since this article was first drafted, (Harde and Salby 2022) has been published. Unfortunately the newer paper contains the same fatal flaws as the earlier papers already discussed. We can use it to summarize those flaws:

- (Harde and Salby 2022) focuses on tropical temperature dependent outgassing, a regional

analysis. No doubt it contains some insights. But like the other papers critiqued it ignores the well-established fact that Net *Global* Uptake is solidly positive in the current era as the authors should know. All credible models need to be constrained by this simple observation.

- (Harde and Salby 2022) correctly notes that natural emissions and absorptions approximately balance. The mixing that results surely modifies isotope distributions as discussed in Section 3. Isoflux effects explain and dominate the evolution of <sup>14</sup>C distributions, both specific activity and concentration, over the last 100 years as described in Section 3.2. Isoflux effects can change carbon isotope distributions without changing total carbon distributions. These authors ignore them and mistakenly believe that total carbon changes mimic isotope changes. See the appendix for further criticism of the radiocarbon model of these authors.

Scientific progress depends upon original ideas challenging the consensus. Scientific progress also relies on the use of empirical data to weed out ideas that may be original but are also just plain wrong. That progress is helped when the originators of the wrong ideas acknowledge their errors and find constructive ways to contribute. That does not always happen, and it is not happening in the present case. The unconventional ideas critiqued here have been around for well over a decade. It should not have been necessary for this article to refer to a 2011 paper to, once again, refute them.

As an example of authors' clinging to old and discredited ideas, we will describe (Harde and Salby 2021)'s attempt to salvage Harde's model by designing (inventing?) a background to transform the true concentration curve (red in Figure 2) to the curve they originally thought was the concentration (green in Figure 2). As the analysis here is not part of the main argument of this paper, it is relegated to an appendix. For a credible model of the evolution of the bomb pulse, see Section 3.2 or the quantitative and peer-reviewed analyses cited.

### **Appendix: Clinging to wrong ideas: the radiocarbon model of Harde and Salby**

The two curves in Figure 2 describing atmospheric radiocarbon history were confused by (Berry 2019) and by (Harde 2017;2019), as pointed out by (Andrews 2020). They wrongly believed that concentration had followed the green curve rather than the red curve. Theories built around the false belief that concentration had returned to its pre-bomb test value now had to explain why, after initially falling quickly after atmospheric nuclear testing ended, <sup>14</sup>C concentration had since roughly stabilized 30% or more above the pre-1950 plateau that had persisted for centuries. If the ~30% increase does not include lingering bomb carbon as it certainly appears to, what caused it? (Berry 2021) does not address this obvious question, though he drops the many references to <sup>14</sup>C data contained in (Berry 2019). (Harde and Salby 2021) recognize their need to address the baseline shift, because (Harde 2019) had used <sup>14</sup>C data, wrongly interpreted, to supposedly validate his questionable model.

(Harde and Salby 2021) follows (Harde 2019) in hypothesizing that not only the flow of carbon from atmosphere to oceans, but the fast cycle flow in the other direction, from the oceans to the atmosphere, also depends only on the carbon concentration *in the atmosphere* (!?) They put this into their model through their Equation 6:  $e_{R,14} = \beta C_{14}/\tau$  This term represents the rate of reemission to the atmosphere of <sup>14</sup>C that has been taken up by, say, the ocean. Since  $\beta$  and  $\tau$  are treated as constants determined from fits in this analysis, equation 6 asserts that reemission is proportional to  $C_{14}$ , the concentration *in the atmosphere*. The higher the concentration in the atmosphere, say Harde and Salby, the higher the flow *into it*. This conjecture defies common sense, but its consequences to Harde and Salby's analysis are clear. If the flows both to and from the atmosphere were indeed each proportional to atmospheric concentration, then the net flow would also be proportional to atmospheric concentration, and a single time constant exponential function would of course describe the net exchange, as it does in their model.

(Harde and Salby 2021) treat showing that <sup>14</sup>C concentration fell with a simple exponential form



as the crucial empirical question. To transform the red curve in Figure 2 to the green curve, their solution is to postulate new sources of atmospheric <sup>14</sup>C that started after about 1964. These are the components of the  $e'_{NB,14}(t)$  term in their Equation 10. From their fits to the concentration data assuming an effective time constant of 10 years, they arrive at an evaluation of its magnitude:

$$e'_{NB,14}(t) = 123 \text{ \%}/\text{yr} + .3 \text{ \%}/\text{yr}^2 \times (\text{year} - 1990) \quad \text{valid after 1964} \quad (2)$$

