Supplementary Material to
Using the Suess effect on the stable carbon isotope to
distinguish the future from the past in radiocarbon

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Model Description

In this study I use the well tested Box model of the Isotopic Carbon cYCLE (BICYCLE),
which has been applied in several case studies on impacts of both natural and
anthropogenic climate change on the evolution of the global carbon cycle (Köhler
et al. 2005, Köhler, Hartmann & Wolf-Gladrow 2010). The model consists of a scheme,
how prescribed changes in the physics of the climate system, e.g. ocean circulation, sea
ice coverage, temperature, external input of the micro-nutrient iron, lead to variations
in carbon fluxes between the various reservoirs, including changes in the carbon pumps
that bring C and associated nutrients from the surface to the deep ocean and therefore
to variable carbon budgets. Within the 10 oceanic, 1 atmospheric and 7 terrestrial boxes
of the model not only C content, but also both its isotopic signatures, $^{13}$C, $^{14}$C, are
traced. Furthermore, in the ocean total alkalinity, oxygen and PO$_4^{3-}$ concentration are
state variables, that change due to the variable physical boundary conditions. The model
also consists of a simplistic scheme how terrestrial carbon content in vegetation and soil
pool might alter due to a changing global temperature and atmospheric CO$_2$ concentration
and considers differences in isotopic fractionation due to C$_3$ or C$_4$ photosynthesis. The
terrestrial scheme is neglecting permafrost and peatland carbon pools and is not spatially
resolved, thus it might only act to guide some very simplistic zero order changes in the
carbon distribution between land, atmosphere, and ocean. However, it has been shown
recently (Köhler et al. 2015) that the CO$_2$ fertilization which might be realized within
such a simple scheme of the terrestrial biosphere leads to much too high land carbon
uptake for some RCP emissions scenarios. I therefore restrict my analysis in the following
to an atmosphere-ocean only system by keeping the terrestrial carbon content constant,
but I will show some results including the dynamical terrestrial biosphere for the historical
period.
BICYCLE also contains a time-delayed response function of changes in deep ocean carbonate ion concentration, that mimics the carbonate compensation effect (Broecker & Peng 1987), which is the response of the deep ocean - sediment fluxes of carbonate dissolution / accumulation to any changes in the carbon cycle. The impact of the carbonate compensation is on the time scales of interest (some centuries) small (simulated atmospheric CO$_2$ varies by less than 1%), but the process is included here for the sake of completeness.

Since my model-setup does not contain the physical part of the climate system, the global temperature change $\Delta T$ (relevant for both atmosphere–ocean gas exchange and the turnover time of carbon in terrestrial reservoirs) connected with a change in atmospheric CO$_2$ is calculated using the transient climate sensitivity (TCS) for CO$_2$ doubling, which has been obtained from more sophisticated climate models, and which has been recalculated to $\text{TCS} = 2$ K recently by a data-based approach (Storelvmo et al. 2016). In detail, I calculate $\Delta T = \text{TCS} \times \Delta R_{\text{CO}_2}/\Delta R_{2\times\text{CO}_2}$ with $\Delta R_{\text{CO}_2} = 5.35 \text{ W/m}^2 \cdot \ln(\text{CO}_2/278 \text{ ppmv})$ (Myhre et al. 1998). Changes in sea surface temperature (SST) are assumed to follow $\Delta T$ and changing SST will influence via Henry’s Law the CO$_2$ solubility in the ocean and isotopic fractionation during gas exchange (Zeebe & Wolf-Gladrow 2001).

The simulated time period contains the bomb spike in $^{14}$C in the second half of the 20th century and the depletion in both $\delta^{13}$C and $\Delta^{14}$C according to the historical Suess effects. In order to match observed variations in $\Delta^{14}$C as good as possible the $^{14}$C production rate is prescribed from (Roth & Joos 2013) varying around a mean production rate of 440 mol per year (Fig. S1C). The previous study (Graven 2015) also considered $^{14}$C production from the nuclear industry with assumed $^{14}$C emissions being constant at the 2005 level following a recent inventory (Graven & Gruber 2011). These nuclear industry $^{14}$C emissions were shown to be on the order of 10% of the natural $^{14}$C production rate. Here, I refrain from assuming any $^{14}$C emissions from nuclear industry, since its evolution in the future is difficult to propose. However, I estimate the size of its impact on the $^{14}$C cycle in BICYCLE in a sensitivity run, in which for RCP8.5 $^{14}$C production rate gradually rose from year 1980 onward to +10% in year 2005 CE (or to a relative $^{14}$C production rate of 1.1), and constant thereafter (Fig. S1C). The simulated atmospheric $\Delta^{14}$C based on this revised $^{14}$C production rate was 5% and 10% higher in year 2100 and 2500, respectively. Also note that the reconstructed size of the $^{14}$C emission from the nuclear industry is on the same order of magnitude as the variation in the natural $^{14}$C production rate in the industrial period (Fig. S1C), but smaller than its variability over the last 10,000 years (Roth & Joos 2013).

