# Getting the Cause of Atmospheric CO2 Rise Wrong

David Evans Andrews<sup>1</sup>

<sup>1</sup>University of Montana

November 24, 2022

#### Abstract

Papers published in Science of Climate Change by Harde and Salby (2021), and by Berry (2021) contend that the well documented growth of atmospheric CO2 in recent decades is predominantly a natural phenomenon rather than human caused. Both papers are wrong on two counts. First, they focus only on their own (suspect) calculations of anthropogenic atmospheric carbon levels, rather than on measured total carbon levels. The present concentration of anthropogenic carbon in the atmosphere, even correctly calculated, does not establish the human contribution to the rise in total atmospheric carbon. Second, the models these authors develop for determining present anthropogenic atmospheric carbon levels are incompatible with 14C data. The sources of background "anomalous 14C" postulated by Harde and Salby fall well short of being large enough to salvage their model. A highly questionable assumption built into Harde and Salby's model is explicitly identified.

#### Getting the Cause of Atmospheric CO<sub>2</sub> Rise Wrong

David E Andrews Department of Physics and Astronomy (Retired) The University of Montana (April 10, 2022)

#### Abstract

Papers published in Science of Climate Change by Harde and Salby (2021), and by Berry (2021) contend that the well documented growth of atmospheric CO<sub>2</sub> in recent decades is predominantly a natural phenomenon rather than human caused. Both papers are wrong on two counts. First, they focus only on their own (suspect) calculations of anthropogenic atmospheric carbon levels, rather than on measured total carbon levels. The present concentration of anthropogenic carbon in the atmosphere, even correctly calculated, does not establish the human contribution to the rise in total atmospheric carbon. Second, the models these authors develop for determining present anthropogenic atmospheric carbon levels are incompatible with <sup>14</sup>C data. The sources of background "anomalous <sup>14</sup>C" postulated by Harde and Salby fall well short of being large enough to salvage their model. A highly questionable assumption built into Harde and Salby's model is explicitly identified.

#### **1** Introduction

This paper is a commentary on: "What Controls the Atmospheric  $CO_2$  Level" by Harde and Salby, Science of Climate Change vol. 1, no. 1, Aug 30 2021, pp 54-69, (hereafter "Harde and Salby") and on "The Impact of Human  $CO_2$  on Atmospheric  $CO_2$ " by Berry, Science of Climate Change, vol. 1, no.2, Nov 11 2021, pp 1-46 (hereafter "Berry"). Reviewing these two papers together is informative. They reference each other and come to similar conclusions about the role of anthropogenic carbon emissions in atmospheric  $CO_2$  increase. But they also contradict each other on essential points, as will be discussed. They cannot both be correct, but they most certainly can both be wrong.

Both papers challenge the consensus view that the rise in atmospheric  $CO_2$  is the direct result of human activities, mainly the burning of fossil fuels. The 2013 IPCC report on the carbon cycle (Ciais et al. 2013) provides a concise description of the carbon cycle. Some useful points from this report follow:

- Large, natural, two-way exchanges of carbon occur between the atmosphere and the oceans and land biomass. The fluxes each way exceed the one-way flux into the atmosphere of anthropogenic carbon from fossil fuels.
- New CO<sub>2</sub> added to the atmosphere from fossil fuel burning becomes part of this "fast cycle" exchange, on a time scale sometimes called the "residence time." This time is less than a decade and can be thought of as the time it takes carbon put into the atmosphere from whatever source to move, often temporarily, to either a land or ocean sinks.
- Atmospheric carbon, having gotten to another reservoir in about one residence time, has a significant probability of returning to the atmosphere. But there are now a large range of timescales possible. There is certainly no a priori reason to think that the time scale for reemission is the same as the time scale for the original absorption. If an atmospheric carbon atom was captured in seasonal foliage, it likely gets back in the atmosphere within a year or two. If it goes into the bark of a young redwood tree, it could be there for centuries. The IPCC therefore discusses multiple residence times. Others talk of a longer effective "adjustment" time, for example associated with the slow mixing of surface water and deep oceans. Processes by

which carbon is incorporated into limestone or other rock are said to remove carbon from the fast cycle entirely as millennia will pass before that carbon returns, if ever, to the atmosphere. The number of parameters necessary to describe this complexity with any desired precision is an empirical question, determined by how well models using them describe measurable quantities.

- Human activities generate more than two times the CO<sub>2</sub> than is necessary to account for the atmospheric carbon rise, as will be discussed in Section 2. This simple fact is well known and not disputed by the authors of these papers but may not be known to many of their readers.
- The IPCC reports do not attempt to analyze human and natural carbon separately. This paper will make clear why they have little motivation to do so.

