Comment on "World Atmospheric CO_2, Its ^{14}C Specific Activity, Non-fossil Component, Anthropogenic Fossil Component, and Emissions (1750–2018)," by Kenneth Skrable, George Chabot, and Clayton French

Dear Editors:

Recently Skrable et al. (2022) published a study based on analysis of $^{14}CO_2$ (radiocarbon) in the atmosphere that concludes that CO_2 emitted from fossil-fuel combustion, which contains no $^{14}CO_2$, contributes only 36% to the observed increase in atmospheric CO_2 over the industrial era, with the balance due to non-fossil-fuel sources. In this Comment we demonstrate that the paper by Skrable et al. is fundamentally flawed in at least four respects: (1) erroneous history of $^{14}CO_2$ in air that is at odds with direct observations; (2) neglect of the consequences of the large input of $^{14}CO_2$ into the atmosphere from nuclear weapons tests in the 1950s and 1960s; (3) failure to account for isotope exchanges between the atmosphere, ocean, and land biosphere that occur independent of net change in amount of atmospheric CO_2; and (4) neglect of multiple independent lines of evidence that CO_2 emitted from fossil-fuel combustion is the principal contributor to the increase of atmospheric CO_2 over the industrial era. We detail these flaws here and conclude that the paper of Skrable et al. should be retracted in its entirety.

Erroneous history of $^{14}CO_2$ in air that is at odds with direct observations

The amount of atmospheric $^{14}CO_2$ in the industrial period as presented by Skrable et al. and as determined from dendrochronologically dated wood samples and by direct measurements are compared in Fig. 1. Here the two vertical axes represent this amount as specific activity (activity of a sample per amount of total carbon in the sample, as employed by Skrable et al. 2022) and as decay- and fractionation-corrected enrichment relative to a standard, as generally reported by the atmospheric science community (Stuiver and Polach 1977); the two measures are linearly related (Levin et al. 2010). The specific activity given by Skrable et al. at the beginning of the record (1750), denoted by the square brown marker in Fig. 1, is erroneously high, mainly because of their use of an outdated and erroneous value for the cosmogenic production rate of ^{14}C in Earth's atmosphere. The several values of specific activity given by Skrable et al. toward the end of the record (2002–2012), shown in the figure by round brown markers, were obtained from contemporaneous measurements in air. The intermediate and subsequent values were obtained by an interpolation formula. It is clear that the historical specific activity given by Skrable et al. is completely at odds with the measurements over virtually the entire record and thus cannot be relied upon in apportionment of the source of the increase of atmospheric CO_2 over the industrial period to fossil-fuel and non-fossil-fuel sources or for any other purpose.

Neglect of the consequences of the large input of $^{14}CO_2$ into the atmosphere from nuclear weapons tests in the 1950s and 1960s

The large increase in atmospheric $^{14}CO_2$ from nuclear weapons testing (so-called bomb $^{14}CO_2$) is the most prominent feature in the measurement record of atmospheric $^{14}CO_2$ and has been widely documented from numerous observational locations by multiple research groups around the world (Fig. 1). Although the source of $^{14}CO_2$ from weapons tests ceased almost entirely in 1964 on account of the test ban treaty, the signal of bomb $^{14}CO_2$ in atmospheric $^{14}CO_2$ has persisted substantially to the present time. Although Skrable et al. (2022) expressed cognizance of the input of $^{14}CO_2$ into the atmosphere from weapons testing, they state, based on a citation to Wikipedia, that $^{14}CO_2$ from the atmospheric bomb tests would be significant only to about 2005 and therefore that no correction to their apportionment anthropogenic fossil and non-fossil components of CO_2 would be required for measurements subsequent to 2005. The contribution of bomb $^{14}CO_2$ to atmospheric $^{14}CO_2$ was explicitly examined in a modeling study (Graven et al. 2020) in which the source of bomb $^{14}CO_2$ was omitted, shown also in Fig. 1; the signal of bomb $^{14}CO_2$ is given by the difference between the measurements and the model in the absence of the bomb $^{14}CO_2$ source. This difference shows that residual bomb $^{14}CO_2$ cannot be dismissed over the years 2004–2012 used by Skrable et al. in their analysis. Failure to include this residual bomb $^{14}CO_2$ in their apportionment of the increase of the increase of atmospheric CO_2 to fossil fuel and non-fossil fuel sources completely vitiates this apportionment.

