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## Reply on RC1

Stephen E. Schwartz

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Author comment on "Observation Based Budget and Lifetime of Excess Atmospheric Carbon Dioxide" by Stephen E. Schwartz, Atmos. Chem. Phys. Discuss.,  
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## Response to Reviewer

I thank Dr. Archer for his signed review. The paper is indeed long, but I would disagree with Dr. Archer's assessment that the findings are "buried under many pages of scholarship." Because I reached a conclusion that differs substantially from the picture that has evolved from many prior studies, I felt compelled to provide chapter and verse for how I obtained my results. Consequently, I endeavored to make my approach clear and transparent, with much of the "scholarship" in Appendices. That said, however, if my study is fundamentally wrong, the journal wants to know in what way, and I want to know that too.

In brief, I believe all of the concerns raised by Dr. Archer are rebuttable and I present those rebuttals here. I also refer Dr. Archer and others to Appendix F of the manuscript, where some of the concerns raised by Dr. Archer are addressed more fully.

First I would like to address the concern over the definition of what constitutes the lifetime of a substance in a reservoir, and more specifically of excess CO<sub>2</sub> in the atmosphere, where excess refers to the amount above preindustrial. In my study I adopted what I consider the very reasonable qualitative definition of lifetime (more specifically, *adjustment* time) advanced by Archer et al. in their 2009 paper in *Annual Reviews*, paraphrased slightly here, as the time that would be required, following cessation of anthropogenic emissions, for the amount of CO<sub>2</sub> in the atmosphere to decrease substantially to its preindustrial value. As the influences of excess CO<sub>2</sub> on Earth's radiation budget or on ocean acidity that trigger concern over excess CO<sub>2</sub> would scale more or less with the amount of excess CO<sub>2</sub>, the time scale over which this excess CO<sub>2</sub> would decrease substantially is the time scale of primary importance. Determination of this adjustment time was a major objective of my study.

To briefly recapitulate my study: In this study I developed a budget for anthropogenic CO<sub>2</sub>, extending and updating the budget that has been used in several IPCC reports and which goes back to Sarmiento and Gruber's 2002 article in *Physics Today* (all references in the manuscript). Based on the stocks and fluxes in this budget I defined and evaluated a *turnover* time for excess CO<sub>2</sub> that takes into account rapid exchange between the atmosphere and the mixed-layer (ML) ocean and between the atmosphere and the labile biosphere (LB). This budget permitted evaluation of transfer coefficients describing the rate of transport of carbon between the several compartments; so far as possible the

transfer coefficients were developed using information that was not specific to CO<sub>2</sub>; for example transfer coefficients between the ML and the deep ocean (DO) are based on the piston velocity describing the rate of heat transport between these compartments. Using these transport coefficients I constructed a simple numerical model that accurately reproduces atmospheric CO<sub>2</sub> mixing ratio over the Anthropocene and atmospheric and oceanic radiocarbon over the Anthropocene including the abrupt increase and subsequent decrease of production associated with atmospheric weapons testing and the abrupt cessation of those emissions. This model was then used to evaluate the rate of decrease of excess CO<sub>2</sub> following a hypothetical cessation of emissions and from this decay rate to evaluate the *adjustment time* of excess CO<sub>2</sub>. Multiple measures of the adjustment time exhibit close agreement with each other and with the turnover time. This agreement is essential to lend confidence in the understanding and numerical representation of the transfer rates governing evolution of the system.

As Dr. Archer points out, and as found in numerous prior studies including Archer et al. (2009) and in my study, subsequent to cessation of emissions and the relatively rapid decrease of excess CO<sub>2</sub>, there would be a tail of excess CO<sub>2</sub> that would persist for a time much greater than the time scale of substantial reduction. Such a long tail would, depending on averaging time, increasingly influence the average lifetime of excess CO<sub>2</sub>. The concern over an ever-increasing average lifetime of excess CO<sub>2</sub> that results from this long tail has led some investigators, e.g., Tans (1997) to assert that "The CO<sub>2</sub> lifetime concept should be banished." That is why, in Section 2 of the manuscript, I introduced several quantitative measures of the adjustment time that would characterize the time scale of substantial reduction advocated by Archer et al. (2009). These several measures of lifetime are compared in Fig. 12 of the manuscript. In particular in Fig. 12b it is seen that these measures of lifetime abruptly increase when, for the parameters of that example, the fraction of excess CO<sub>2</sub> remaining in the atmosphere has been reduced by about 85% from its value at the time of cessation. Surely the lifetime characterizing this removal of this fraction of the initial excess CO<sub>2</sub> meets the criterion of the time scale of substantial removal advocated by Archer et al. (2008).