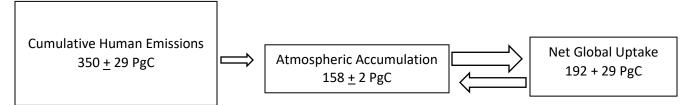
The critique of the two papers will focus on two general issues. In Section 2 their incomplete analysis of the carbon cycle will be emphasized. They have much to say about atmospheric "human carbon", but next to nothing to say about total carbon, and total carbon is both what is observable and what matters. Understanding the cause of atmospheric total carbon increases is not the same as estimating what fraction of carbon in the current atmosphere was once in the form of fossil fuels. In Section 3, the focus is on their <sup>14</sup>C analysis. Both papers respond to Andrews (2020) which pointed out a clear and undisputed error that Harde(2017, 2019) and Berry(2019) had made in earlier papers. The <sup>14</sup>C analysis is particularly central in Harde and Salby. It was central to the argument of Berry (2019) but is deemphasized in his 2021 paper for reasons which will become clear. This paper will show that the models presented in both papers conflict with <sup>14</sup>C data.

## 2. Carbon accounting: Keep it Simple

Measurements of atmospheric carbon inventories, estimates of biomass size, and measurements of increasing ocean acidity all measure total carbon, not "human" or "natural" carbon separately. The conclusions of the two papers about percentages of current atmospheric carbon that originated from human activities are therefore not directly testable. Exploring the implications of observable changes in total carbon is a more fruitful approach. Neither paper attempts this. Harde and Salby argue that the inventories of carbon reservoirs other than the atmosphere are too poorly known to draw any conclusions. On the other hand, the IPCC's estimates of the sizes of those reservoirs are Berry's input data.

Berry models the carbon cycle as analogous to an RC circuit with three resistors and four capacitors. Such a model seems far too simple to elucidate much about the real-world carbon cycle, but that is an issue we will postpone until Section 3. The main oversight of Berry, as well as of Harde and Salby is not excessive simplicity. On the contrary, *they don't simplify enough*. They are distracted by detail irrelevant to the main question: "What is the impact of human emissions on the atmospheric  $CO_2$  level".

To accommodate Harde and Salby's concern about uncertainties in individual reservoir totals, we adopt the method of Ballantyne et al. (2012) who combine land and sea reservoirs into "global uptake". With very good measurements in hand of atmospheric carbon changes between 1960 and 2010, and good estimates of human emissions, carbon conservation allows us to get good estimates of "global uptake" in that period. Ballantyne's Figure 2 numbers are shown schematically in Table 1 below. They are a sobering reminder that over 70% of the rise in atmospheric  $CO_2$  during the Industrial Era has occurred since 1960:



Since the total carbon emissions into the atmosphere from human activity exceed the rise in atmospheric carbon, we know immediately that land and sea reservoirs together have been net sinks, not sources, of carbon during this period. We can be sure of this without knowledge of the detailed inventory changes of individual non-atmospheric reservoirs. Note that this conclusion contains no assumption whatsoever about the constancy of natural carbon in this period. Nor does it rely on treating "human" and "natural" carbon differently as both papers allege. 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 fossil fuel burning than stayed there, it had to have gone somewhere else.

Berry's corresponding numbers for cumulative emissions and atmospheric accumulation are in his Figure 11. His emission numbers for 1960-2010 are somewhat lower than Ballantyne's, perhaps because of different handling of carbon from land use changes by his source. Still, global uptake deduced from his numbers remains solidly positive. There is no disagreement about this fact: over the course of one year or fifty years more carbon has been leaving the atmosphere than entering it via natural exchange processes. These natural processes therefore can hardly be the cause of the increase. While the larger two-way exchanges are *removing* net carbon, the smaller one-way emissions from human activities are *adding* carbon and are therefore the cause of the increase. This is a logical conclusion, not an "IPCC assumption".

The simple analysis above does not tell us the present composition of the atmosphere in terms of human and natural carbon, but that is of secondary importance. It is quite possible that large gross flows of "natural" carbon, from the oceans to the atmosphere and back, could substantially dilute the fraction of "human" carbon in the fast cycle, and therefore also in the atmosphere. But 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. Modeling the fraction of "human" carbon in the fast cycle of "human" carbon in the fast cycle circulation, as both papers attempt, is a distraction from the main question. We want to know first and foremost what caused the increase, and we do. While nature is helping us by removing somewhat over 50% of the carbon we have been adding, *the entire increase is traceable to human emissions*.

