## Comments on "Components of CO<sub>2</sub> in 1750 through 2018 Corrected for the Perturbation of the <sup>14</sup>CO<sub>2</sub> Bomb Spike"

## Dear Editors:

A paper in this journal analyzing atmospheric radiocarbon specific activity by Skrable et al. (February 2022) (hereinafter "Skrable 1") attracted multiple critical letters by atmospheric scientists and others. One of those letters by Schwartz et al. (2022) made four arguments against the paper, briefly summarized here as:

- 1. Use of erroneous data on atmospheric  $CO_2$  history;
- 2. Neglect of the "bomb carbon;"
- 3. Failure to account for isotope exchange between the atmosphere and other reservoirs; and
- 4. Neglect of other lines of evidence that fossil fuel emissions are the source of atmospheric CO<sub>2</sub> rise.

In a new letter, which references a revised paper contained in accompanying supplemental data, Skrable et al. (November 2022) (hereinafter "Skrable 2"), the authors address points 1 and 2. They recompute what they call the "fossil component" of atmospheric carbon with "no bomb scenario" inputs modeled by Graven et al. (2020). Getting similar results to their first analysis, Skrable 2 doubles down on the sweeping conclusions of Skrable 1, but they have not addressed in any meaningful way points 3 and 4. It is not the purpose of this letter to mount new arguments against these papers, as none are necessary. But since evidently Skrable, Chabot, and French have not yet appreciated points 3 and 4, it does appear necessary to spell out in detail their explicit relevance to the Skrable 1 and 2 analyses.

The key equation derived in both Skrable 1 and 2 is a specific activity dilution formula:

$$C_F(t) = C(0) \left[ \frac{S(0)}{S(t)} - 1 \right].$$

This formula correctly gives the amount of cold carbon  $C_F(t)$  which, if added to a quantity C(0) having specific activity S(0), would yield a mixture having specific activity S(t). Using this formula, Skrable 2 argues correctly that the 2018 atmosphere cannot be a simple mixture of 276.44 ppm of

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S(0) carbon (1750 values) and 128.96 ppm (the CO<sub>2</sub> rise from 1750 to 2018) of cold carbon because the present value of S(t) is too high. That much cold carbon in the atmosphere would have diluted the specific activity more. So far, so good. But the authors then wrongly jump to the conclusion that the small value of their calculated "fossil component" implies anthropogenic emissions are "much too low to be the cause of global warming." Before making this erroneous statement, they should have thought more about points 3 and 4.

Although the authors acknowledge the existence of continuous two-way carbon exchanges between the atmosphere and land/sea reservoirs, the subject of Point 3, in their response published with the Schwartz et al. letter, they assert with no justification that "exchange among the reservoirs was inconsequential for our work." The effects of exchanges on specific activity are in fact very consequential to their work and easily understood. Since fractionation effects are minor, the exchanges to good approximation simply mix atmospheric carbon with land/sea inventories. Therefore, if something causes an isotope ratio (specific activity) in one reservoir to temporarily differ from its value in another, then mixing from the two-way exchanges will tend over time to erase the difference. Even when no net carbon is exchanged, net <sup>14</sup>C will be transferred from the higher specific activity reservoir to the lower one because of the mixing. Specialists call these transfers "isofluxes." They are illustrated by the bomb pulse.

In 1964, the specific activity of atmospheric carbon was nearly twice that of the oceans because nuclear testing had left additional <sup>14</sup>C in the atmosphere. But after atmospheric testing ceased, specific activities in different reservoirs converged toward similar numbers because atmospheric and land/sea inventories mixed. Plots of the bomb pulse specific activity, for example Fig. 1 of Schwartz et al. (2022), indicate a time constant for the mixing of roughly a decade. In like manner, specific activity deficits in the atmosphere caused by injections of fossil fuel carbon are reduced by isofluxes of <sup>14</sup>C flowing toward the (lower specific activity) atmosphere. The net effect of mixing is therefore to oppose the decline of S(t) resulting from emission of cold fossil fuel CO2. The measured Suess effect (specific activity dilution by cold carbon) is smaller than it would have been without carbon exchanges. This was noted by Seuss himself: "The decrease [in specific activity] can be attributed to the introduction of a certain amount of  $C^{14}$ -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 Seuss began the rich history of using radiocarbon to learn about atmospheric processes when noting "the rate by which this  $CO_2$  exchange must be greater than previously assumed" (Seuss 1955). A modern quantitative analysis of the qualitative argument given here can be found in Graven et al. (2020). Without carefully considering exchanges, Graven et al. would not have been able to untangle the opposing effects of specific activity increasing bomb carbon, specific activity decreasing fossil fuel emissions, and the associated isofluxes to arrive at the "no bomb scenario" plot used in Skrable 2's reanalysis.

