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Comment on *"Scrutinizing the carbon cycle and CO_2 residence time in the atmosphere"* by H. Harde

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Abstract

Harde (2017) proposes an *alternative accounting scheme* for the modern carbon cycle and concludes that only 4.3% of today's atmospheric CO_2 is a result of anthropogenic emissions. As we will show, this *alternative scheme* is too simple, is based on invalid assumptions, and does not address many of the key processes involved in the global carbon cycle that are important on the timescale of interest. Harde (2017) therefore reaches an incorrect conclusion about the role of anthropogenic CO_2 emissions. Harde (2017) tries to explain changes in atmospheric CO_2 concentration with a single equation, while the most simple model of the carbon cycle must at minimum contain equations of at least two reservoirs (the atmosphere and the surface ocean), which are solved simultaneously. A single equation is fundamentally at odds with basic theory and observations. In the following we will (i) clarify the difference between CO_2 atmospheric residence time and adjustment time, (ii) present recently published information about anthropogenic carbon, (iii) present details about the processes that are missing in Harde (2017), (iv) briefly discuss shortcoming in Harde's generalization to paleo timescales, (v) and comment on deficiencies in some of the literature cited in Harde (2017).

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1. Residence time versus adjustment time

The global carbon cycle is currently not in a steady state as shown, for example, by measurements of atmospheric CO_2 concentration at Mauna Loa (Hawaii) and at the South Pole since 1958 (Dlugokencky et al., 2016). The main reason for this increase is the addition of 'anthropogenic CO_2' by burning of coal, oil, and gas, industrial processes and land use change (Le Quéré et al., 2016). In the case of non-steady state conditions one can ask the question: How long will the perturbation (here: higher atmospheric CO_2 concentration) stay? If one adds a certain amount of anthropogenic CO_2 to the atmosphere at time t_0, the concentration will increase suddenly and than fall off following a complicated function that depends on the response of the various active carbon reservoirs (surface ocean, intermediate and deep ocean, marine sediments, terrestrial biosphere). The time connected to such an relaxation in atmospheric CO_2 concentration is the **adjustment (or equilibration) time** and the timescale of interest for the problem at hand. The function how CO_2 relaxes after such an initial perturbation can be approximated by the sum of a few exponential functions with different characteristic timescales (e.g. Archer et al., 1997; Lord et al., 2016). Simple one-box models suggest adjustment times of about 70 years, but these models ignore many relevant processes and consequently under-estimate this timescale (Cawley, 2011). More complex models suggest adjustment (equilibration) times of well over 100 years, and that it depends on total anthropogenic emissions (Archer et al., 2009; Joos et al., 2013; Lord et al., 2016). When anthropogenic CO_2 is added continuously one has to run a global carbon cycle model that takes into account the responses by the various reservoirs mentioned above (e.g. Friedlingstein et al., 2006; Jones et al., 2013).

The timescale determined in Equations (7) and (8) in Harde (2017) is actually an approximation of the **residence time**, i.e. the average length of time for which an individual molecule of CO_2 remains in the atmosphere before being taken up by the ocean or terrestrial biosphere. Given the fluxes into, and out of, the atmosphere, we would expect a CO_2 molecule to only remain in the atmosphere for a few years, before being replaced by a molecule from one of the other reservoirs. The usual misunderstanding is that this is CO_2 leaving the atmosphere, rather than mostly just being exchanged, leaving no change in atmospheric CO_2 concentration. Even though the numbers presented in Harde (2017) are reasonable approximations for the residence time, they are largely irrelevant for what the paper later presents.

Changes in atmospheric CO_2 concentration following the initial perturbation depend on the net CO_2 flux out of the atmosphere, rather than - as in the case of the residence time - depending only on the flux into the natural sinks. Note that the residence and adjustment times refer to different and distinct aspects of the carbon cycle and have different definitions; a distinction clearly made in the IPCC First Assessment Report (Houghton et al., 1990, §1.2.1) as well as in more recent reports (Stocker et al., 2013, p. 1457). Thus to conflate residence time and adjustment time is a fundamental misunderstanding of the carbon cycle.