The argument made here is not at all a new one, having been made by Cawley (2011) and by Richardson (2013) among others. Harde (2019) attempted a rebuttal of those critiques in his Section 5.3 "Environment as a Net Sink". He argues opaquely that "As long as any fraction of human emissions is involved, the environment is always a net sink. This is true per definition, since up to now no artificial uptake exists." This is nonsense. That net global uptake is positive in the current era (human emissions exceed atmospheric accumulations) is an empirical fact, not a logical necessity. One can easily imagine, as Harde himself and others have, that warming and outgassing oceans make the environment a net source, rather than a sink. But if that were the case in the current era atmospheric accumulation would exceed human emissions. Data show it does not. Though the present 400+ ppm level has likely not been reached in 3 million years,  $CO_2$  level variations have been seen in the past. When levels rose about

140,000 years ago (much more slowly than now), we can be sure that atmospheric accumulations were then exceeding the (zero) human emissions, and the environment was then a net source.

Harde (2019) goes on to point to his Figure 10 as evidence that "*nature is always a net sink*". What his Figure 10 illustrates is only that a model can be constructed that is compatible with positive net global uptake and which still allows accumulated natural carbon to dominate accumulated human carbon in the present atmosphere. Again, that is of secondary importance and does not change conclusions about the cause of atmospheric carbon increase.

We are zeroing in on a basic conceptual error common to both papers: they mistakenly infer the cause of the atmospheric carbon increase from their calculated current human carbon abundance. But the statement "Human carbon in the present atmosphere is only 50% of the Industrial Age increase" is not the same as "Human emissions caused only 50% of the increase." As the disconnect between current inventories and fundamental causes is subtle, an analogy may be helpful for understanding it. Cawley 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 can be a small percentage of carbon in the present atmosphere, yet 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.

Analyses of human and natural carbon separately have produced untestable conclusions, wrong inferences, and no useful insights. Perhaps that is why they are not included in IPCC reports.

## 3.0 The <sup>14</sup>C Analysis and Data

Human activities drive the increase in atmospheric CO<sub>2</sub>, regardless of the exact current abundance of anthropogenic carbon in the atmosphere. But are human carbon levels correctly calculated in these papers? The calculations cannot be directly compared with data because measurement of the human component alone is not possible. Radiocarbon 14 movements between reservoirs, on the other hand, can be traced with measurements. This section expands on the conflict between <sup>14</sup>C data and the models constructed by these authors, conflicts that were pointed out in Andrews (2020) and which are inadequately addressed in the new papers.

Harde and Salby sum up the main argument of their paper in the second sentence of their abstract: "*The* exponential decline of anomalous  ${}^{14}CO_2$  establishes that absorption of  $CO_2$  is determined not by extraneous reservoirs of carbon, but autonomously by the atmosphere". In other words, in their model the rate at which the oceans (or land) take in carbon from the atmosphere depends only on the concentration of carbon in the atmosphere. It is, they say, independent of the concentration of carbon

already in the oceans, or in land biomass, or anything else except the atmospheric concentration. This result is, they say, empirically confirmed by  ${}^{14}C$  data.

Berry makes a similar statement: "outflow is proportional to level", but Harde and Salby take it further. They hypothesize 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<sub>14</sub>, 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, then the net flow would of course describe the net exchange, as it does in their model. Harde (2019), Berry (2019), and Harde and Salby claim that this unlikely hypothesis is empirically confirmed with <sup>14</sup>C data. We show below that an error in the earlier papers and inappropriate data manipulations in the current paper negate this conclusion.

Note that Berry makes the more reasonable assumption that the more human carbon there is in the (surface) ocean, the more will flow from it into the atmosphere. He describes a *serial* process: carbon goes from the atmosphere to the oceans, then back to the atmosphere. As a result, Berry's is not a single time constant model, as will be shown in Section 3.2. Only parallel processes are allowed in Harde and Salby's one-time-constant logic.

In summary: To establish their unlikely hypothesis, Harde and Salby need to make a convincing case that data show the decline of anomalous <sup>14</sup>C is described by a simple one time-constant exponential function. Putting a theory to the test is good science. Before discussing their present attempt to do this, let us look at the relevant <sup>14</sup>C data, and describe Harde and Berry's past attempts to interpret it.

## 3.1 <sup>14</sup>C Data and Past Mistakes Interpreting It

Figure 1 shows a plot in green of " $\Delta^{14}$ C" (left axis), the standard variable by which the <sup>14</sup>C community presents data, from 1920 to 2015. Only northern hemisphere data from Graven et al. (2017) is shown, to align with the data used by Harde and Salby. 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, described by both papers being reviewed. Harde and Salby are correct that the bomb pulse presented an invaluable opportunity to use <sup>14</sup>C as a tracer to study the dynamic behavior of atmospheric carbon and the rate at which it is exchanged with land and ocean sinks.

