

Radiocarbon in the land and ocean component of the Community Earth

System Model

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Key Points: (< 140 characters)

The uptake of bomb-produced ¹⁴C by the ocean and land is simulated with the Parallel Ocean Model POP2 and the Community Land Model CLM5

¹⁴C uptake by CLM5 is lower than observational estimates and carbon allocation and overturning in forest ecosystems are biased.

The deep ocean of POP2 is ventilated too slowly and radiocarbon ages are several centuries older than estimates from observations

Abstract (< 250 words) 242 words

Large amounts of the carbon-isotope ¹⁴C, entering Earth's carbon cycle, were produced in the atmosphere by atomic bomb tests in the 1950s and 1960s. Here, we forced the ocean and land components of the Community Earth System Model with atmospheric ¹⁴CO₂ over the historical period to constrain overturning time scales and fluxes. The uptake of bomb ¹⁴C by the land model is lower than observation-based estimates. This mismatch is likely linked to too low ¹⁴C uptake by vegetation as the model overestimates ¹⁴C/C ratios of modern soils indicating model biases in forest productivity or wood carbon allocation and turnover. The ocean model matches the observation-based global bomb ¹⁴C inventories when applying the Large and Yeager wind data

and the quadratic relationship between gas transfer piston velocity and wind speed of Wanninkhof, 2014. However, ocean bomb ^{14}C inventories are underestimated in simulations with winds from the Japanese Reanalysis Project, calling for an upward revision of the piston velocity by 15% for this wind product. The sum of ocean, land, and atmospheric bomb ^{14}C inventory changes is lower in the 1960s than reconstructed bomb ^{14}C production, likely due to uncertainties in the observational production and atmospheric records and too low land model ^{14}C uptake. Simulated natural radiocarbon ages in the deep ocean are many centuries older than data-based estimates, indicating too slow deep ocean ventilation. Our study suggests that ^{14}C observations are key to constrain carbon fluxes and transport timescales within Earth system models.

Index Terms: 0414 Biogeochemical cycles, processes, and modeling; 0428 Carbon cycling; 1622 Earth system modeling; 1635 Oceans; 4504 Air/sea interactions

Keywords: Radiocarbon, bomb ^{14}C , land biosphere, gas transfer piston velocity, deep ocean ventilation.

1. Introduction

Radiocarbon (^{14}C) is produced naturally in the upper atmosphere by cosmic rays (Masarik and Beer, 2009). In addition, large amounts of ^{14}C were injected into the stratosphere as a byproduct of nuclear bomb tests in the 1950s and 1960s (Enting, 1982). Natural and bomb-produced ^{14}C is quickly oxidized to $^{14}\text{CO}_2$ and becomes part of the Earth's carbon cycle (Siegenthaler, 1989). ^{14}C , unlike the stable ^{12}C and ^{13}C isotopes, decays radioactively with a mean lifetime of 8200 years (Bé et al., 2013). The decay rate serves as a clock once carbon is isolated from the atmosphere. Radioactive decay is too slow to have noticeably affected bomb ^{14}C up to now. However, carbon in the deep ocean and soils is depleted in natural ^{14}C with respect to atmospheric carbon because of radioactive decay (Revelle and Suess, 1957; Oeschger et al., 1975). The slower the carbon exchange with the atmosphere the larger the depletion. Observations of the transient, pulse-like bomb ^{14}C signal and the natural $^{14}\text{C}/\text{C}$ ratio provide therefore constraints on the uncertain time scales of exchange of carbon and other tracers

between and within different Earth system components. These constraints are important as correctly representing the overturning time scales (Bolin and Rodhe, 1973) and exchange fluxes of carbon, heat, and other tracers is a prerequisite to reliably project the fate of anthropogenic CO₂ and ultimately global warming.

The goal of this study is to evaluate the representation of key carbon fluxes and transport time scales in the ocean and land components of the Community Earth System Model (CESM) with ¹⁴C observations. We rely on ¹⁴C-enabled simulations over the historical and preindustrial periods. Model results are compared to the observational evidence for the redistribution of bomb ¹⁴C among the atmosphere, ocean, and land biosphere, and the observation-based natural ¹⁴C age of the deep ocean. The Earth system budget of bomb ¹⁴C is established by comparing bomb ¹⁴C production statistics and inventory changes in the atmosphere, ocean, and land biosphere.

The natural cycle of ¹⁴C is strongly perturbed by human activities. Atmospheric ¹⁴C/C varied relatively little over the past few millennia before the first nuclear bombs detonated (Reimer et al., 2020;Hogg et al., 2020). The tropospheric ¹⁴C/C ratio almost doubled within a decade before a nuclear test ban treaty was set in place in 1963 (Naegler and Levin, 2009a). Over time, this bomb ¹⁴C signal entered the ocean, vegetation, and soils (Lawrence et al., 2020;He et al., 2016;Hesshaimer et al., 1994;Joos, 1994;Broecker et al., 1985) which caused a decline in the tropospheric¹⁴C/C ratio and the ¹⁴C inventory of the atmosphere after 1963. The emissions of ¹⁴C-free fossil CO₂ forced an additional negative trend in atmospheric ¹⁴C/C (Graven, 2015;Naegler and Levin, 2006;Suess, 1955). Small anthropogenic ¹⁴C sources stem from the nuclear power industry (Zazzeri et al., 2018;Graven and Gruber, 2011). For convenience, the terms “bomb” and “excess” are used interchangeably in this manuscript to describe the changes in ¹⁴C/C and ¹⁴C inventories due to all processes, including natural variability, since 1945.

The ¹⁴C variations of the recent past and the modern distribution of ¹⁴C in the Earth system are documented by compilations from various climate archives such as tree rings and corals (Reimer, 2020;Dentith et al., 2019;Graven et al., 2012;Naegler and Levin, 2009a;Grottoli and Eakin, 2007;Druffel, 2002), and modern sampling in the atmosphere, ocean, and land biosphere (Turnbull et al., 2017;Levin et al., 2010;Key et al., 2004;He et al., 2016;Lawrence et al., 2020;Shi et al., 2020). For comparison with observations and improved process understanding, ¹⁴C has been implemented in box models (Oeschger et al., 1975;Broecker et al.,

1985;Siegenthaler and Joos, 1992;Naegler and Levin, 2006), dynamic ocean circulation models (Jahn et al., 2015;Mouchet, 2013;Toggweiler et al., 1989;Rodgers et al., 2004), spatially-resolved land biosphere models (Koven et al., 2013;Roth and Joos, 2013;Randerson et al., 2002), and atmospheric transport models (Rodgers et al., 2011;Krakauer et al., 2006;Randerson et al., 2002;Braziunas et al., 1995). The ocean modeling community established protocols to simulate the uptake and distribution of natural and bomb ^{14}C (Orr et al., 2017;Orr et al., 1999) for the evaluation of transport time scales and water mass age (Khatiwala et al., 2012).