Given this difference between the residence time (years) and adjustment time (centuries to millennia) we would also not expect an enhancement in atmospheric CO_2 to be entirely composed of molecules of directly anthropogenic origin, even if the cause for such an enhancement were entirely anthropogenic. Therefore, the claim in Harde (2017) that the anthropogenic contribution makes up only 15% of the increase since the industrial era - even if correct - is not an indication that the increase is not entirely anthropogenic.

2. Most recent anthropogenic carbon inventory

Total anthropogenic CO_2 from fossil fuel and cement production emitted between 1750 and 2010 has accumulated to 365 PgC (Le Quéré et al., 2016). A further 153 PgC was emitted in the same time interval from land use changes (Le Quéré et al., 2016). In 2010 the atmospheric CO_2 concentration was approximately 390 ppm (Dlugokencky et al., 2016), a value that features prominently in the calculations of Harde (2017).

Anthropogenic carbon in the ocean can be tracked by various methods, e.g. the ^{13}C Suess effect (Eide et al., 2017), ΔC^* method (Gruber et al., 1996), or anthropogenic produced substances, such as CFCs (Lauvset et al., 2016). Various approaches have shown that the oceanic sink accounts for 48% of the total fossil-fuel and cement-manufacturing CO_2 emissions of 118 PgC emitted between 1800 to 1994 (Sabine et al., 2004). Landschützer et al. (2016) calculated an increase in anthropogenic carbon in the ocean of about 60 PgC released by fossil-fuel and cement-manufacturing CO_2 emissions between 1982 and 2012. Taking the temporal overlay of both studies into account, we find an anthropogenic carbon inventory of the fossil-fuel and cement-manufacturing CO_2 emissions 1800–2012 in the ocean of about 150 PgC. This is 41% of the accumulated emission from fossil fuel and cement production, or 29% of the total emissions including land use change. In this scenario, the rise in atmospheric CO_2 from a preindustrial value of 278 ppm (= 589 PgC) before 1750, to 390 ppm (= 827 PgC) in 2010 is solely due to anthropogenic emissions. They overprint any potential natural CO_2 outgassing from the ocean (see the decomposition of anthropogenic and natural fluxes between ocean and atmosphere in Gruber et al. (2009)). The rise in the atmospheric carbon reservoir by 112 ppm, or 238 PgC, corresponds to an airborne fraction of 46% of the total anthropogenic emissions of 518 PgC. The missing residual of the anthropogenic emissions of 130 PgC is assigned to terrestrial carbon uptake.

According to Harde (2017), 4.3% of the actual atmospheric CO_2 concentration is of anthropogenic origin. With an atmospheric CO_2 concentration of 390 ppm used in Harde (2017), reached in $\sim$2010, this is similar to a proposed anthropogenic CO_2 concentration of about 17 ppm or 36 PgC. It would imply that only 7% of the total anthropogenic emissions remained airborne. The airborne fraction of Harde (2017) is therefore a factor of 6.6 smaller than in the inventory that is supported by observational-based studies. If the approach in Harde (2017) was correct, it would directly asked for evidence where this anthropogenic carbon has been stored. Unfortunately, no further evidence for this storage has been given in the paper and as we have shown above, it cannot reside in the ocean.

3. Why is the Harde model too simple?

The core argument in Harde (2017), section 3, is about the lifetime of anthropogenic CO_2 in the atmosphere, closely related to the airborne fraction that remains after a given time. A framework is then developed in which both natural and anthropogenic carbon fluxes are analysed. In this framework one important part of the carbon cycle, which is of major relevance for the airborne fraction of CO_2, is missing: the carbonate chemistry in the ocean. It is correctly stated that Henry's Law governs the net gas exchange of CO_2 between the surface ocean and the atmosphere, with higher temperatures leading to a higher net flux to the atmosphere. However, within the ocean CO_2 molecules react with water to form carbonic acid (H_2CO_3) which subsequently dissociates into hydrogen ions (H^+) and bicarbonate ions (HCO_3^-). In a second step the HCO_3^--ion dissociates into another H^+-ion and a carbonate ion (CO_3^{2-}). The sum of all these carbon species is typically referred to as dissolved inorganic carbon (DIC). For present day conditions the fraction of carbonic acid is negligible; the majority of DIC (∼90%) is found as HCO_3^-, ∼9% as CO_3^{2-}, and only about 1% is found as dissolved CO_2 (Zeebe & Wolf-Gladrow, 2001). Only this 1% of DIC in the surface ocean, found as dissolved CO_2, can exchange with the atmosphere. Thus, the carbonate chemistry represents a bottleneck for the oceanic uptake of anthropogenic CO_2 emitted to the atmosphere. Note, that the basic knowledge on the marine carbonate system, which is completely neglected in Harde (2017), is at least 60 years old, e.g. see Revelle & Suess (1957) and references therein. Furthermore, different software packages to compute the marine carbonate chemistry have been published in the meantime (e.g. Orr et al., 2015), and are in most cases freely available, e.g. see http://cdiac.ornl.gov/ftp/oceans/co2sys/ for different versions of the package CO2SYS which was widely discussed in Orr et al. (e.g. 2015) or https://www.soest.hawaii.edu/oceanography/faculty/zeebe_files/CO2_System_in_Seawater/csys.html for Matlab routines to Zeebe & Wolf-Gladrow (2001).