Dr. Archer takes me to task for having "tossed out" the long tail of excess CO<sub>2</sub> that can persist for 5 kyr. In rebuttal, I would simply state that in evaluating the lifetime of excess CO<sub>2</sub> that characterizes the substantial decrease, the persistent tail of 15-20% at 700 years subsequent to cessation, shown for several pairs of the model parameters  $S_{pi}^l$  and  $b$  (carbon stock in the preindustrial labile biosphere and fertilization exponent, respectively) is not pertinent to the time scale characterizing the rate of decrease of the first 80 to 85%.

I turn now to some specifics raised in the Review.

**Paragraph 1** of the Review states: "If one takes the excess carbon from fossil fuels in the atmosphere, about 200 Gtons, and divides it by the ocean uptake rate of carbon, about 2 Gton / yr, one arrives at the erroneous result that the fossil carbon will go away in 100 years," and goes on to state "This result, analogous to that presented in this paper, is wrong because as the carbon invades the ocean, it alters the buffer chemistry of the ocean, and hence further uptake of CO<sub>2</sub>, by depleting CO<sub>3</sub>(<sup>2-</sup>) ion."

I concur with the Reviewer that a lifetime of excess CO<sub>2</sub> evaluated in this way would be an erroneous measure of lifetime, but not solely for the reason given by the Reviewer.

First, however, I emphatically state in my manuscript I did not use the measure of lifetime presented by the Reviewer. In the manuscript much attention was paid to identifying the leaving compartments and leaving fluxes that best represent the turnover time, taking into account near equilibration of the atmosphere (AC) and the ML ocean and near steady state between the AC and the labile biosphere (LB). Because of these close couplings the

turnover time pertinent to excess CO<sub>2</sub> was defined (Eq 5.7) as the quotient of the sum of the stocks in these compartments upon the sum of the net fluxes into the deep ocean and the obdurate biosphere, both of which exhibit return times to the AC + ML + LB that are long compared to the equilibration/steady-state times. It is the turnover time evaluated in this way for present stocks and fluxes (Eq 5.8) that was shown to be essentially the same as the adjustment time, defined above, all of which are compared in Fig. 12a.

The quotient of the present-day excess stock in the AC as given by the budget in my paper, Fig. 2, as 269 Pg, upon the net leaving flux into the ocean, 2.2 Pg yr<sup>-1</sup>, the measure of the turnover time cited by the Reviewer as yielding an erroneous measure of lifetime, is 122 yr. This value is coincidentally about the same as the turnover time evaluated as described above and other measures of the lifetime shown in Fig. 12. However I underscore that I did not evaluate the turnover time in this way.

The Reviewer goes on to state that evaluation of the turnover time in the manner he describes would be erroneous, not for the reasons just outlined but because "as the carbon invades the ocean, it alters the buffer chemistry of the ocean, and hence further uptake of CO<sub>2</sub>, by depleting CO<sub>3</sub>(<sup>2-</sup>) ion." To be sure the buffer chemistry of the ocean plays a role in the dynamics of the system, but the turnover time evaluated from the budget is based on stocks and fluxes; consequently although the turnover time is affected by the buffer chemistry, evaluation of the turnover time from the budget need not and should not take cognizance of the buffer chemistry. In contrast, the decay rate of excess CO<sub>2</sub> subsequent to cessation of emissions and the adjustment time evaluated from this decay rate are influenced by the buffer chemistry, as outlined below, but this influence is rather small, and in any event is explicitly accounted for in the model developed and exercised in this work.