Measurements documenting bomb ^{14}C in vegetation and soils are relatively scarce and do not permit to directly establish the bomb ^{14}C inventory of the global land biosphere. Naegler and Levin (2009a) estimated the bomb ^{14}C evolution in the land biosphere by the difference between total ^{14}C production estimated from bomb-test statistics, the observed change in the atmospheric inventory and the observation-constrained uptake by the ocean. Naegler and Levin (2009b) used their bomb inventory reconstruction in combination with a 3-box land biosphere model to estimate global net primary productivity (NPP) to 41 to 64 GtC yr^{-1} . The representation of NPP and carbon turnover time scales is essential to simulate the terrestrial sink of anthropogenic carbon (Thompson et al., 1996). The global observational constraint of Naegler and Levin (2009a) has, however, not yet been applied by others to evaluate NPP and carbon turnover of spatially-resolved state-of-the-art terrestrial models.

Observations of the bomb ^{14}C tracer in the ocean are particularly useful to constrain the air-sea gas transfer piston velocity (Broecker et al., 1985;Wanninkhof, 1992;Siegenthaler, 1989). The piston velocity is a key parameter for the observational and modeling community and used to compute air-sea fluxes of various gases as well as the marine carbon sink from observations of the air-sea CO_2 partial pressure difference (Wanninkhof, 1992;Watson et al., 2020;Iida et al., 2020). The piston velocity is typically parameterized as a function of wind speed (Woolf et al., 2019). The scaling coefficients of these parameterizations have been determined with reduced and intermediate complexity and transport matrix models by minimizing differences between the observed and simulated bomb ^{14}C signal (Wanninkhof, 2014;Naegler, 2009;Müller et al., 2008;Sweeney et al., 2007;Krakauer et al., 2006;Naegler et al., 2006) but uncertainties in the piston velocity of order 20% remain (Wanninkhof, 2014; Naegler 2009). The values of the scaling coefficients are closely tied to the applied wind product and need potentially to be adjusted for different wind products. The application of different wind products in ^{14}C -enabled

ocean models, as in this study, offers the opportunity to evaluate the applied piston velocity parameterization and its coefficients for specific wind products against observation-based global ocean bomb ^{14}C inventory estimates.

The Earth system budget of bomb ^{14}C allows one to assess global exchange fluxes of carbon between the atmosphere, ocean, and land biosphere and to test overall consistency of model results and observations (Hesshaimer et al., 1994; Naegler and Levin, 2009a). Bomb ^{14}C production records combined with observation-based estimates of the changes in the stratospheric and tropospheric bomb ^{14}C inventories reveal the combined bomb ^{14}C uptake by the land and ocean from the atmosphere.

While many studies are addressing ^{14}C in dynamic ocean models, ^{14}C simulations with spatially-resolved land biosphere models are scarce and simulations with state-of-the-art Earth System Models addressing the budget of bomb ^{14}C and air-sea and air-land fluxes are missing. Thus far, the models participating in the Coupled Model Intercomparison Project (Eyring et al., 2016), which are used for carbon cycle and climate projections in the assessments of the Intergovernmental Panel on Climate Change, have generally not been evaluated against ^{14}C observations.

Recently, carbon isotopes were added to the marine (Jahn et al., 2015) and land biosphere components (Koven et al., 2013) of the Community Earth System Model (CESM). Shi et al. (2020) compared $\Delta^{14}\text{C}$ of soil carbon as simulated with CLM5 with their soil $\Delta^{14}\text{C}$ data. Here, we build on this earlier work and present results from ^{14}C -enabled simulations with the standard version of POP2 with a nominal horizontal resolution of 1° and with the latest version 5 of CLM for the preindustrial state and the historical period. POP2 and CLM5 are standard components of CESM version 2. POP2 simulations over the historical period are performed with two wind products, the Large and Yeager Normal Year Forcing (NYF, Large and Yeager (2009)) and the Japanese Reanalysis project (JRA55, Kobayashi et al. (2015)) data, to investigate the sensitivity of results to the wind forcing and the related piston velocity.

We analyze the bomb ^{14}C budgets of the Earth, the ocean, and the land biosphere to evaluate the land and ocean components of CESM2. The modeled evolution of the bomb ^{14}C inventory of the land biosphere is compared with the observational estimate of Naegler and Levin (2009a). We further compare estimates of the global ocean bomb ^{14}C inventory established from marine

measurements, the temporal evolution of surface ocean $^{14}\text{C}/\text{C}$ during the bomb period as recorded by corals and bivalves, and the gridded bomb $^{14}\text{C}/\text{C}$ data from the Global Ocean Data Analysis Project (GLODAP) with model results. The observed changes in the atmospheric bomb ^{14}C inventory and the bomb ^{14}C production record allow us to estimate whether the combined bomb ^{14}C uptake by the ocean and the land biosphere models is simulated in agreement with observations. Finally, we compare modeled and measured $^{14}\text{C}/\text{C}$ in the deep ocean to assess the model's ventilation time scales.

2. Methods

2.1 Isotopic Notation

We adopt conventional isotopic notation. The $^{14}\text{C}/\text{C}$ ratio ($^{14}R_{\text{sample}}$) of a pool or sample is reported as a deviation from a standard ratio ($^{14}R_{\text{std}}=1.176 \cdot 10^{-12}$) in permil (‰) using the so-called Δ -notation. It holds $\Delta^{14}\text{C}_{\text{sample}}=(^{14}R_{\text{sample,N}}/^{14}R_{\text{std}} -1) \cdot 1000$. Δ -values are corrected for fractionation by normalizing $^{14}R_{\text{sample}}$ to a fixed $^{13}\text{C}/^{12}\text{C}$ ratio of -25 ‰. The normalized ratio is $^{14}R_{\text{sample,N}}=^{14}R_{\text{sample}} (1-2 (\delta^{13}\text{C}_{\text{sample}}+25)/1000)$. $\delta^{13}\text{C}_{\text{sample}}$ is the deviation of the $^{13}\text{C}/^{12}\text{C}$ ratio of the sample from a standard ratio ($^{13}R_{\text{std}}=0.0112372$) in permil and, similarly, $\delta^{14}\text{C}_{\text{sample}}$ is the deviation of the $^{14}\text{C}/\text{C}$ ratio of the sample from $^{14}R_{\text{std}}$. It holds $\delta^i\text{C}=(^iR_{\text{sample}}/^iR_{\text{std}} -1) \cdot 1000$ with index i indicating the isotope. $\Delta^{14}\text{C}$ is then related to $\delta^{14}\text{C}$ and vice versa (Stuiver and Polach, 1977):

$$\Delta^{14}\text{C} = \delta^{14}\text{C} - 2(\delta^{13}\text{C} + 25)(1 + \frac{\delta^{14}\text{C}}{1000}) \quad (1a)$$

$$\delta^{14}\text{C} = \frac{\Delta^{14}\text{C} + 2(\delta^{13}\text{C} + 25)}{1 - \frac{2(\delta^{13}\text{C} + 25)}{1000}} \quad (1b)$$

The isotopic fractionation α is related to the fractionation factor ε in permil units by $\varepsilon=(\alpha-1) \cdot 1000$ (Mook, 1986).