They chose to express concentration as a dimensionless variable showing deviation from a standard, analogous to the definition  $\Delta^{14}\text{C}$ . We will put this into a more transparent form for comparison with other studies. Since a  $\Delta^{14}\text{C}$  standard is  $1.176 \times 10^{-12}$  (moles <sup>14</sup>C)/ (moles of total C) and taking 315.8 ppm as the total CO<sub>2</sub> abundance in 1959 (Harde and Salby's chosen year to define a standard) their <sup>14</sup>C concentration standard is  $371.4 \times 10^{-12}$  ppm. Taking the total atmosphere to contain  $1.77 \times 10^{20}$  moles, then this standard can also be expressed as 65.71 kmoles of <sup>14</sup>C. The emission rate in (2) is then equivalent to:

$$e'_{NB,14}(t) = 8.08 \text{ kmoles}/\text{yr} + .0197 \text{ kmoles}/\text{yr}^2 \times (\text{year} - 1990) \quad \text{valid after 1964} \quad (3)$$

From Figure 2 we see that prior to 1950, the atmospheric abundance of <sup>14</sup>C was approximately constant around  $355 \times 10^{-12}$  ppm. If it is removed with an effective time constant of 10 years, as Harde and Salby argue, then its production rate needs to be  $35.5 \times 10^{-12}$  ppm/year, or 6.28 kmoles/year. (The equilibrium level equals the time constant times the emission rate.) So Harde and Salby would say:

$$e'_{NB,14}(t) = 6.28 \text{ kmoles}/\text{yr} \quad \text{before 1964 (assumes their 10 yr time constant)} \quad (4)$$

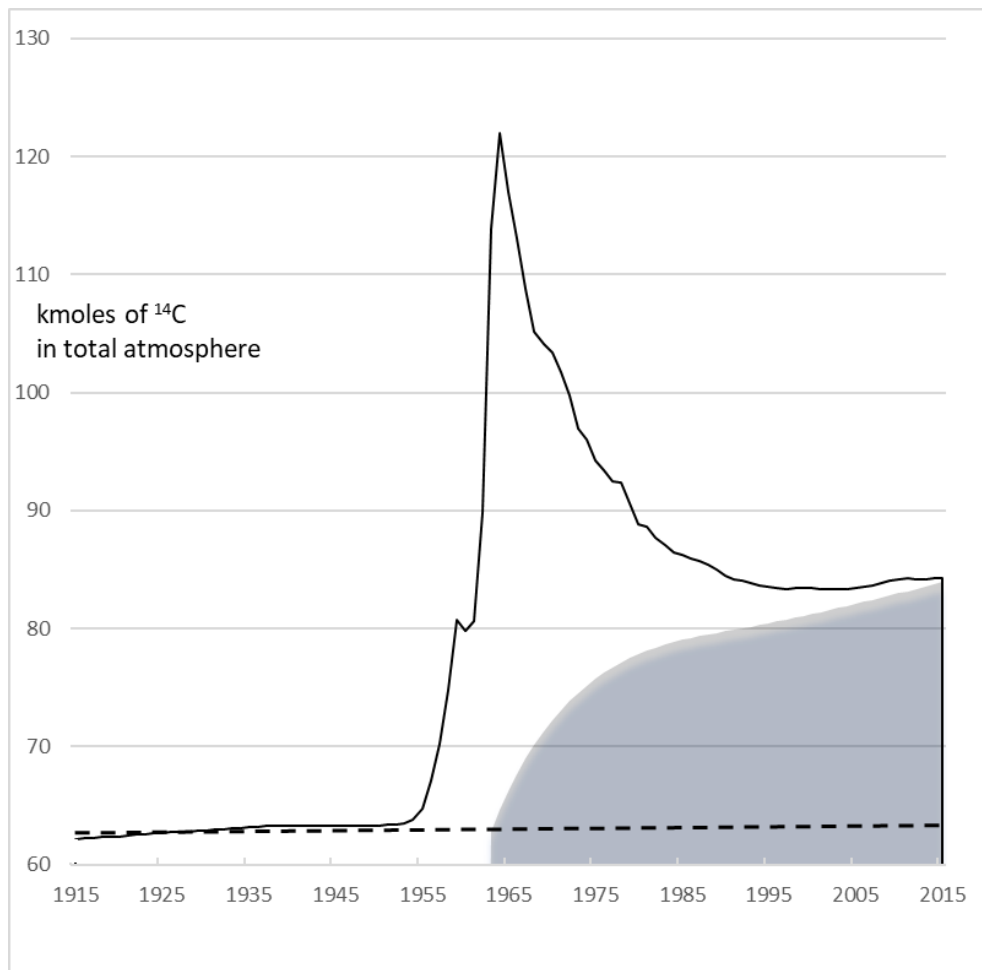


Fig 3 The Engineered Background of Harde and Salby

Figure 3 shows the now familiar bomb pulse concentration curve. This is the same data as in Figure 2, but with the y axis now converted to kmoles in the total atmosphere, and with the background generated by Harde and Salby's emission function (3) shown as the shaded areas. The dashed line is the background level generated by emission function (4), based on the pre-test data. As in Harde and Salby's model, the background is being depleted with a 10-year time constant. It is maintained at the emission rate (4) until (3) switches on in 1964. The new postulated sources increase the background towards a new equilibrium level of 80.8 kmoles. Then in 1990 the linearly increasing "cosmic ray term" in (3) switches on.