Briefly, as the Reviewer notes and as is well known, uptake of CO<sub>2</sub> by the ocean over the Anthropocene is altered from that at preindustrial (PI) times by shift of the equilibrium chemistry in the CO<sub>2</sub>-bicarbonate-carbonate system that reduces the equilibrium solubility of CO<sub>2</sub> and that affects the uptake of CO<sub>2</sub> by the world ocean. There are two contributions to the net atmosphere-ocean flux: that due to increasing concentration of dissolved inorganic carbon (DIC) in the ML itself, and that due to the flux from the ML to the DO, Fig. 2. Because of near equilibrium between the AC and the ML the net fractional rates of both these processes depend on the differential equilibrium constant  $K'_{ma}$  (Appendix B, Eq B15). The increase of  $K'_{ma}$  over the Anthropocene due to increasing CO<sub>2</sub> (Fig. B1) results in a 30% decrease in the fractional net removal rate of anthropogenic DIC from the AC + ML compartment into the DO. Subsequent to cessation of emissions these fractional transfer rates would return nearly to their PI values. The effects of changing solubility as a function of atmospheric CO<sub>2</sub> (or of DIC concentration) are explicitly represented in the set of differential equations that constitute the numerical model used to calculate the time evolution of CO<sub>2</sub> throughout the time period examined and specifically after hypothetical cessation of emissions used to calculate the decay rate and adjustment time. These calculations make use of the equilibrium constant (and differential equilibrium constant) between atmospheric and ML stocks calculated off-line at every ppm of atmospheric CO<sub>2</sub> by the widely used program of Lewis and Wallace (1998). Solution of the differential equations evaluates these quantities by interpolation, and calculates the net flux from the partial pressure difference, thus explicitly taking cognizance of the buffer chemistry.

The Reviewer goes on to state "Complete drawdown of the CO<sub>2</sub> awaits first restoration of ocean pH by CaCO<sub>3</sub> balance, and ultimately CO<sub>2</sub> fluxes from volcanic emissions and weathering of silicate rocks." Here I would simply reiterate that the focus of this study is not on complete drawdown of excess CO<sub>2</sub>, but only on the time period of substantial removal of excess CO<sub>2</sub>, which time scale is much shorter than that influenced by volcanic emissions and weathering of silicate rocks.

In **paragraph 2** the Reviewer states " The time scale ( $\tau$ ) estimate one gets from the analysis presented in this paper, e.g. equations 2.1, 2.2, 5.1, 5.5, essentially fits an exponential to the present-day slope and extrapolates it forward, with no awareness of changes in ocean chemistry."

In response, I note first that all these equations deal with *turnover time* (quotient of stocks over fluxes, as described above) and as such are instantaneous properties of the system at any time and do not characterize the rate of decay of CO<sub>2</sub> subsequent to cessation of emissions (exponential or not), which rate of decay is characterized by the *adjustment time*.

The reviewer goes on to state "The real expected curve diverges from the exponential." Clearly this sentence is referring to the model calculations subsequent to cessation of emissions. The Reviewer then elaborates on the role of buffer chemistry.

Regarding decay of excess CO<sub>2</sub> subsequent to abrupt cessation of emissions in the model developed in this study several approaches are introduced to determine the *adjustment time*. In all of these the adjustment time is obtained from the rate of decay of excess CO<sub>2</sub> subsequent to abrupt cessation of anthropogenic emissions as calculated using the numerical model developed as described above and detailed in the manuscript. Evaluation of this rate of decay in the model, and indeed of the stocks of excess atmospheric CO<sub>2</sub> and other stocks and their rates of change, takes full cognizance of the buffer chemistry, as described above.

The Reviewer goes on to state that departure from exponential decay may be expected by future changes in ocean chemistry arguing that "future changes in ocean chemistry are larger than those that have already happened." Here I would note that subsequent to the abrupt cessation of emissions examined here, atmospheric CO<sub>2</sub> would decrease toward its preindustrial value, and hence that changes in ocean chemistry relative to preindustrial would be smaller than the present change, not larger.

This close conformance of the decay curves to an exponential function is manifested by the nearly straight lines of several decay curves on a semi-log plot, Fig. 1, over the initial 100 years subsequent to cessation, although some departure from exponential may be noted by the upward curvature in these plots. This close conformance to exponential decay is seen also in Fig. 12a. The close conformance to exponential decay results essentially from net flux out being proportional to stock with (nearly) constant coefficients; departure from exponential can come from non constancy of the transfer coefficient  $k'_{ma}$ , which decreases as the amount of DIC in the ML decreases, so that would lead to super-exponential decay) and from increase in return flux to the leaving compartment as the system approaches the preindustrial condition. Here the latter dominates the latter dominates, as is manifested by the upward curvature of the curves on the semi-log plots. It should be emphasized that exponential decay is not imposed on the system; the near-exponential decays results simply from solution of the set of (slightly nonlinear) differential equations.