All simulations are started in year 10,000 BP to allow the $^{14}$C cycle to adjust to variable production rates. From 1950 CE onward the $^{14}$C production rate is kept constant, but was perturbed in individual years of the 1950ies to 1970ies by high peaks in $^{14}$C production caused by nuclear bomb testing (Naegler & Levin 2006) (Fig. S1C). The cumulative bomb-$^{14}$C production leads to the injection of $1.2 \cdot 10^6$ g $^{14}$C into the atmosphere after 1950, 15% smaller than suggested, because the natural background $^{14}$C production
rate in BICYCLE is also only 85% of that chosen previously (Naegler & Levin 2006).

Model Evaluation

For evaluation of the model performance in the historical period (Fig. S2) dynamics of $^{14}$C in the time windows 1820–1950 (historical $^{14}$C Suess effect) and 1950–2010 (bomb-$^{14}$C) have to be distinguished, since the impact of the Suess effect on $^{14}$C is after 1950 superimposed by bomb-$^{14}$C.

The time window 1820–1950 covers the full data set of one of the first reconstructions of the $^{14}$C Suess effect from tree ring data (Stuiver & Quay 1981). In this period all atmospheric carbon variables using a constant terrestrial biosphere (experiment TB–; my standard setup) have a small offset in the simulations from the data (Fig. S2), while their dynamic trends meet the evolution seen within the data: CO$_2$ rises by 30–35 ppmv, $\delta^{13}$C falls by 0.6–0.7‰, $\Delta^{14}$C falls by 20–25‰ after year 1900 superimposed on some decadal-scale variability, which was probably caused by changes in the $^{14}$C production rate (Roth & Joos 2013). The carbon cycle dynamics of the data are even better met by the model simulations which includes an active terrestrial biosphere (experiment TB+ in Fig. S2): a slightly smaller rise in CO$_2$, smaller decrease of $\delta^{13}$C more in line with the data, and hardly any offset in $\Delta^{14}$C.

In the 60 years including the bomb radiocarbon (1950–2010) the simulated CO$_2$ rises by 108 ppmv in experiment TB–, which is more than the observed rise by 80 ppmv (Fig. S2), but well within the uncertainty band of the C$^4$MIP results (Friedlingstein et al. 2006). This offset is certainly caused by the fixed terrestrial carbon pools in my setup. In scenarios with active terrestrial biosphere simulated CO$_2$ rises by 71 ppmv between 1950 and 2010, agreeing with the lower range of the C$^4$MIP range of results. In the historical period the land carbon is the least known pool and its change is typically derived from the residual after observed and modeled change in atmosphere and ocean have been subtracted from the anthropogenic emissions and during the historical period this residual land carbon sink took up about a fourth of the emissions (Le Quéré et al. 2015). The decreasing trend in simulated atmospheric $\delta^{13}$C was with $-2.12‰$ in TB+ larger than the decrease of about $-1.4‰$ in the data (Fig. S2B). This model-data mismatch is also caused by the missing terrestrial carbon sink, since the simulated trend of $-1.16‰$ in atmospheric $\delta^{13}$C in TB– agrees better with the trend in the data. Since simulated CO$_2$ in the long term agrees reasonable well with CMIP5 data (Figs. 1D, 2C) I judge this misfit in atmospheric $\delta^{13}$C to be only of minor importance for the overall conclusions.

The global mean atmospheric $\Delta^{14}$C peaks in the data in the mid 1960s at $700\pm200‰$ and declines towards $+50‰$ in year 2010 thereafter. The simulated peak in bomb-$^{14}$C is with $+900‰$ at the upper end of the range of reconstructions, decaying thereafter to $+5‰$ in year 2010 (Fig. S2C). The decay of the $\Delta^{14}$C peak in atmosphere is faster in the model than in the data which indicates that the vertical mixing between surface and deep ocean in the model operates faster than in nature. This is a phenomenon well known for box models, but less pronounced in BICYCLE than in other box models (Köhler et al. 2005, Broecker et al. 1999).
Simulated ocean acidification represented by a fall in surface ocean pH is difficult to compare with data, because observations exist only for a few sites since about 1990 (Doney et al. 2009). Nevertheless, the decline of \( \sim 0.02 \) pH units per decade over less than 20 years detected in these data is in agreement with the BICYCLE simulations shown here (Fig. 1G). The time series of the pH data are so short that I do not show them in the figures.