 $\Delta^{14}$ C measures the fractional deviation of the *specific activity* of a sample from a standard, in parts per thousand (‰):

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

That is, it *always* 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 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/kg<u>C</u> or 1.176x10<sup>-12</sup> mole<sup>14</sup>C/<u>moleC</u>. See (Stuiver and Polach 1977, Stenstrom et al. 2011, Andrews 2020).

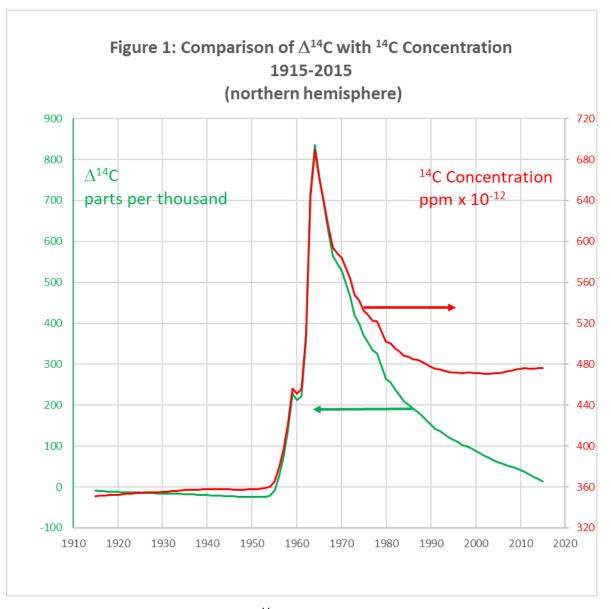


Figure 1 also shows in red the atmospheric <sup>14</sup>C *concentration* (right axis) in ppm during the same period, calculated from the same  $\Delta^{14}$ C and data on total atmospheric carbon concentration from the US Energy Information Administration, EIA (2022). After the "bomb pulse", the <sup>14</sup>C concentration did not return to its 1950 value. It remains about 30% higher. How this arises from the data is easily understood. The isotope ratio measured by  $\Delta^{14}$ C has (nearly) returned to its old value, but in the meantime total carbon has increased by about 30%.

Essenhigh (2009), Harde (2017), and Berry (2019) were motivated by the green  $\Delta^{14}$ C curve in Figure 1 to develop their models. They all wrongly thought that  $\Delta^{14}$ C measured concentration, rather than an isotope ratio; see Andrews (2020). Salby shows that he was making the same mistake in a 2015 video. (Reference 4 in Harde (2019)). The green curve can be well fit between 1970 and 2010 by a single time constant exponential. (It diverges from an exponential in more recent years.) If the *concentration* followed that curve, their models would be interesting, but instead it follows the red curve. These authors all subsequently concluded that anthropogenic carbon plays a minor role in the rise in atmospheric CO<sub>2</sub>.

Conventional analyses in which this mistake was not made reached a different conclusion. Berry, and Harde and Salby in their new papers all now concede that in fact concentration follows the red curve.

To correct their earlier mistake, Harde and Salby define a new dimensionless variable " $(\Delta^{14}C)_c$ " to measure concentration, and Berry defines "14CR" for the same purpose. Their two variables are similar, but not identical. The former fixes " $(\Delta^{14}C)_c$ " to equal  $\Delta^{14}C$  in 1959, and the latter fixes "14CR" to equal  $\Delta^{14}C$  in 1970. Both dates are poor choices to fix a standard, because  $\Delta^{14}C$  was changing rapidly in those years. In 1959 the southern hemisphere  $\Delta^{14}C$  lagged the value of the northern hemisphere (where the testing was concentrated) by 87 parts per thousand. There is little motivation to measure concentration with a dimensionless variable, but if that were deemed desirable one could simply define a standard concentration. In most of the analysis to follow I will convert these variables to the abundance, or concentration, in ppm in the interest of transparency. [Note: Plotting data in terms of a dimensioned variable instead of a dimensionless ratio obligates one to get the normalization constant right. In Andrews (2020) the y axis in Figure 2 is off by a factor of 1.38, accounting for the difference between that plot and Figure 1 here, now corrected. Also, in that plot world averages rather than northern hemisphere value were shown.]