2.2 Model description

The Community Earth System Model version 2 (CESM2) (Danabasoglu et al., 2020;Hurrell et al., 2013) is a state-of-the-art Earth System Model developed by the National Centre for Climate Research (NCAR), USA. Recently, the carbon isotopes ^{13}C and ^{14}C were added to the ocean (Jahn et al., 2015) and land component (Keller et al., 2017;Koven et al., 2013;Oleson et al., 2013) of CESM2. Jahn et al. applied a version of the Parallel Ocean Model version 2 (POP2) with a horizontal resolution of about 3° to simulate the distribution of carbon, ^{13}C , and ^{14}C within the ocean for the preindustrial state and the historical period. Koven et al. (2013) described the implementation of a radiocarbon tracer within the Community Land Model version 4.5 (CLM4.5) and compared modeled and observed ^{14}C soil profiles for a range of sites. In this study, we applied the carbon isotope-enabled POP2 ocean (Danabasoglu et al., 2012;Danabasoglu et al., 2020) and the most recent version 5.0 of CLM (Lombardozzi et al., 2020;Kennedy et al., 2019;Lawrence et al., 2019). Both components are used in a so-called stand-alone mode and driven by atmospheric forcing data (see section 2.3). A brief description of the isotope-enabled POP2 and CLM5.0 models is provided below.

2.2.1 Ocean model: POP2/MARBL

POP2 are run on the standard model grid with 60 vertical layers and a horizontal resolution of about 1° and finer resolution around the equator. The marine biogeochemical cycle is based on the Biogeochemical Elemental Cycling (BEC) model (Moore et al., 2004;Moore et al., 2002) and handled by the Marine Biogeochemistry Library (MARBL). It represents the cycling of carbon, the carbon isotopes ^{13}C and ^{14}C , nitrogen, phosphorus, iron, silica, oxygen, and alkalinity. Carbon isotopes are exchanged between the seven tracers: dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), calcium carbonate (CaCO_3), and three different phytoplankton (small phytoplankton, diatoms, diazotrophs) and one zooplankton pool.

The net air-to-sea gas flux of CO_2 , $F_{a,s,\text{net}}$, is modeled as the product of the piston velocity and the difference between the saturation, C_{sat} , and surface water, C_s , concentration of dissolved CO_2 following Wanninkhof (2014):

$$F_{a,s,\text{net}} = (1 - a_{\text{ice}}) \cdot a \cdot u_{10}^2 \cdot \left(\frac{660}{Sc_{\text{CO}_2}} \right)^{-0.5} \cdot (C_{\text{sat}} - C_s) = k \cdot (C_{\text{sat}} - C_s) \quad (2)$$

a_{ice} is the fraction of a grid cell covered by ice, a is a scaling factor, u_{10} is the wind speed at 10 m above sea level, and Sc_{CO_2} the Schmidt number of CO_2 , and k the piston velocity with respect to the liquid phase. A similar approach is used for computing the air-to-sea gas fluxes of the isotopes of CO_2 . The scaling factor a is set to $0.251 \text{ cm h}^{-1} / (\text{m s}^{-1})^2$ as in Wanninkhof (2014), substantially lower than the value of $0.31 \text{ cm h}^{-1} / (\text{m s}^{-1})^2$ assumed by Jahn et al. (2015). This downward revision of the piston velocity is broadly in agreement with earlier re-assessment of the bomb-produced radiocarbon distribution within the ocean (Müller et al., 2008; Sweeney et al., 2007; Peacock, 2004). The value of the coefficient a of $0.251 \text{ cm h}^{-1} / (\text{m s}^{-1})^2$ was derived using the Cross-Calibrated Multi-Platform (CCMP) wind product at 0.25° and 6-h resolution (<http://podaac.jpl.nasa.gov/datasetlist?search=ccmp>) by matching observations of bomb $DI^{14}C$ in an inverse ocean transport model approach (Wanninkhof, 2014). We note that the value of a is tied to the applied wind product and its use with other wind products may lead to different air-sea fluxes and, in turn, bomb ^{14}C inventory and $\Delta^{14}C$ values.

^{14}C is, as ^{13}C and carbon, cycled between the atmosphere and ocean and between all marine ecosystem carbon pools. Fractionation of ^{13}C versus ^{12}C is implemented, as described by Jahn et al. (2015), following Zhang et al. (1995) for air-sea gas exchange and following Laws et al. (1995) for photosynthesis, and fractionation of 2 ‰ is assumed for the formation of calcium carbonate. Fractionation for ^{14}C is twice as large as for ^{13}C . Besides, a so-called “abiotic” ^{14}C tracer is implemented in POP2 (Jahn et al., 2015). Abiotic ^{14}C does not cycle through the organic matter pools and the fractionation factors for air-sea gas exchange fluxes are set to unity. The abiotic implementation is a simplification, but requires much less computing resources than the biotic implementation. We will compare simulated $\Delta^{14}C$ between the two implementations to test the validity of the abiotic implementation. ^{14}C decays in POP2 with a half-life of 5730 years. We note that the value of the half-life has recently been updated based on new measurements to 5700 years (Orr et al., 2017), but this small revision has not yet been implemented in CESM. The standard ratios for $^{13}C/^{12}C$ and $^{14}C/C$ are set to unity in the POP2 model. Model units are converted by applying the standard $^{14}C/C$ ratio of $1.176 \cdot 10^{-12}$ for the computation of ^{14}C inventories.