One integrated approach to evaluate my model performance is to plot the calculated temperature change \( \Delta T \) as a function of cumulative CO\(_2\) emission (Fig. S3). When compared with CMIP5 results, which are here restricted to scenarios with CO\(_2\) emissions only (neglecting global warming connected with anthropogenic emissions of CH\(_4\), N\(_2\)O, or any aerosol effects) I find my box model simulations very well in the middle of the uncertainty range spanned by simulation results of the Earth system models (ESM) contributing to CMIP5. Until the year 2100 I would find in RCP8.5 (about 2500 PgC of cumulative CO\(_2\) emissions) a warming of 4 K, which rises to a maximum of 5.7 K for the cumulative CO\(_2\) emissions of 5300 PgC. The slight decline towards 5.5 K for even higher cumulative CO\(_2\) emissions (nearly 6000 PgC) is due to the small annual emission rate of 1.5 Pg C yr\(^{-1}\) during the last 250 simulated years within RCP8.5 which allows the ocean to absorb more CO\(_2\) than is emitted, therefore lowering atmospheric CO\(_2\) and global warming. Also note, that in my simple modeling approach \( \Delta T \) is not a linear function of cumulative CO\(_2\) emission (Fig. S3). Such a non-linear relationship between \( \Delta T \) and cumulative CO\(_2\) emission has already been found for results based on Earth system models of intermediate complexity (EMICs) (Allen et al. 2009), while state-of-the-art ESM contributing to CMIP5 find this relationship to be rather linear, not only for the 21st century (IPCC 2013), but also for cumulative emissions up to 5000 PgC (Tokarska et al. 2016). For comparing my simple carbon cycle model with these results based on more complex models, one needs to be aware that no warming beyond that caused by CO\(_2\) is contained in my results. Furthermore, it is even not yet clear why the results based on ESMs and EMICs differ for high cumulative CO\(_2\) emissions (Frölicher 2016).

Another evaluation method for carbon cycle models is the simulation of a CO\(_2\) pulse response (Joos et al. 2013). The model response to the instantaneous injection of 100 PgC into the atmosphere for modern background conditions (here: atmospheric CO\(_2\) concentration of 389 ppmv) is then investigated. The airborne fraction \( f \) of this CO\(_2\) pulse decays over time. In my atmosphere-ocean version of the BICYCLE model with constant terrestrial biosphere I find \( f \) of 0.45 after one century to decline towards 0.20 after one millennium, well in agreement with results from more complex models \((f = 0.41 \pm 0.13 \text{ (2\sigma)}\) and \(f = 0.25 \pm 0.09\) after 100 and 1000 years, respectively) which contributed to the intercomparison study (Joos et al. 2013).

If compared directly with the previous study (Graven 2015) one needs to keep in mind that here the whole carbon cycle including the carbon isotopes are freely evolving in response to changing boundary conditions (implying that I prescribe natural and bomb-\(^{14}\)C production of radiocarbon), while in the previous approach the measured atmospheric \( \Delta^{14}\)C data for the historical period have been prescribed. As result of this difference in the
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setup, I am here able to compare simulated $\Delta ^{14}$C with data for the past to test the model performance, while this is *per se* not possible in Graven (2015). The radiocarbon age and the corresponding atmospheric $\Delta ^{14}$C in year 2100 are in my simulations 2343 years ($-253\%$) in RCP8.5, 1516 years ($-172\%$) in RCP6.0, 758 years ($-90\%$) in RCP4.5 and 261 years ($-32\%$) in RCP2.6. My simulated age for RCP2.6 is slightly older ($\Delta ^{14}$C smaller) than in (Graven 2015), while all other results agree well with this previous study. All-together, I conclude that both modeling approaches are similar in complexity and produce comparable results.