Failure to account for isotope exchanges between the atmosphere, ocean, and land biosphere that occur independent of net change in amount of atmospheric CO_2

The Skrable et al. (2022) framework assumes that carbon in the environment can be divided into two categories: (1)

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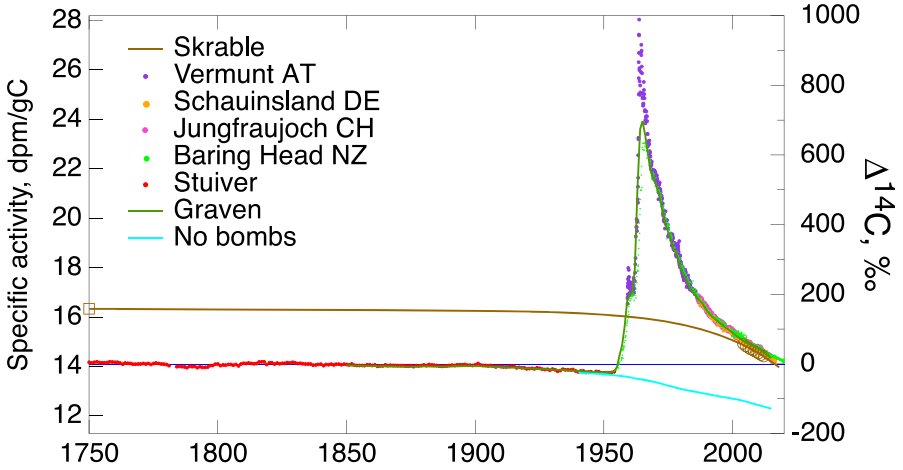

Fig. 1. Specific activity of atmospheric CO_2, disintegrations per minute left axis and corresponding decay- and fractionation-corrected isotopic enrichment of ^{14}C, per mil (‰), right axis. Brown curve shows specific activity as given by Skrable et al. (2022); square brown marker denotes value used by those investigators as initial value for industrial period; round brown markers denote measurements of $^{14}CO_2$ in air also used by those investigators. Brown curve denotes interpolation formula. Points denote enrichment of atmospheric $\Delta^{14}CO_2$ as determined from direct atmospheric samples and dendrochronologically aged wood samples and corresponding specific activity (Stuiver et al. 1998; Levin et al. 1985, 2013; Turnbull et al. 2017). Green curve denotes composite $\Delta^{14}CO_2$ (and corresponding specific activity) as compiled by Graven (2017). Cyan curve shows specific activity and enrichment calculated from atmospheric $^{14}CO_2$ as modeled in the absence of the perturbation due to nuclear weapons testing, so-called no-bombs scenario (Graven et al. 2020). Blue line denotes preindustrial specific activity corresponding to $\Delta^{14}C$ = 1.2 ‰ and CO_2 mixing ratio 278 ppm.

preindustrial carbon, which is taken as uniform in the $^{14}C/^{12}C$ ratio, and (2) fossil carbon, which is devoid of radiocarbon. They further assume that these categories maintain their identities as carbon is exchanged between the atmosphere, ocean, and land biosphere. In fact, the flows of different carbon isotopes are not connected as assumed by Skrable et al. Each carbon atom and isotope is exchanged independently. Thus, importantly here, ^{14}C can be exchanged between the atmosphere and ocean with no net exchange of carbon as a whole. An important consequence of this independence is that impact of emissions of fossil-fuel CO_2 on the $^{14}C/^{12}C$ ratio of atmospheric CO_2 is much greater than the impact of these emissions on atmospheric CO_2 as a whole. Dilution of atmospheric CO_2 by the ^{14}C-free fossil-fuel carbon is therefore not a straightforward proxy for the impact of fossil-fuel on the buildup of atmospheric CO_2. Correct handling of these independent exchanges requires information on the sizes and exchange rates between different carbon reservoirs, as has been clear since the 1950s (Bolin and Eriksson 1959). Additionally, the isotopic ratio of preindustrial carbon is not uniform, with older reservoirs such as the carbon in the deep ocean having lower $^{14}C/^{12}C$ ratio. These differences are the basis of radiocarbon dating. These critical aspects were not considered by Skrable et al., causing their calculations to underestimate the input of fossil-fuel CO_2.