Both Harde and Salby, and Berry convert  $\Delta^{14}$ C to their chosen concentration variables and plot it. The concentration curve in Figure 1 differs from Hardy and Salby's Figures 2, 3, and 5, and from Berry's Figure 18, in the scale and intercept, but not in the shape of its evolution. It also differs in the range plotted. Figure 1 here extends the data plotted back to 1915, to better show the "bomb pulse" in context. Harde and Salby's plots start in 1959 after the testing had already begun and D<sup>14</sup>C was already some 20% above the pretest baseline. Their plots omit the plateau prior to 1957 and give the false impression that the pre-test baseline for "( $\Delta^{14}$ C)<sub>c</sub>" was 200, when in fact it was around -20.

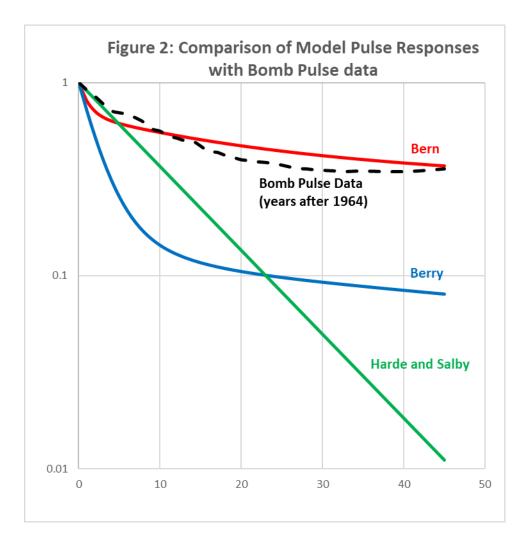
The focus of carbon transport studies is on the decline in <sup>14</sup>C concentration, from about 1965 onwards. Because Harde and Salby, and Berry are primarily about rates of removal of carbon from the atmosphere, having now realized their error, their new models should require extensive modifications from the earlier papers (Berry (2019), Harde (2017,2019)). Remarkably, very relevant data changes have not affected their models. This is discussed in the next section.

## 3.2 Understanding the <sup>14</sup>C data

Like dye added to a water system, <sup>14</sup>C can be used to trace geographical details of flows through the atmosphere and under the ocean surface (Levin and Hesshaimer 2000, Turnbull et al. 2009). Its utility here is to assess the response function of simplified models to a quick injection of new atmospheric carbon. The models being reviewed assume no geographical variations and well mixed atmospheres. Since atmospheric nuclear testing continued for the better part of a decade, and northern and southern hemispheres mix on a time scale of one or two years, the "bomb pulse" is an imperfect "pulse" for these models. Nevertheless, it presented an opportunity to observe the response of the carbon cycle to a perturbation. The response function to an injection of "anomalous carbon" (to use Harde and Salby's term) for Berry's model is explicitly displayed in his Figure 17, where it is compared with the response function of the IPCC's Bern Model (Joos 2013). The same two response functions plotted in Berry's Figure 17 are plotted here in Figure 2, using the year 1964 as the approximate time of the pulse to allow comparison with data.

Harde and Salby's response function, a 10-year single time constant exponential, is also added in Figure 2. Note that the vertical axis here has a logarithmic scale, whereas in Berry's plot the scale was linear. On a logarithmic scale, a single time constant exponential function forms a straight line. The break in the slope around 1975 (year 10) in Berry's model confirms the observation made in section 2.0 that his model

has more than one time constant. But comparing models with each other is of less interest than comparing them with data. The <sup>14</sup>C concentration curve in Figure 1 shows a long, approximately stable baseline before 1950. Its value around 355 x  $10^{-12}$  ppm is established by a balance between atmospheric <sup>14</sup>C production by cosmic rays, its natural distribution into land and sea sinks, and eventual nuclear decay. The dashed line in Figure 2 is the "anomalous <sup>14</sup>C": the concentration data of Figure 1 with the 355 x  $10^{-12}$ ppm plateau subtracted, then normalized to the 1964 value. It should be no surprise that the Bern Model fits the data better than the models being reviewed. It was built with the <sup>14</sup>C data in hand, and with the relationship between  $\Delta^{14}$ C and concentration correctly understood.