2.2.2 Land model: CLM5.0

CLM5.0 represents terrestrial carbon and nitrogen dynamics and features various vegetation, litter, and soil organic matter pools (Oleson et al., 2013). Each grid cell is covered by different land-use classes. Each class has its own set of plant functional types (PFTs) and soil columns. Vegetation is described by 15 different PFTs which either follow the C3 or C4 photosynthesis pathway. Twenty carbon (C) pools per PFT store C in vegetation. C is tracked for leaf, live stem, dead stem, live coarse root, dead coarse root, and fine root pools and corresponding storage pools representing, respectively, short-term and long-term storage of nonstructural carbohydrates. Decomposition of fresh litter material into progressively more recalcitrant forms of soil organic matter is formulated as a cascade of transformations between decomposing coarse woody debris, three litter, and three soil organic matter pools. These pools are vertically resolved by 10 layers over a depth of 3.8 m. Discrimination of ^{13}C is implemented following (Farquhar et al., 1989) considering kinetic fractionation during the diffusion of CO_2 across the leaf boundary layer and into the stomata and during enzymatic fixation for C3 plants. Unlike in POP2, ^{14}C fractionation is not explicitly considered and CLM carries fractionation corrected ^{14}C . The standard ratio for $^{13}\text{C}/^{12}\text{C}$ and $^{14}\text{C}/\text{C}$ are set to 0.0112372 and 10^{-12} in CLM5.0. Model results in gram- ^{14}C are converted by multiplication with $1.176 \cdot 14 / 12.011 = 1.3707$ to compute ^{14}C inventories with 1.176 representing the ratio between the real and the model standard ratio, and 14 g/mol and 12.011 g/mol the molar mass of ^{14}C and C, respectively. ^{14}C decays in CLM5.0 with a half-life of 5730 years.

2.3 Setup of simulations

Eight simulations were performed. These include a spin-up (SPIN) as well as a simulation over the historical period 1850 to 2015 (HIST) and an associated 165-yr control run (CTRL) for both the land biosphere (LN) and the ocean-sea ice components (OC) with NYF. In addition, POP2 was run with an alternative climate forcing (JRA55) over the historical period and in a corresponding control.

Spin up and control runs were forced with 1850 conditions for atmospheric CO_2 (284.7 ppm), $\delta^{13}\text{C}$ (-6.61 ‰), and $\Delta^{14}\text{C}$; prescribed atm. $\Delta^{14}\text{C}$ is distinguished between 3 latitudinal bands ($>30^\circ\text{N}$: -2.3‰; 30°S - 30°N : -4.0‰; $<30^\circ\text{S}$: -5.8‰) following Graven et al. (2017). The

historical period simulation is forced with prescribed, transient atmospheric CO_2 , $\delta^{13}\text{C}$, and $\Delta^{14}\text{C}$ following Meinshausen et al. (2017) and Graven et al. (2017).

In the code provided by NCAR, atmospheric ^{14}C boundary values are read from input files. However, atmospheric boundary values are expected in units of $\delta^{14}\text{C}$ for the ^{14}C tracer in POP2, whereas atmospheric boundary values are expected in units of $\Delta^{14}\text{C}$ for the abiotic ^{14}C tracer in POP2 and by CLM5. We revised the code to account for this difference between $\delta^{14}\text{C}$ and $\Delta^{14}\text{C}$ of around 37‰ ($2 \cdot (\delta^{13}\text{C} + 25) \approx 2 \cdot (-6.6 + 25)$).

The land component was forced with data from the Global Soil Wetness Project (GSWP3) (Dirmeyer et al., 2006) which provides data from 1901 onwards. Spin up (SPIN_LN), control simulation (CTRL_LN), and the first 50 years of the historical period simulation (HIST_LN) were forced by repeatedly prescribing the climate data for the period 1901 to 1920. The GSWP3 climate data for 1901 to 2015 were prescribed for the same period in HIST_LN. Land use is prescribed following the Land Use Harmonized version 2 data set (Hurtt et al., 2020).

The ocean model was forced with the Coordinated Ocean Research Experiments (CORE.v2) Normal Year Forcing (NYF; Large and Yeager (2009)) during simulation SPIN_OC, CTRL_OC, and HIST_OC. Climate data from the Japanese Reanalysis project (JRA55, Kobayashi et al. (2015)) were used in a sensitivity simulation over the historical period (HIST_OC_JRA) and a corresponding control run (CTRL_OC_JRA). The NYF and JRA55 data allow us to investigate the influence of two different wind products and related changes in the piston velocity on simulated bomb ^{14}C . NYF data are repeated annually. The JRA55 data cover the period from 1958 onwards. The JRA55 data capture the influence of global warming while allowing us to prescribe a relatively constant climate in CTRL_OC_JRA and for the pre-1958 period in HIST_OC_JRA. Specifically, the first twenty years of JRA55 (1958 to 1977) are applied repeatedly in the simulations CTRL_OC_JRA. Similarly, the first twenty years of JRA55 data are applied repeatedly during the years 1850 to 1957 in HIST_OC_JRA, followed by the JRA55 data for the post-1958 period. The global mean surface air temperature in JRA55 varies within $\pm 0.2^\circ\text{C}$ in the control and the pre-1958 period of HIST_OC_JRA. This is followed by global warming of about 0.7°C over the period 1958 to 2015, mainly realized after 1989, in HIST_OC_JRA.

SPIN_LN was started using the initial files downloaded from NCAR and run for 750 years. CTRL_LN and HIST_LN were continued from the end of SPIN_LN. The remaining model drifts in global carbon and radiocarbon are small after the spin-up. Global vegetation carbon remains constant and the global soil carbon inventory changes by less than 1 GtC or about 0.5‰ over the control simulation. $\Delta^{14}\text{C}$ of vegetation carbon remains constant during the control CTRL_LN. Drift in $\Delta^{14}\text{C}$ are typically modest for soil carbon ($< \pm 2\text{‰/century}$, except in northern Siberia and Canada and parts of the Sahara, where $\Delta^{14}\text{C}$ of soil carbon shows a substantial drift as these soils continue to age. The total ecosystem ^{14}C inventory decreases from 251 kmol to about 249 kmol over the control simulation.

SPIN_OC was started using the initial files downloaded from NCAR and run for 1350 years. Initial values for ^{14}C and ^{13}C are missing in the files from NCAR and these missing values were specified as follows. $\delta^{13}\text{C}$ of DIC was prescribed following the gridded, observation-based preindustrial distribution of Eide et al. (2017); $\delta^{13}\text{C}$ values for the top 200 m are missing in the Eide et al. data and were set to the values at 200 m depth. $\delta^{13}\text{C}$ of DOC is set to -20 ‰. ^{14}C of DIC is initialized using the information for abiotic $\Delta^{14}\text{C}$ in the NCAR initial files. We apply equation 1b) with $\delta^{13}\text{C}=0$ to set $\delta^{14}\text{C}_{\text{biotic}} = (\Delta^{14}\text{C}_{\text{abiotic}} + 50) / 0.95$. $\Delta^{14}\text{C}$ of DOC is set to -100 ‰.