This effect of the carbonate chemistry on the carbon cycle is not a theoretical concept, but an observed quantity also known as the Revelle (or buffer) factor R. This is a fundamental property of the marine carbonate system and is implicitly considered in marine carbon cycle models underlying the analyses summarized in the IPCC-AR5, the 5th Assessment Report of the Intergovernmental Panel on Climate Change (Stocker et al., 2013). The carbonate chemistry in seawater describing these processes in detail is well known (compare, for example Dickson et al., 2007; Zeebe & Wolf-Gladrow, 2001). The Revelle factor is defined as the ratio of the relative change of dissolved $\mathrm{CO_2}$ to the relative change of DIC and can be readily calculated:

$$R = \frac{\Delta \mathrm{CO_2}/\mathrm{CO_2}}{\Delta \mathrm{DIC}/\mathrm{DIC}} \tag{1}$$

From open ocean data it is known that R varies between 8 and 15 (Sabine et al., 2004). A Revelle factor of 8, for example, leads to a DIC increase by only 12.5% for a doubling of dissolved $\mathrm{CO_2}$. A rise in atmospheric and oceanic carbon content goes along with an increase in the Revelle factor, a phenomenon which is already measurable (e.g. Hauck et al., 2010). This implies that the oceanic uptake of anthropogenic carbon will become slower if we continue to increase anthropogenic $\mathrm{CO_2}$ emissions. This is already seen in all CMIP5 model simulations (Jones et al., 2013). The scientific literature describing the marine carbonate chemistry, which, if complete, automatically includes the Revelle effect, is based on decades of laboratory experiments, field observations and theoretical understanding of the underlying chemical processes and is very well established. The books by Dickson et al. (2007); Zeebe & Wolf-Gladrow (2001) are only two examples of the state of knowledge in this field.

The carbonate chemistry is the most relevant part of the carbon cycle, which is of importance on the timescale of interest, yet ignored in the erroneous approach of Harde (2017). The uptake of anthropogenic $\mathrm{CO_2}$ by the terrestrial part of the carbon cycle is also relevant (e.g. Joos et al., 2013), and nowadays approximately of similar size as the marine carbon uptake (Le Quéré et al., 2016). However, terrestrial carbon uptakes are in the global carbon budgets of Le Quéré et al. (2016) still estimated from the unexplained residual, after fossil fuels and emissions from industry and land-use change have been balanced by constraints on changes in carbon pools in the atmosphere and the ocean. We therefore refrain from a more in-depth discussion of terrestrial carbon uptake processes here. Further processes with different impact on the airborne fraction of anthropogenic CO_2, which are of relevance if longer timescales are of interest (e.g. necessary for the generalization and application to the paleo data) are ocean overturning, carbonate compensation and continental weathering rates (Lord et al., 2016). If implemented in a model this results in an airborne fraction of anthropogenic CO_2 emissions of around 40% on a 100-year timescale falling to ~18% on a 1000-year timescale (Joos et al., 2013) and down to 5% and 2% on timescales of 10^5 and 10^6 years, respectively (Lord et al., 2016).

Remark: Most of these details above on the role of the carbonate chemistry have been taken from another comment some of the authors published as part of the online discussion on another, overly simplistic, and therefore biased approach to explain the modern carbon cycle (http://www.earth-syst-dynam-discuss.net/6/C813/2015/esdd-6-C813-2015.pdf).