Table 2 in the Skrable 2 supplement contains all the data necessary to further discuss Schwartz et al.'s Point 4. In addition to presenting C(t) in ppm from 1750 to 2018, it presents DE(t), anthropogenic CO<sub>2</sub> emissions, in units of billion metric tons (GTons of CO<sub>2</sub>) per year. DE(t) is not used in the paper's calculation of  $C_F(t)$ , even though the magnitude of emissions is obviously relevant to atmospheric CO<sub>2</sub> growth. By inexplicably failing to express C(t) and DE(t) in similar units, Skrable 1 and 2 miss important information. The conversion factor between ppm and total atmospheric CO<sub>2</sub> mass is 7.8 GT CO<sub>2</sub>/ppm, based on a well-mixed dry atmosphere containing  $1.76 \times 10^{20}$  moles of gas. (This conversion factor should not be confused with the 2.124 GT C/ppm factor, which yields the total mass of atmospheric carbon rather than CO<sub>2</sub>.) Then the following table can be constructed wherein quantities expressed in ppm in Skrable 2's Table 2 are converted to GTons of CO<sub>2</sub> (Table 1).

Table 1. quantities expressed in ppm in Skrable 2's Table 2 are converted to GTons of  $\mathrm{CO}_2$ .

Quantity	ppm	GTons CO2
Increase in atmospheric CO <sub>2</sub> (1750–2018)	128.96	1,006
Skrable 2 "Fossil component" of increase	41.28	322
Skrable 2 "Non-fossil component" of increase	87.68	684
Cumulative emissions (1750–2018)		1,590
Cumulative emissions (2009–2018)		346

We note the following:

1. Cumulative emissions during the Industrial Era (1,590 GTons) substantially exceed the increase in atmospheric CO<sub>2</sub> (1,006 GTons), a well-known fact cited by Schwartz et al. and confirmed by Skrable 2's data if not by its conclusions. This implies that during the Industrial Era, the ocean and land reservoirs have together been net sinks of carbon, not sources, leaving emissions as the source of the rise. Perhaps no other fact is as compelling in demonstrating human responsibility for atmospheric CO<sub>2</sub> growth. Ocean acidification and some stimulus of terrestrial plant growth are consequences of the "overflow" of anthropogenic carbon out of the atmosphere. Any model such as the one presented in Skrable 2 that concludes that human emissions have played a minor role in the rise in atmospheric  $CO_2$  during the Industrial Era is falsified by this observation.

2. Skrable 2's estimate of a "fossil component" is but 20% of total 1750-2018 emissions, or roughly the cumulative emissions between 2009 and 2018. This result can be understood with a crude estimate. Recall that a decade was about the mixing time observed with the bomb pulse. We can then expect the magnitude of the dilution of atmospheric specific activity by cold fossil fuel carbon to be dominated by the most recent decade's emissions. The dilution from earlier decades' emissions has largely been erased by mixing. Therefore, an analysis that naively infers a "fossil component" from the observed dilution may be expected to return a number approximating one decade's worth of emissions, and Skrable 2 does just that. Their 321 GTon "fossil component" is close to the 346 GTon's of CO<sub>2</sub> estimated to have been emitted between 2009 and 2018. Because of this dynamic situation, the very notion of "fossil" and "non-fossil" atmospheric components is not useful, accounting for the inability of the authors to find tabulations of these quantities in the peer-reviewed literature.

Fossil fuel emissions more than account for the entire rise in atmospheric  $CO_2$ , but only a small fraction of the cold carbon added to the carbon cycle since 1750 remains in the atmosphere. One could summarize: "What happens in the atmosphere does not stay in the atmosphere."

Like Skrable, Chabot, and French, the author of this letter is concerned that important public policy decisions be based on sound science. The advantage of the scientific method over other approaches to obtaining knowledge is in its ability to use empirical evidence to cull innovative and creative but wrong ideas. In most instances, peerreview brings broadened viewpoints that expose weaknesses in arguments early in the communication process. Peer-review failed the authors in this case, leaving them with the unpleasant responsibility of making things right after publication. We thank them for acknowledging data errors in Skrable 1. But the conceptual errors clearly pointed out by Schwartz et al. and detailed further here remain in Skrable 2, nullifying their conclusions. Health Physics, along with Skrable, Chabot, and French, needs to decide whether to continue calling the issues discussed here "controversial" or acknowledge that human responsibility for atmospheric CO<sub>2</sub> rise during the Industrial Era is as settled as science gets.

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