Neglect of multiple independent lines of evidence that CO_2 emitted from fossil-fuel combustion is the principal contributor to the increase of atmospheric CO_2 over the industrial era

Finally, the present understanding of the controls on atmospheric CO_2 buildup importantly rests on many convergent strands of evidence in addition to radiocarbon. From the well quantified rates at which CO_2 is building up in the atmosphere and rates of CO_2 release from fossil-fuel burning, it is clear that around 50% of the emitted carbon remains in the atmosphere, with the balance absorbed by other reservoirs, of which the oceans and the land biosphere are the most important. The ocean and land biosphere are thus together acting as a major sink not a source of CO_2 (Denning 2022). The excess CO_2 in the ocean is now also well measured, and the uptake rate is consistent with a wide body of other evidence on rates of mixing and carbon chemical properties of seawater. This understanding of the rate at which excess carbon is being redistributed into the ocean and land is independently supported by measurements of trends in atmospheric O_2 and $^{13}C/^{12}C$ ratio in addition to radiocarbon (Keeling and Graven 2021).

On the basis of the several arguments presented here, we conclude that the paper of Skrable et al. (2022) should be retracted in its entirety.

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REFERENCES

Bolin B, Eriksson E. Changes in the carbon dioxide content of the atmosphere and sea due to fossil fuel combustion. In: Bolin B, ed. The atmosphere and the sea in motion (Scientific contributions to the Rossby memorial volume). New York: Rockefeller Institute Press in association with Oxford University Press. 1959; 30–142.

Denning AS. Where has all the carbon gone? Annual Rev Earth Planetary Sci 50:55–78; 2022.

Graven H, Allison CE, Etheridge DM, Hammer S, Keeling RF, Levin I, Meijer HA, Rubino M, Tans PP, Trudinger CM, Vaughn BH. Compiled records of carbon isotopes in atmospheric CO_2 for historical simulations in CMIP6. Geosci Model Dev 10: 4405–4417; 2017. DOI:org/10.5194/gmd-10-4405-2017.

Graven H, Keeling RF, Rogelj J. Changes to carbon isotopes in atmospheric CO_2 over the industrial era and into the future. Global Biogeochem Cycles 34(11):1–21; 2020.

Keeling RF, Graven HD. Insights from time series of atmospheric carbon dioxide and related tracers. Annual Rev Environ Res 46:85–110; 2021.

Levin I, Kromer B, Schoch-Fischer H, Bruns M, Münnich M, Berdau D, Vogel JC, Münnich KO. 25 years of tropospheric 14C observations in central Europe. Radiocarbon, 27: 1–19; 1985.

Levin I, Naegler T, Kromer B, Diehl M, Francey R, Gomez-Pelaez A, Steele P, Wagenbach D, Weller R, Worthy D. Observations and modelling of the global distribution and long-term trend of atmospheric $^{14}CO_2$. Tellus B: Chem Physical Meteorol 62: 26–46; 2010.

Levin I, Kromer B, Hammer S. Atmospheric $\Delta^{14}CO_2$ trend in Western European background air from 2000 to 2012. Tellus B: Chem Physical Meteorol 65:20092; 2013.

Skrable K, Chabot G, French C. World atmospheric CO_2, its ^{14}C specific activity, non-fossil component, anthropogenic fossil component, and emissions (1750–2018). Health Phys 122:291–305; 2022.

Stuiver M, Polach HA. Discussion. Reporting of ^{14}C data. Radiocarbon 19:355–363; 1977.

Stuiver M, Reimer PJ, Braziunas TF. High-precision radiocarbon age calibration for terrestrial and marine samples. Radiocarbon 40:1127–1151; 1998.

Turnbull JC, Mikaloff Fletcher SE, Ansell I, Brailsford GW, Moss RC, Norris MW, Steinkamp K. Sixty years of radiocarbon dioxide measurements at Wellington, New Zealand: 1954–2014. Atmospher Chem Phys 17:14771–14784; 10.5194/acp-17-14771-2017; 2017.

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