One of the approaches to determining the adjustment time fits a decaying exponential (with baseline taken as preindustrial CO<sub>2</sub> mixing ratio) to the decay curve of CO<sub>2</sub> mixing ratio as calculated with the model to obtain the time constant of the exponential fit to the decay curve, denoted (line 1075 of the manuscript) as  $\tau_{CO_2fit}$ . This comparison to exponential decay should not be interpreted as imposing an exponential decay upon excess CO<sub>2</sub>. The close conformance of the decay curve to an exponential function, over the first 100 years subsequent to cessation, during which time atmospheric CO<sub>2</sub> has decayed to about 40% of its initial value, Fig. 12, is not imposed on the model but rather is a result of the model calculation yielding a nearly exponential decrease over this time

period. The measure of the adjustment time obtained in this way agrees closely with other measures of the adjustment time and with the turnover time suitably defined, Fig. 12a.

The Reviewer concludes his second paragraph by characterizing the use of a minimal model in this study as "willful ignorance," indicating that the more complex carbon models to which the present model is compared are "underlain by simple thermodynamics and mass balance and can't be dismissed as due to the whims of complicated, chaotic, uncertain models." In response I would note that the model developed here is indeed built on thermodynamics and mass balance, as detailed in the derivation of the transfer coefficients as backed up by Appendices B and A, respectively.

**Paragraph 3** of the Review deals again with the long small tail that would remain subsequent to cessation of CO<sub>2</sub> emissions. The reviewer erroneously states that the "the long tail doesn't arise in the calculations presented in the paper, so I wonder why it is acknowledged in the abstract as at least real."

In response, the floor that is reached in the model calculations at time scale greater than 300-400 years is discussed in Section 7.9 and shown explicitly in Fig. 16 and implicitly in Fig. 12b.

The Reviewer goes on to assert that the 5 kyr time scale of the tail is "tossed out." As discussed above this time scale is well beyond the time scale over which the CO<sub>2</sub> substantially decreases to its preindustrial value and is thus beyond the scope of this paper.

**Paragraph 4** elaborates on the impact of the long small tail.

In response, without going into the premises of the Reviewer's concerns (band saturation of CO<sub>2</sub> absorption, impacts of long-duration forcing), I would simply reiterate that the long tail and its effects are beyond the scope of this paper.

**Paragraph 5** of the Review takes issue with the statement in the Abstract that speaks of "substantial recovery of CO<sub>2</sub> toward its preindustrial value in less than a century" in the absence of anthropogenic emissions. This language rests on the three decay curves presented in Fig. 1 that bound the region in parameter space that is consistent with observations, best estimate reduction at 100 years after cessation, 59%; range 51% - 67%. Such reduction would certainly be substantial by any fair reading of the word "substantial." So it would seem that the disagreement with the statement in the Abstract reflects disagreement with the finding, which is based on the model calculations, not on the presence of the statement in the Abstract. I have presented in the manuscript full detail of the calculations and on determination of the transport coefficients employed in the model. The Reviewer has raised concerns over the approach taken in this study, but I feel that these concerns have been addressed in this Response. I thus stand by the statement given in the Abstract.

The Reviewer concludes by recommending against publication because he believes "it will just sow confusion." It would seem that the bases for this recommendation are 1) disagreement with definition of lifetime (adjustment time) as a measure of the time period of substantial reduction of excess CO<sub>2</sub>; 2) lack of inclusion of the long tail and long-time processes in evaluation of the lifetime; 3) concern over definition of turnover time; 4) concern that exponential decay is imposed on the modeled decay; and 5) concern that buffer chemistry and thermodynamics are not included in the model. I would hope that this Response to Reviewer has adequately resolved these concerns. This manuscript, presenting the budget of excess carbon in the several reservoirs, determination of transfer coefficients from that budget, inference of the turnover time of excess carbon, development of a numerical model accurately describing evolution of carbon and

radiocarbon over the Anthropocene, and use of that model to determine the adjustment time of excess carbon, should be viewed not as sowing confusion, but as substantially contributing to elucidation of this complex and important problem.