Harde's flawed 1-box carbon cycle: One key element of Harde's carbon cycle is the assumption of a simple absorption/decay process, which is unsuitable for the problem at hand. Harde's Eq. (11) reads:

$$\frac{dC_{\mathrm{CO_2}}}{dt} = e_T - \alpha \cdot C_{\mathrm{CO_2}} \ , \tag{2}$$

where $C_{\mathrm{CO_2}}$ is the atmospheric CO_2 concentration, e_T is a total emission rate, $\alpha = 1/\tau$ is an absorption efficiency, and τ is Harde's CO_2 "lifetime". Thus, Harde assumes that $C_{\mathrm{CO_2}}$ can be predicted by solving only a single rate equation of the carbon cycle (other reservoirs may exist but their time evolution is ignored). However, at any given time t, the CO_2 fluxes into and out of the atmosphere depend on, for instance, the atmosphere-ocean disequilibrium, which in turn depends on simultaneous changes in ocean carbon inventory and seawater chemistry, as explained above. Thus, even the simplest carbon cycle model must at minimum comprise two boxes for atmosphere and ocean (including Revelle factor), whose equations are solved simultaneously. For investigations of timescales longer than centuries (e.g. in paleo applications as done in the generalization) processes which export carbon from the surface to the deep ocean (so-called carbon pumps, see Volk & Hoffert, 1985) also need to be taken into consideration, asking for at least another deep ocean box. Yet, Harde (2017) ignores this fact (and many others) that have been established in over 60 years of carbon-cycle research (a few examples include Revelle & Suess, 1957; Oeschger et al., 1975; Heimann & Maier-Reimer, 1996; Archer et al., 2009; Joos et al., 2013). As a result, the approach in Harde (2017) leads to fundamentally flawed mass balances, CO_2 "lifetimes", and thus erroneous conclusions.

Note also that the posited analogy to radiocarbon (and other isotopes) is incorrect because changes in bulk inventory (total atmospheric CO_2) are confused with changes in isotopic $^{14}C/C$ ratio typically expressed as $\Delta^{14}C$. In detail, the concentration of CO_2 in the atmosphere, when perturbed by anthropogenic emissions, largely depends on the net oceanic CO_2 uptake rate, and therefore on the bottleneck of the carbonate chemistry as explained in section 3 above, while any changes in the isotopic $^{14}C/C$ ratio of atmospheric CO_2 also depends on the gross gas exchange rates (e.g. see Joos et al., 1996; Naegler & Levin, 2006). Furthermore, the record of atmospheric radiocarbon is perturbed/depleted by the emission of ^{14}C-free CO_2 from fossil fuels — the so-called ^{14}C Suess effect (Suess, 1955; Köhler, 2016) — and points to much longer atmospheric adjustment time on the order of 100 years for 1985–2005 (LEVIN et al., 2010).

4. Harde's generalization including approximations based on paleo reconstrucions

Finally, Harde (2017) generalizes the results that incorrectly model the modern carbon cycle to draw, again, erroneous conclusions about the paleoclimate record (section 3.3 and Figure 3). Here, again, various shortcomings invalidate the conclusions. The main ones are the following:

(a) **Glacial temperature:** The assumed surface temperature change for glacial times of -8 K is wrong, as the source cited (Petit et al., 1999) approximates not global temperature change, but that over East Antarctica. According to some recent studies the global temperature change at the last glacial maximum with respect to preindustrial times was -4.0 ± 0.8 K (Annan & Hargreaves, 2013). It is furthermore not clear to the reader how the data points in Figure 3 were generated with one data point for a temperature of 8, 10, 12, 14°C, while the underlying paleo data from the Vostok ice core contain several thousand data points of the last 420,000 years, also including periods which have been warmer than the preindustrial climate.