SPIN_OC was stopped in year 675 to adjust the isotopic carbon pools of DIC and DOC to speed up equilibration. The difference in the isotopic pools between years 675 and 475 was added to the pools. In other words, the trend over years 475 to 675 was extrapolated for another 200 years. SPIN_OC was then continued for another 675 years with the updated isotopic concentrations.

At the year 1350, there were still trends in most ocean variables. Trends are typically modest in the upper ocean and more substantial in the deep ocean. The mean ocean trend in $\Delta^{14}\text{C}$ is -0.32‰ per century and horizontally-average trends are between -5‰/century in the deep Pacific and +3‰/century in the upper Pacific and smaller for other basins. The trends in the upper ocean are small in comparison to the anthropogenic ^{14}C and $\Delta^{14}\text{C}$ changes simulated in HIST_OC and HIST_OC_JRA.

2.4 Drift correction

We correct the results from historical runs (HIST_LN, HIST_OC, HIST_OC_JRA) for long-term trends by subtracting the drift from control runs (CTRL_LN, CTRL_OC, CTRL_OC_JRA). For example, the change in a variable at time t is estimated by the difference in the results from HIST_OC(t) and CTRL_OC(t). The bomb radiocarbon inventory is evaluated as follows for both the ocean and the land:

$$^{14}I_{\text{bomb}}(t) = [^{14}I_{\text{IND}}(t) - ^{14}I_{\text{IND}}(t=1945)] - [^{14}I_{\text{CTRL}}(t) - ^{14}I_{\text{CTRL}}(t=1945)], \quad (3)$$

where $^{14}I_{\text{bomb}}(t)$ represents the change in ocean ^{14}C inventory since 1945. This change is mainly driven by the ^{14}C production from atomic bomb tests but is also influenced by other small anthropogenic ^{14}C sources, as well as the impact of fossil fuel burning, land use, and climate change.

2.5 Establishing the global budget of excess ^{14}C

The transient evolution in the Earth system inventory of excess ^{14}C is estimated from the sum of the land and ocean excess ^{14}C inventories simulated by CLM5.0 and POP2 plus the tropospheric and stratospheric excess ^{14}C inventories estimated from measurements of $\Delta^{14}\text{C}(\text{CO}_2)$, $\delta^{13}\text{C}(\text{CO}_2)$, and CO_2 on atmospheric samples as described by Naegler and Levin (2009a).

The two excess ^{14}C production records of Naegler and Levin are estimated from the compilations of atomic bomb test explosions by Yang et al. (2000) or Rath (1988) and cover the bomb period up to 2004. These compilations are scaled to match an estimate of the total bomb ^{14}C inventory in the mid-1960s. The contribution of the ^{14}C release by the nuclear industry is included in these production records. The uncertainty in cumulative production up to 1980 is estimated to be around 6 kmol ^{14}C and the uncertainty in the stratospheric inventory to about 2 to 2.5 kmol ^{14}C , with additional smaller uncertainties associated with natural ^{14}C productivity, the release by the nuclear industry, and the tropospheric ^{14}C inventory (Naegler and Levin, 2009a, 2006). Any difference between the Earth system inventory and the cumulative production of excess ^{14}C represents an accumulated budget imbalance. Imbalances larger than uncertainties in the atmospheric and production data, point to a mismatch in simulated uptake of ^{14}C from the atmosphere by POP2 and CLM5.

3 Results

3.1 The global budget of bomb ^{14}C

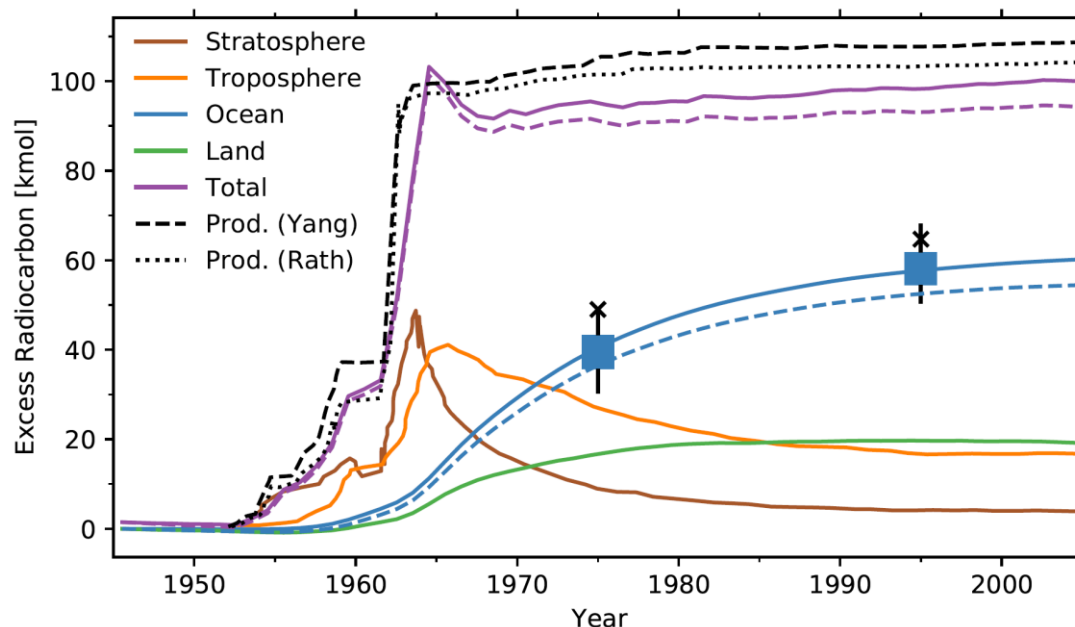


Figure 1: The Earth system budget of excess radiocarbon: observational estimates versus model results. The evolution of ^{14}C inventories are shown for the stratosphere (brown), troposphere (orange), ocean (blue), and land biosphere (green). Their total (magenta) is compared to two estimates of the excess ^{14}C production (black; dotted, dashed). Data for the ocean and land biosphere are from simulations with POP2 and CLM5. Dashed blue and dashed magenta show results obtained with the JRA55 instead of NYF in POP2. The ocean bomb ^{14}C inventories simulated by Jahn et al. (2015) are indicated by crosses. All other data are from Naegler and Levin (2009a). The range of best estimates (Sweeney et al., 2007; Peacock, 2004; Müller et al., 2008; Key et al., 2004) for the ocean inventory for 1975 and 1995 is given by the filled box and the bar represents the overall uncertainty of these estimates as summarized by Naegler and Levin (2009a). Production records are extrapolated from 2004 to 2010.