A feature of the  $\Delta^{14}$ C data in Figure 1 is its slow decrease to negative values before 1950, beginning from a preindustrial value of 0. This is the "Suess Effect" (Suess 1955). Since fossil fuels have been in the ground much longer than the <sup>14</sup>C lifetime, they are devoid of <sup>14</sup>C. When burned they add to the <sup>12</sup>C in the atmosphere without adding any <sup>14</sup>C, lowering  $\Delta^{14}$ C. Berry argues that the return of  $\Delta^{14}$ C to near zero, but still positive values, implies that the isotopic composition of new carbon added to the atmosphere matches that of oceanic carbon, not fossil fuel carbon which would take it negative. But  $\Delta^{14}$ C is not done changing. As evident from a close examination of Figure 1, since ~2000 it has been falling approximately linearly, not exponentially, as anthropogenic carbon accumulates in the atmosphere. It will assuredly go negative in the current decade, as predicted by Caldiera, et al. (1998), even as the concentration of <sup>14</sup>C continues to increase. We can take Berry's comment as a testable prediction that in his model,  $\Delta^{14}$ C will remain positive. Berry's forecast also contrasts with forecasts in a recent review on the topic by Graven, Keeling, and Rogeli (2019), which agree with Caldeiera. We are confident Berry's prediction will be falsified.

The most important feature of the <sup>14</sup>C concentration that needs to be explained is that, after initially falling quickly when atmospheric nuclear testing ended, it has since roughly stabilized 30% or more above the pre-1950 plateau that had persisted for centuries. Both Figures 1 and 2 show this. If the  $\sim$ 30% increase does not include lingering bomb carbon as it certainly appears to, what caused it? Berry does not address this obvious question and merely fits an exponential to the red curve for the years between 1970 and 1990. Unlike Berry, Harde and Salby recognize the need to address the baseline shift.

Harde and Salby's 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:

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

We will put this into more transparent units to compare with other studies. Since a  $\Delta^{14}$ C standard is 1.176 x 10<sup>-12</sup> (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) their <sup>14</sup>C concentration standard is 371.4 x 10<sup>-12</sup> ppm. Taking the total atmosphere to contain 1.77 x 10<sup>20</sup> moles, then this standard can also be expressed as 65.71 kmoles of <sup>14</sup>C. When the total atmosphere contains this much <sup>14</sup>C, its "( $\Delta^{14}$ C)<sub>c</sub>" = 0. The emission rate in (2) is then equivalent to:

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

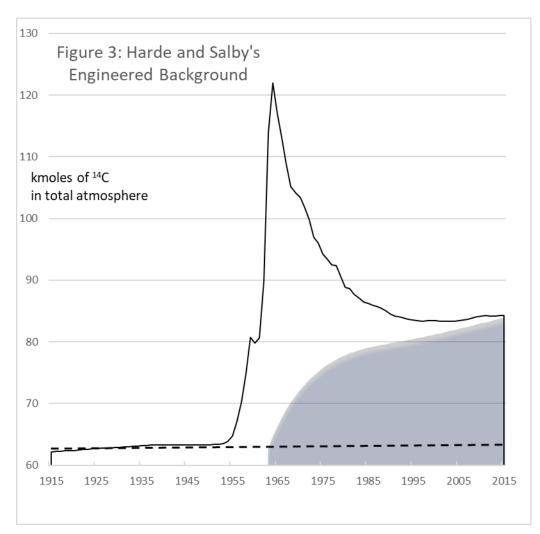
From Figure 1 we see that prior to 1950, the atmospheric abundance of <sup>14</sup>C was constant around 355 x  $10^{-12}$  ppm. If it is removed with an effective time constant of 10 years, as Harde and Salby argue, then its production rate needs to be 35.5 x  $10^{-12}$  ppm/year, or 6.28 kmoles/year. (The equilibrium level equals the time constant times the emission rate.) So Harde and Salby would say:

(4)  $e'_{NB,14(t)} = 6.28$  kmoles/yr valid before 1964 (or at least before 1950. But note that this value assumes their 10-year time constant.))

Figure 3 shows the now familiar bomb pulse concentration curve. This is the same data as in Figure 1, but with the axis now labeled in kmoles, 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 source(s) grow 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 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 need 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  $\Delta^{14}$ 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  $\Delta^{14}$ C and calibrate the age vs specific activity curve (Damon and Peristykh (2000)). Detailed plots of historical values of  $\Delta^{14}$ 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 propose a step change in cosmic ray flux close to 30 %. For this to have happened without being noticed elsewhere is unlikely to say the least. 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  $\Delta^{14}$ C, not fluctuations in concentration. But  $\Delta^{14}$ 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 not likely.

Note that Harde and Salby'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 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 as they have done.