References


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Figure S1: Detailed forcing of the historical simulations. A: Anthropogenic emissions, total and subdivided in those based on fossil fuels or land use change (Meinshausen et al. 2011). Note, that fossil fuel emissions also contains CO$_2$ release from cement production. B: The related $\delta^{13}$C signatures of the land use change (internally calculated), fossil emissions (Andres et al. 2000, Andres et al. 2015) and the mean $\delta^{13}$C of the total emission flux. Black broken line shows $\delta^{13}$C signature of fossil fuel emissions following a gradually increase in cement production (to 100% in year 2250) in the fossil fuel source mix used in scenario RCP8.5@cement. C: $\Delta^{14}$C of the total anthropogenic emissions and the relative change in the $^{14}$C production rate (Roth & Joos 2013). Broken line (1980 – 2000) indicates a rise in $^{14}$C production rate by 10% in the year 2005 (and constant thereafter) due to the nuclear industry (Graven & Gruber 2011), whose impact is tested in a sensitivity study. Dots in panel C are anthropogenic (bomb-based) increases in $^{14}$C production rate derived from a closure of the $^{14}$C cycle (Naegler & Levin 2006) on its own y-axis.
Figure S2: Evaluating the historical simulations. Comparing atmospheric (A) CO$_2$, (B) $\delta^{13}$C, (C) $\Delta^{14}$C of historical simulations of the BICYCLE carbon cycle model with data. In the BICYCLE simulations the terrestrial biosphere is either passive (=constant) (TB-) or active (TB+). Vertical line in (C) indicates the break in the y-axis in $\Delta^{14}$C at 1950 CE. CO$_2$: instrumental (Mauna Loa) (Keeling & Whorf 2005) and Law Dome ice core (Rubino et al. 2013); $\delta^{13}$C: instrumental (Point Barrow, South Pole) (Keeling et al. 2001), Law Dome and WAIS Divide ice cores (Rubino et al. 2013, Bauska et al. 2015); $\Delta^{14}$C: pre-bomb reconstructions of $\Delta^{14}$C (IntCal13 (Reimer et al. 2013)) including the historical $^{14}$C Suess effect (Stuiver & Quay 1981) and the $^{14}$C-bomb peak (global mean and range) (Hua et al. 2013). Monthly mean data of the instrumental periods were aggregated into annual mean values.
**Figure S3:** Global mean surface temperature increase as a function of cumulative global CO\(_2\) emissions. Colored lines are own simulation results with the BICYCLE model for the four different RCP emission scenarios with passive terrestrial biosphere using the net CO\(_2\) emissions. Simulation results show changes from the beginning of the emissions (year 1765) until year 2100 (thick lines), and thereafter (2101–2500, thin lines). For the BICYCLE results I directly calculate \(\Delta T\) from CO\(_2\) using a transient climate response of 2 K as given in the methods. For comparison the multi-model mean and range simulated by CMIP5 models, forced by a CO\(_2\) increase of 1% per year is given by the broken black line and gray area (after Figure SPM 10 of (IPCC 2013)). These simulations exhibit lower warming than those driven by RCPs within CMIP5, which include additional non-CO\(_2\) forcings and therefore lead to higher temperature changes. For the CMIP5 results \(\Delta T\) until the year 2100 is calculated relative to the 1861–1880, CO\(_2\) emissions relative to 1870.
**Figure S4:** Analysis of the combined Suess effects on both $^{14}$C and $^{13}$C for oceanic surface and deep reservoirs: (A) surface North Atlantic (same data as in Fig. 3B); (B) surface Equatorial Atlantic; (C) deep Atlantic; (D) surface Southern Ocean; (E) deep Southern Ocean; (F) surface North Pacific; (G) surface Equatorial Indo-Pacific; (H) deep Indo-Pacific (same data as in Fig. 3C). Here, deep ocean boxes are all water masses below 1000 m; surface water boxes are 100 m deep in the equatorial region and 1000 m deep in the high latitudes; North Atlantic (Pacific) is north of 50° N (40° N); Southern Ocean is south of 40° S. A more detailed description of the definition of the different reservoirs including water mass fluxes is found elsewhere (Köhler, Fischer & Schmitt 2010). Scatter plots of simulated $\Delta^{14}$C versus $\delta^{13}$C showing the historical and future Suess effect and the influence of bomb-$^{14}$C, future CO$_2$ emissions and carbon dioxide reduction (CDR) approaches (BECCS, DAC, EW) on both variables. Also included in dotted lines are results for RCP2.6, RCP4.5 and RCP6.0, which all contain a prescribed contribution of BECCS (see Fig. 1 for details). For comparison, also the available paleo knowledge is added. I show the data range obtained from sediment cores in deep ocean $\delta^{13}$C (Peterson et al. 2014) for a fixed value of $\Delta^{14}$C = 100‰ obtained for the Last Glacial Maximum (LGM) and the late Holocene (HOL). For the surface ocean $\Delta^{14}$C in Marine13 (Reimer et al. 2013) is plotted. Additionally, the range in both isotopes in previously published (imperfect) simulations using the BICYCLE model covering the last 50,000 year (50 ka) (upper limit of scenario S3x ($^{14}$C production rate based on $^{10}$Be) and lower limit of scenario S4x ($^{14}$C production rate based on reconstructions of the geomagnetic field strength GLOPIS-75) as used before (Köhler et al. 2006)). The gray broken line in all subplots crosses values for year 2020 with a slope $m = 50$ (see text for further explanation).
Figure S4: Caption on previous page.