We first address the Earth system budget of bomb ^{14}C to evaluate the combined bomb ^{14}C model flux to the ocean and land. Estimates of cumulative production are compared to the Earth system inventory of excess ^{14}C in Fig. 1. The change in the Earth system ^{14}C inventory, determined by the sum of modeled ocean and land biosphere uptake plus observation-based tropospheric and stratospheric inventory changes, is generally lower than estimates of the bomb ^{14}C production (magenta versus black lines in Fig. 1).

Both cumulative production records show a steep increase to 96-99 kmol until 1963, when the bomb-test ban treaty was set in place, followed by a modest increase of 7 kmol until 1980, and

near-constant values after 1980. The estimate based on Rath remains around 4 kmol lower than the estimate based on Yang after 1963.

The stratospheric and tropospheric bomb inventory strongly increased in the 1950s to peak at 49 kmol in 1963 and 41 kmol in 1965. Afterward, the stratospheric inventory declined steeply, to 14 kmol in 1970 and 4 kmol in 2005, while the tropospheric inventory shows a more gradual decline to 17 kmol in 2005. Modeled land and ocean uptake of bomb ^{14}C are largest in the 1960s. The land inventory is 14 kmol in 1970, further increases to 19 kmol in 1980 and remains roughly stable thereafter. The ocean inventory is 30 kmol in 1970, further increases to 48 kmol in 1980 and reaches 59 kmol in 2000. The modeled inventory is lower when POP2 is forced with the JRA55 instead of the NYF and the difference between the two simulations grows to about 5.5 kmol at 2000.

The total Earth system inventory of excess ^{14}C reproduces the rapid initial increase in cumulative production of around 100 kmol ^{14}C , though with a delay of a few years. The Earth system inventory peaks in 1964 and declines in the next three years by about 10 % from 103 to 92 kmol. This is in contrast to the cumulative production records that show little changes in this period. Afterward, the inventory follows the evolution of the cumulative production records, albeit at a lower absolute value. The combined bomb ^{14}C inventory shows an offset of 4 to 7 kmol to the cumulative production record based on Rath and of about 9 kmol to the record based on Yang during the period 1970 to 2005. In this period, the change in total production (5 to 7 kmol) and the change in the Earth system inventory (7.5 kmol) remain small and comparable. Thus, the budget imbalance mainly accrues before 1970. The accumulated budget imbalance of 4 to 7 kmol (Rath) and of about 9 kmol (Yang) is also similar or larger than the uncertainty in the cumulative production record of 6 kmol. The budget imbalance is larger when the total inventory is determined using results from the POP2 simulations with JRA55 forcing (dashed, magenta in Fig. 1) instead from the simulation with NYF.

The differences between the ^{14}C production records and the Earth system inventory estimates suggest that the simulated uptake of excess ^{14}C from the atmosphere is underestimated. This is primarily the case during the 1960s when simulated ocean and land uptake is the largest. After 1970, the accumulated budget imbalance does not further increase and the combined modeled land and ocean uptake is consistent with the observational records. A too low combined uptake by POP2 and CLM5 in the 1960s could explain the delay in increase between the production and

total inventory in the early 1960s and the offset between inventory and cumulative production after 1970 when uncertainties in the atmospheric inventory become small.

We now discuss potential reasons for the decline in the Earth system inventory between 1964 and 1967 (Fig. 1 magenta line). Production from bomb tests is small during this short period. The decline by 12 kmol is either due to a too low simulated ^{14}C uptake from the atmosphere by CLM5 and/or POP2 or a too large decline in the atmospheric inventory as reconstructed from observations, or a combination of these factors. The simulated ocean uptake over these 3 years would need to be more than doubled to avoid the decline. Such a large upward revision would not be compatible with marine ^{14}C observations because modeled and observation-inferred bomb ^{14}C inventories closely agree and uncertainties in observational estimates are about 20% (Fig. 1 blue symbols and line). Simulated land uptake amounts to 5 kmol over these three years and would need to be more than tripled to avoid the decline. Therefore, it is difficult to explain the decline by shortcomings of CLM5 alone. The prescribed atmospheric inventory declines by 27 kmol from 1964 to 1967. Most of this decline is realized in the stratosphere. Uncertainties in the stratospheric inventory arise from sparse sampling and large spatio-temporal variations in stratospheric ^{14}C concentrations, while the tropospheric inventory is well known. However, the decline of 12 kmol is substantially larger than the uncertainty (2-2.5 kmol) given for the stratospheric inventory data (Naegler and Levin, 2009a, 2006). A scenario that would avoid the decline in the Earth system ^{14}C inventory and the related budget imbalance during the period 1964 to 1967 likely needs to combine a downward revision of the stratospheric decline and an upward revision of modeled CLM5 land biosphere uptake and, perhaps to a lesser extent, of POP2 ocean uptake. The inferred decline provides further evidence that the simulated ^{14}C uptake from the atmosphere is too low during the 1960s. Importantly, this evidence for too low uptake is independent of the production records and associated uncertainties.

During the 1950s, the uptake of bomb ^{14}C from the atmosphere is relatively small, because the changes in tropospheric $\Delta^{14}\text{C}$ were rather modest, and the Earth system inventory is in good agreement with the inventory record based on Rath. The small land and ocean uptake imply that remaining budget imbalances during this early period are likely due to uncertainties in the production and atmospheric data. In summary, the combined uptake by POP2 and CLM5 is too low during the 1960s, while the combined uptake over the period 1970 to 2004 is consistent with production estimates and atmospheric data.