In sharp contrast to Harde and Salby's approach of making unjustified background adjustments, Caldeira et al. (1998) were able to account for the <sup>14</sup>C concentration curve with no manipulations. Even better, they *predicted* the rise in atmospheric <sup>14</sup>C concentration after 2000. In their model, about half of the increase in atmospheric <sup>14</sup>C concentration since 1950 is from residual bomb carbon. The nuclear testing initially resulted in high <sup>14</sup>C/<sup>12</sup>C ratios in the atmosphere compared to the land and sea sinks. As the inventories in the fast carbon cycle mixed, both the isotope ratio and the <sup>14</sup>C concentration fell in the atmosphere and rose elsewhere. Anthropogenic carbon put into the atmosphere subsequently reversed the isotope ratio gradient via the Seuss effect, and the fast carbon cycle returned net <sup>14</sup>C from land and sea sinks back into the atmosphere. Successful predictions are persuasive.

#### **4** Discussion

While the rhetoric of these papers suggests they have "proofs" absolving humans of responsibility for atmospheric CO<sub>2</sub> increases, what the papers actually provide are inaccurate models of a mostly irrelevant quantity, the present concentration of atmospheric anthropogenic carbon. Both models complicate carbon accounting with their separation of human from natural carbon, but both are too simple to explain the <sup>14</sup>C data. Because <sup>14</sup>C lingers in the fast cycle longer than either of their models indicates, we can conclude that more anthropogenic carbon remains in the atmosphere than these papers estimate. From the fact that net global uptake is positive, we can conclude that the exact amount of human carbon in the atmosphere is of secondary importance in determining the cause of the increase.

Neither of these papers was written for a scientific audience. Scientists, skeptical by nature, are not fooled by engineered backgrounds. Both papers were written instead to influence lay opinion on an

important public policy issue. Both papers target readers who focus on results they want to believe, but who lack either the stamina or the tools to critically evaluate for themselves the arguments presented. Perhaps the most telling argument against these papers is this: when the  $\Delta^{14}$ C error was discovered and data effectively changed, their theories did not. Theories which are insensitive to data can only be described as ideological, not scientific.

Berry acknowledges a couple of prominent skeptics, perhaps suggesting that they endorse his model. Here is what they actually say:

Ralph Alexander: "However, it's an open-and-shut case to me. There's no question in my mind that Ed's [Berry's] model is falsified by the observations of <sup>14</sup>C and needs to be modified before it can be taken seriously." Scienceunderattack blog, October 19, 2020, The Scientific Method at Work: The Carbon Cycle Revisited.

William Happer: "[the rising CO2] appears to be mostly due to burning fossil fuels... that's the most logical explanation" How to Think About Climate Change, Clintel Lecture in Amsterdam, November 15 2021.

Those somehow motivated to challenge the consensus view that human activities are causing climate change should look elsewhere. That fossil fuel burning and other human activities are the direct cause of atmospheric  $CO_2$  increases is as settled as science gets.

#### Acknowledgements

The author received no financial support for this work and has no conflicts of interest. All data used in this paper is publicly available in the references cited.

## References

- Andrews, D.E.(2020) "Correcting an Error in Some Interpretations of Atmospheric <sup>14</sup>C", *Earth Sciences*. Vol. 9, No. 4, pp. 126-129. doi: 10.11648/j.earth.20200904.12.
- Ballantyne, A. P. Alden, C.B., Miller, J.B., Tans, P.P. (2012), "Increase in observed net carbon dioxide uptake by land and oceans during the past 50 years", *Nature*, vol 488 pp 70-72. doi:10.1038/nature11299
- Berry, E. X. (2019). "Human CO<sub>2</sub> emissions have little effect on atmospheric CO<sub>2</sub>." *International Journal* of Atmospheric and Oceanic Sciences, 3(1), 13-26. doi: 10.11648/j.ijaos.20190301.13
- Berry, E.X. (2021) "The Impact of Human CO<sub>2</sub> on Atmospheric CO<sub>2</sub>", *Science of Climate Change*, vol. 1, no.2, pp 1-46.
- Caldeira, K., Raul, G. H., and Duffy, P. B. (1998). "Predicted net efflux of radiocarbon from the ocean and increase in atmospheric radiocarbon content." *Geophysical Research Letters*, 25(20), 3811-3814.
- Cawley, G. C. (2011). "On the atmospheric residence time of anthropogenically sourced CO<sub>2</sub>." *Energy Fuels* 25, 5503–5513, <u>http://dx.doi.org/10.1021/ef200914u</u>.
- Ciais, P., C. Sabine, G. Bala, L. Bopp, V. Brovkin, J. Canadell, A. Chhabra, R. DeFries, J. Galloway, M. Heimann, C. Jones, C. Le Quéré, R.B. Myneni, S. Piao and P. Thornton, 2013: Carbon and Other Biogeochemical Cycles. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- Damon, P.E and Peristykh, A.N. (2000), "Radiocarbon Calibration and Application to Geophysics, Solar Physics, and Astrophysics", Radiocarbon Vol 42, Nr 1, pp 137-150.