(b) **Explaining paleo CO_2:** Harde (2017) proposes that the complete glacial/interglacial change in CO_2 can be explained by a reduction in surface temperature. However, it is nowadays well established, that glacial/interglacial changes in atmospheric CO_2 can not be explained by one single process (e.g. Köhler et al., 2005; Brovkin et al., 2007; Kohfeld & Ridgwell, 2009). Significant change in atmospheric CO_2 on glacial/interglacial timescales are expected from a rise in sea surface temperature, rising sea-level, reduced marine export production, and responses from carbonate compensation, together with changing land carbon storage (Kohfeld & Ridgwell, 2009). Although models still disagree on the contribution of individual processes, the common consensus is, that the glacial/interglacial rise in temperature (more precisely sea surface temperature) might be responsible for a rise in atmospheric CO_2 mixing ratio of 20-30 ppm. The arguments in Harde (2017) are rather vague, but also seem to assume, that the temperature change might also trigger a change in terrestrial carbon storage. This concept would therefore need to have higher terrestrial carbon storage in cold periods, that might then be released during deglacial warming. However, the glacial terrestrial carbon storage is nowadays found to be smaller (not larger) in glacial times than during the preindustrial period (Ciais et al., 2012). So, again evidence contradicts what is needed to support the concept set forth in Harde (2017). One might now argue, that all mentioned processes vary synchronously with temperature, and therefore the chosen temperature-dependency in Harde (2017) might be a possible simplification. However, this would largely ignore the complexity of the carbon cycle-climate system including the fact that the paleorecords also contain interglacial periods with higher than preindustrial global surface temperature, but similar atmospheric CO_2 concentrations, such as the last interglacial about 130,000 years ago (e.g. Bakker et al., 2013; Past Interglacials Working Group of PAGES, 2016).

(c) **Paleo CO_2 data:** Furthermore, Harde (2017) argues that due to distortion and diffusion the CO_2 data from ice cores are rather imprecise leading to large error bars for CO_2 shown in Figure 3 and to 20–30 ppm lower values than reconstructions based on fossil stomata. It must be clarified that ice core based CO_2 perfectly overlaps with the instrumental measurements of atmospheric CO_2 which started in 1958 and we therefore see no support for the contention that they might be biased to lower values (e.g. Ahn et al., 2012; Rubino et al., 2013; Köhler et al., 2017). Furthermore, short term variations seen in stomata-based CO_2 during the Holocene have been heavily criticized, and when averaged for known enclosure characteristics of gas bubbles in ice cores have not been confirmed in ice core-based records (e.g. Ahn et al., 2014; Köhler et al., 2015). Together, the scientific arguments are in favour of the ice core-based CO_2 records and not of that based on fossil stomata.

(d) **Impact of strong dependency of CO_2 on surface temperature:** Finally, the dependency of the atmospheric CO_2 concentration on the surface temperature, as depicted in Harde's Figure 3 and his Equation 17 would imply — as also discussed in Harde (2017) — a large contribution of CO_2 from the oceans for rising temperatures (but also some contributions from land). A large CO_2 outgassing contribution from the oceans, however, is at odds with the atmospheric oxygen records (e.g. see Keeling & Shertz, 1992).

5. Citations and some details on IPCC

Harde (2017) cites throughout various parts of the contribution of Working

Group 1 (physical science basis) to the IPCC-AR5 (Stocker et al., 2013) and proposes *alternative views* on the impact of anthropogenic CO_2 emissions and related global temperature increase. This approach is inappropriate because it fails to address the actual underlying literature of the IPCC-AR5. The IPCC summarizes the state of the art in the peer-reviewed literature. Hence neither the residence time nor the adjustment time are assumptions or interpretations of the IPCC-AR5, but robust outcomes of the underlying science, which is summarized in the report. In presenting *alternative concepts* to the view presented in the IPCC-AR5 the article of Harde (2017) ignores and is in contradiction to the state of knowledge in the field, most obviously in the conflation of residence and adjustment times.

Some of the citations in Harde (2017) are inappropriate, including video presentations, and do not meet the standards of the peer-reviewed literature.

Further, Harde (2017) cites two papers (Essenhigh, 2009; Humlum et al., 2013) that were subject to highly critical peer-reviewed comments (Cawley, 2011; Masters & Benestad, 2013; Richardson, 2013; Kern & Leuenberger, 2013), none of which are referenced in Harde (2017). In fact, Harde (2017) repeats many of the same arguments that have already been refuted.

6. Conclusion

In conclusion, Harde (2017) does not provide an *alternative view* of the carbon cycle, but uses a too simplistic approach, that is based on invalid assumptions, and which leads to flawed results for anthropogenic carbon in the atmosphere. We suggest that the paper be withdrawn by the author, editor or publisher due to fundamental errors in the understanding of the carbon cycle.

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