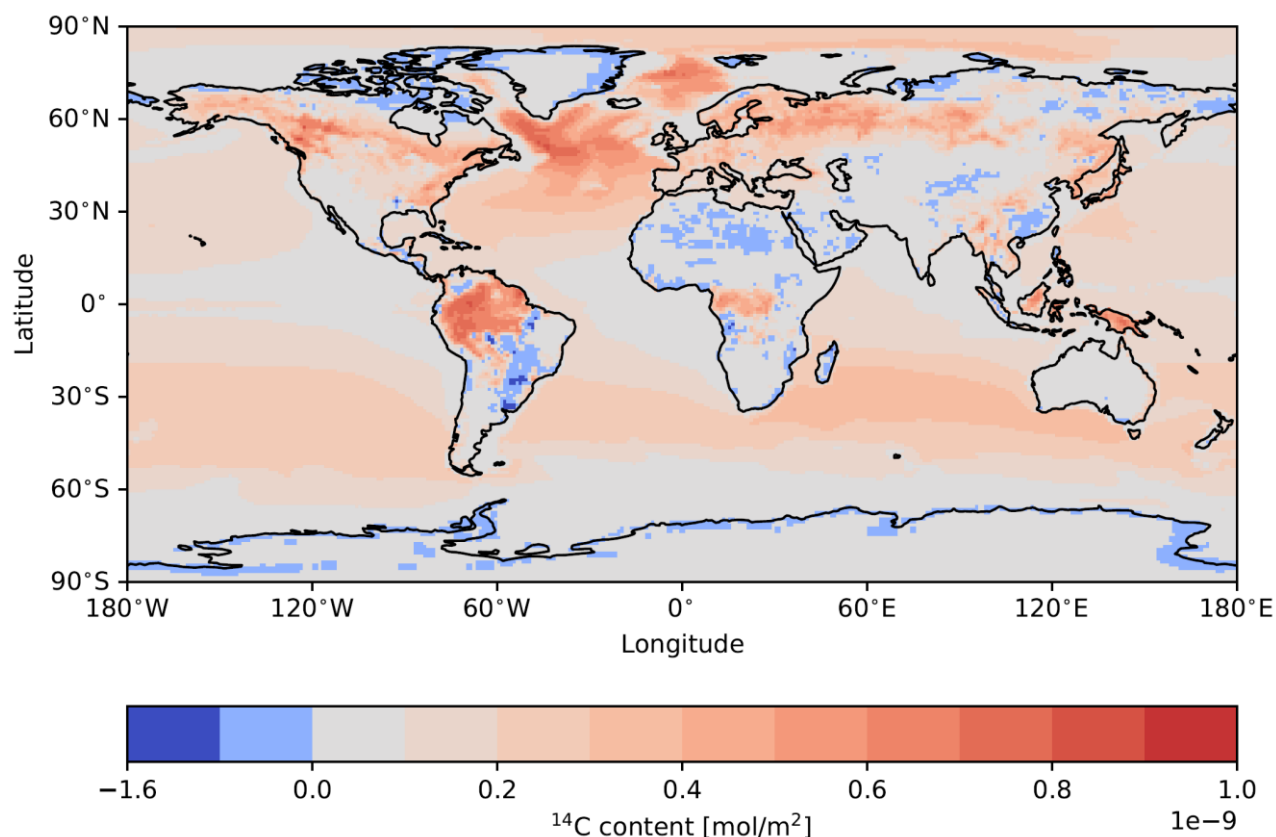


Figure 2: Changes in the ^{14}C column inventory in the ocean and on land from 1945 to 1995 simulated by POP2 with normal year forcing (HIST_OC) and CLM5 (HIST_LN).

3.2 Excess ^{14}C in the POP2 ocean model

3.2.1 The global ocean bomb ^{14}C inventory

Next, we assess whether the Earth system budget imbalances arise from ocean model biases. The global ocean bomb ^{14}C inventory simulated by POP2 agrees well with observation-based estimates (Fig.1 blue line vs symbols). POP2, driven by NYF, simulates an ocean inventory of 41.3 kmol in 1975 and 57.9 kmol in 1995. These estimates are well within the range of recent central estimates of the ocean's excess ^{14}C inventory of 36 to 44 kmol (overall uncertainty range: 30.3 kmol to 49.3 kmol) for 1975 (Müller et al., 2008; Sweeney et al., 2007; Peacock, 2004) and 54.5 to 62 kmol (overall uncertainty range: 52 to 68 kmol) for 1995 (Sweeney et al., 2007; Müller et al., 2008; Peacock, 2004). This good agreement between POP2 and observational estimates suggests that the budget imbalances discussed in the previous section are likely not caused by too low modeled ocean uptake, but rather linked to too low uptake by the land biosphere.

The good agreement between the observation-inferred inventories and POP2 results supports the representation and time scales of air-sea gas transfer and surface-to-thermocline transport on the global scale for POP2 with NYF. The air-sea flux of bomb ^{14}C is given by the piston velocity multiplied by the perturbed air-sea gradient in dissolved $^{14}\text{CO}_2$ (Eq. 2). The global ocean uptake of bomb ^{14}C before the first global survey of ^{14}C by the Geochemical Ocean Section Study (GEOSECS, 1972 to 1978) primarily depends on the magnitude of the piston velocity, whereas uncertain ocean transport has a marginal influence on the large air-sea $^{14}\text{CO}_2$ gradient during the time of the bomb peak (Müller et al., 2008; Siegenthaler, 1989). Thus, the GEOSECS bomb ^{14}C data provide a particularly strong constraint on the piston velocity. On the other hand, uncertainties in the overturning time scales within the ocean become important for the modeled ^{14}C uptake during more recent decades and the model inventory in 1995.

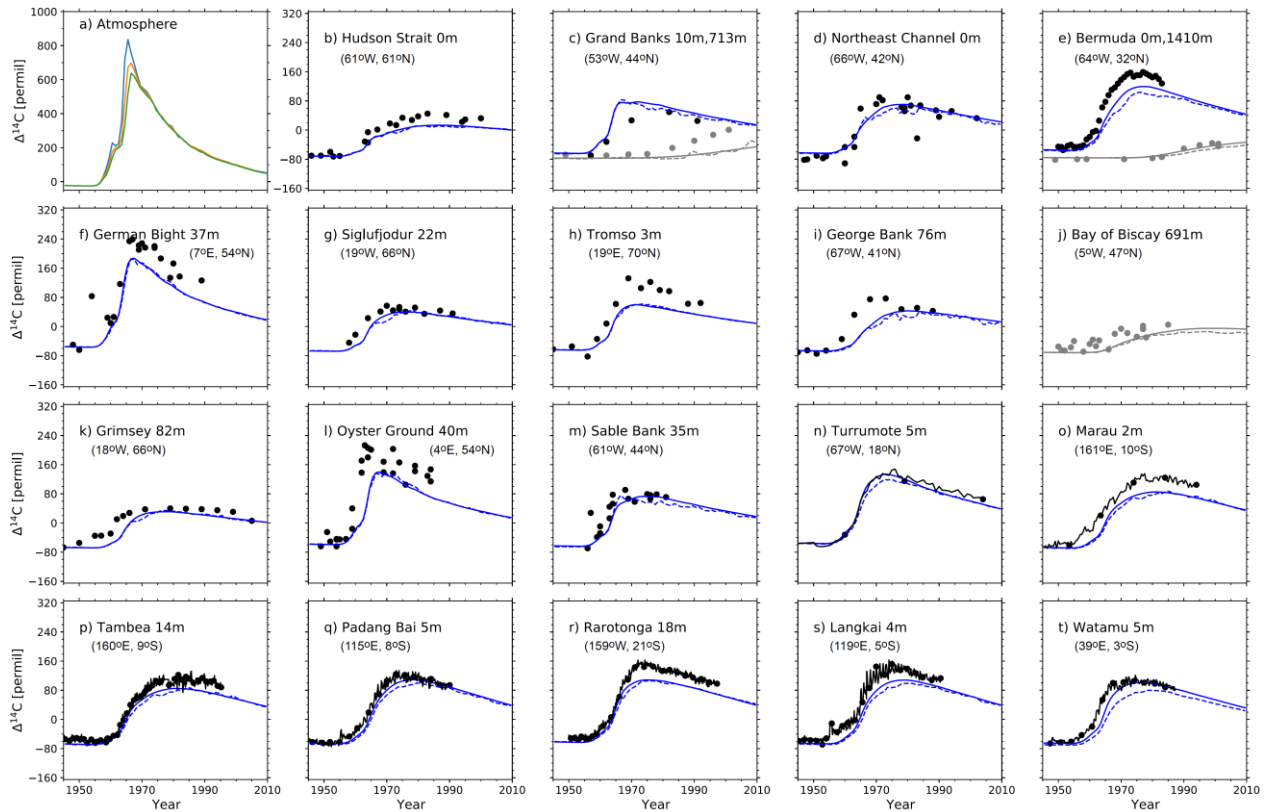
The ocean ^{14}C uptake appears biased low when POP2 is forced by JRA55 instead of NYF (Fig 1, blue dashed line). The simulated uptake is only 35.7 kmol for 1975 and 52.3 kmol for 1995. Both values are in the lower half of the observational range and at or below the central estimates from different studies. As discussed in the previous section, the budget imbalance is considerably larger when JRA55 instead of NYF is applied in POP2. These results suggest that the piston velocity in the POP2 stand-alone model version with JRA55 forcing is likely too low on the global scale, though uncertainties in the observational ocean inventory estimates remain. Further, ocean circulation is also different under JRA55 NYF and this may contribute to the low bomb ^{14}C inventory simulated for 1995 with JRA55 forcing.