- EIA. US Energy Information Administration. <u>http://www.eia.gov/energyexplained/energy-and-the-environment/greenhouse-gases-and-the-climate.php</u>. Accessed 20 January 2022.
- Essenhigh, R. H. (2009). "Potential dependence of global warming on the residence time (RT) in the atmosphere of anthropogenically sourced carbon dioxide". *Energy & Fuels*, 23, 2773–2784.
- Graven, H., Allison, C. E., Etheridge, D. M., Hammer, S., Keeling, R. F., Levin, I., et al. (2017)
  "Compiled records of carbon isotopes in atmospheric CO<sub>2</sub> for historical simulations in CMIP6," *Geosci. Model Dev.*, 10, 4405–4417, https://doi.org/10.5194/gmd-10-4405-2017.
- Graven, H., Keeling, R.F., & Rogelj, J. (2020) "Changes to carbon isotopes in atmospheric CO<sub>2</sub> over the industrial era and into the future" *Global Biochemical Cycles*, 34, e2019GB006170. https://doi.org/10.1029/2019GB006170
- Harde, H.(2017) "Scrutinizing the carbon cycle and CO<sub>2</sub> residence time in the atmosphere". *Global and Planetary Change*, 152,19-26.
- Harde, H. (2019) "What humans contribute to atmospheric CO2: Comparison of carbon cycle models with observations". *Earth Sciences*. 8(3), 139-159. doi: 10.11648/j.earth.20190803.13
- Harde H., and Salby,M "What Controls the Atmospheric CO<sub>2</sub> Level" (2021), *Science of Climate Change* vol. 1, no. 1, Aug 30, 2021 pp 54-69.
- Joos, F., R. Roth, J. S. Fuglestvedt, G. P. Peters, I. G. Enting, W. von Bloh, V. Brovkin, E. J. Burke, M. Eby, N. R. Edwards, T. Friedrich, T. L. Frölicher, P. R. Halloran, P. B. Holden, C. Jones, T. Kleinen, F. T. Mackenzie, K. Matsumoto, M. Meinshausen, G.-K. Plattner, A. Reisinger, J. Segschneider<sup>6</sup>, G. Shaffer, M. Steinacher, K. Strassmann, K. Tanaka, A. Timmermann, and A. J. Weaver(2013) "Carbon dioxide and climate impulse functions for the computation of greenhouse gas metrics: a multi-model analysis", *Atmospheric Chemistry and Physics*, vol 13, pp 2793-2825. doi:10.5194/acp-13-2793-2013.
- Levin, I. and Hesshaimer, V. (2000). "Radiocarbon: A unique tracer of global carbon cycle dynamics", *Radiocarbon*, 42(1), 69–80.
- Naegler, T, Levin, I. (2006) "Closing the global radiocarbon budget 1945–2005", *Journal of Geophysical Research* 111:D12311. doi: 10.1029/2005JD006758
- Richardson, M. (2013), "Comment of "The phase relation between atmospheric carbon dioxide and global temperature" by Humlum, Stordahl, and Solheim", *Global and Planetary Change*, vol 107, pp 226-228.
- Stenstrom, K. E., Skog, G., Gerogiadou, Genberg. J., Johansson, A. (2011). "A guide to radiocarbon units and calculations". Lund University, LUNFD6(NFFR-3111)/1-17/(2011). <u>https://www.hic.ch.ntu.edu.tw/AMS/A%20guide%20to%20radiocarbon%20units%20and%20cal</u> <u>culations.pdf</u>
- Stuiver, M. and Polach, H.(1977). Discussion: Reporting of <sup>14</sup>C data". *Radiocarbon*, 19(3), 355-363. https://journals.uair.arizona.edu/index.php/radiocarbon/article/viewFile/493/498
- Suess, H. E. (1955), "Radiocarbon concentration in modern wood", Science, 122, 415-417.
- Turnbull, J., Rayne, P., Miller, J., Naegler, T., Ciais, P., Cozic, A. (2009). "On the use of <sup>14</sup>CO<sub>2</sub> as a tracer for fossil fuel CO<sub>2</sub>: Quantifying uncertainties using an atmospheric transport model", *Journal of Geophysical Research*, 114, D22302. <u>https://doi.org/10.1029/2009JD012308</u>
- Zazzeri, G., Acuña Yeomans, E., & Graven, H. D. (2018). "Global and regional emissions of radiocarbon from nuclear power plants from 1972 to 2016". *Radiocarbon*, 60, 1067–1081. https://doi.org/10.1017/RDC.2018.42