To defend their model, Harde and Salby needed to show that the bomb pulse abundance had fallen to near 0 by 2015, some 4 of their 10-year time constants after cessation of testing. They did this by engineering the background to be essentially the same as the data in 2015, as shown. The difference between the solid line and the shaded area is, they say, the real signal, not the difference between the solid line and the dashed line. But (Harde 2019) used the dashed curve as the background. (Harde and Salby 2021) say that the new background accounts for nuclear power plant emissions, continued nuclear testing, and a change in the natural background. *Their justification is only qualitative, yet the size of this engineered background is completely responsible for their claimed "success" in showing that the bomb carbon had gone away by 2020. Their method simply assumed this result. It did not determine it.*

Even though Harde and Salby provide no quantitative estimates of <sup>14</sup>C released by nuclear power plants, this has been studied and documented. Figure 1 in (Zazzeri et al. 2018) shows the estimated global <sup>14</sup>C emissions of nuclear power plants from 1972 through 2016, by year and by country. Rough integration of that plot through 2012 shows about 3920 TBq of <sup>14</sup>C had been emitted globally, some as <sup>14</sup>CO<sub>2</sub> and some as <sup>14</sup>CH<sub>4</sub>. One mole of <sup>14</sup>C has an activity of 2.31 TBq, so about 1.7 kmoles of <sup>14</sup>C have been released into the atmosphere from nuclear power activities by this estimate *in 44 years*. This is slightly less than the 1.8 kmoles (8.08 -1.68) that Harde and Salby's model needs to have emitted above the pre-test baseline *every year*. Nuclear power plant emissions cannot save their model.

(Naegler and Levin 2006) estimate a combined total of about 8 kmoles of <sup>14</sup>C released from nuclear testing, nuclear plant operations, and other industrial activities between 1970 and 2005, i.e., a rate of .23 kmoles/year. (See their Figure 4). This would account for about 14% of the background Harde and Salby got by forcing it to rise to the data.

Nuclear testing and nuclear power plants do not come close to accounting for the baseline shift. Let us finally consider the possibility that changes to the incoming flux of cosmic rays can explain it. <sup>14</sup>C dating started with the assumption of a constant historical value for atmospheric  $\Delta^{14}\text{C}$ , i.e., a constant production rate produced by a constant flux of cosmic rays. But that assumption has been refined. Samples whose age is known, say from counting tree rings, have been used to investigate the history of atmospheric  $\Delta^{14}\text{C}$  and calibrate the age vs specific activity curve (Damon and Peristykh 2000). Detailed plots of historical values of  $\Delta^{14}\text{C}$  over the last 11,000 years do show some variations that are possibly cyclic. But the *maximum* short-term excursions from a constant are about 20 parts per thousand, or two percent. (See their Figure 2.) Harde and Salby, on the other hand, propose a step change in cosmic ray flux close to 30 %. For this to have happened without being noticed elsewhere is highly unlikely.

There is another reason to eliminate cosmic ray increases as an explanation for the failure of the <sup>14</sup>C concentration to return to its pre-test level. Fluctuations in the historical cosmic ray flux have been inferred from fluctuations in  $\Delta^{14}\text{C}$ , not fluctuations in concentration. But  $\Delta^{14}\text{C}$  has not done anything unexpected in recent decades. Has the new <sup>14</sup>C from more cosmic rays been precisely balanced by a flux of <sup>14</sup>C devoid carbon from somewhere? Again, this is hardly likely.

Note that Harde and Salby's model's need to invent so much background anomalous <sup>14</sup>C arises from their belief that it is continually removed on a 10-year time scale. Since the observed <sup>14</sup>C

concentration has not declined since 2000, they must keep adding more to explain the data. Of course, if they were to acknowledge that the relatively short time constant observed in the original bomb pulse corresponds only to the initial mixing of bomb carbon into the “fast cycle”, and that longer time scales are needed to describe the subsequent exchange of atmospheric <sup>14</sup>C with other sinks, their problem would be solved.

Putting in their postulated backgrounds, Harde and Salby go on to label a curve “theory” in their Figure 5. It is a most impotent “theory” that must invent an implausible, ad hoc background to salvage their model’s need for a simple exponential decay of atmospheric <sup>14</sup>C. This was done to “establish” an hypothesis that never made sense in the first place. Harde and Salby’s “fits” to the data are meaningless. Any curve can be transformed into any other curve if one is free to engineer the background. No one who has taken the time to understand the details of what Harde and Salby’s did can take their model seriously.

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