Regionally, the highest column inventories of bomb ^{14}C are simulated in the North Atlantic (Fig. 2). Column inventories are also high in the mid-latitude Southern Hemisphere, where Antarctic Intermediate Water and Subantarctic Mode Water (Talley, 2013) efficiently transports excess ^{14}C to depth. Column inventories are low in the tropical upwelling regions, around Antarctica, and in the northern North Pacific.

3.2.2 The coral $\Delta^{14}\text{C}$ records

Next, we evaluate POP2 performance by comparing the simulated evolution of $\Delta^{14}\text{C}(\text{DIC})$ with $\Delta^{14}\text{C}$ records from corals and bivalves in the North Atlantic, tropical Pacific, and Indian Ocean (Fig. 3). The proxy data show a rapid $\Delta^{14}\text{C}$ increase during the 1960s in surface waters. Peak surface $\Delta^{14}\text{C}$ values are reached in the late 1960s and early 1970s, followed by a slow decline.

468 The amplitudes in surface $\Delta^{14}\text{C}$ change between pre-bomb and peak values range between 85 and
 469 250‰ (Fig. 3k, 3f).



470
 471 **Figure 3:** Simulated evolution of $\Delta^{14}\text{C}(\text{DIC})$ by POP2 (solid and dashed lines) versus $\Delta^{14}\text{C}$ records from corals and
 472 bivalves (filled circles, filled circles with solid lines for high-resolution records (n-t)). Model results are for the
 473 simulation with NYF (solid) and JRA55 (dashed) forcing and represent annual means from the grid cell including the
 474 sampling location and sampling depth (indicated in meters). Data and model results in the thermocline are shown in
 475 grey. Data are from the North Atlantic as compiled by Dentith et al. (2019), Puerto Rico, Turrumote Reef (Kilbourne
 476 et al., 2007), Solomon Islands, Marau (Schmidt et al., 2004), Solomon Islands, Tambea (Guilderson et al., 2004),
 477 Lombok Street, Padang Bai (Guilderson et al., 2009), Rarotonga (Guilderson et al., 2000), Langkai, Makassar Strait
 478 (Fallon and Guilderson, 2008), and off the coast of Kenya, Watamu (Grumet et al., 2002). The atmospheric
 479 $\Delta^{14}\text{C}(\text{CO}_2)$ evolution is shown in panel a) for the northern hemisphere ($>30^\circ\text{N}$; blue), the tropics (orange), and the
 480 southern hemisphere ($<30^\circ\text{S}$; green); note different y-axis scale.

481 The model represents the pre-bomb $\Delta^{14}\text{C}$ proxy data well and generally within the scatter of the
 482 coral data in the surface North Atlantic, with exceptions at Grand Banks (Fig. 3c) and Northeast
 483 Channel (Fig. 3d), where model data are about 20‰ higher, and at Bermuda (Fig. 3e), where
 484 model data are about 10‰ lower than most coral data. Pre-bomb $\Delta^{14}\text{C}$ simulated by POP2 is

typically about 10 to 15‰ lower than the $\Delta^{14}\text{C}$ proxy data from the Pacific and the Indian Ocean (Fig. 3o to 3t).

The increase in surface $\Delta^{14}\text{C}$ during the bomb-test period is underestimated in magnitude at all locations by POP2 compared with the proxy data. In the North Atlantic, proxy peak values are matched within 20‰ at five stations, within 40‰ at eight stations and within 75‰ at all of the 12 surface locations (Fig 3b, to 3h). Deviations in peak surface $\Delta^{14}\text{C}$ range from a few permil to up to 60‰ in the Indian and Pacific surface locations (Fig 3o to 3t). The evolution of $\Delta^{14}\text{C}$ at depth is within the scatter of the coral data at Bermuda (32°N) and a depth of 1410 m (Fig. 3e, grey) and the Bay of Biscay (46 °N) at 691m (3j) and underestimated at Grand Banks (44 °N) at 713 m (3c). Peak $\Delta^{14}\text{C}$ values from the POP2 simulation with JRA55 wind forcing are similar to those obtained with NYF at mid and high latitudes in the North Atlantic, but typically lower than for NYF at Bermuda and Puerto Rico and in the low latitude Pacific and Indian locations. $\Delta^{14}\text{C}$ at pre-bomb times and after 1990 are generally indistinguishable for the POP2 model setups.

The deviation between proxy and model data may arise due to a misrepresentation of piston velocity or ocean transport processes in POP2 or a representation bias by comparing proxy data sampled close to the coasts with grid cell values representing open ocean waters in POP2. We are not in a position to distinguish these different factors. The lower than observed pre-bomb and peak $\Delta^{14}\text{C}$ values in the low latitude Indian and Pacific may point to a too strong upwelling of thermocline waters in POP2 and/or a too sluggish gas transfer at these locations.

3.2.3 The spatial distribution of bomb $\Delta^{14}\text{C}$ in the ocean

The modeled spatial distribution of bomb $\Delta^{14}\text{C}$ is compared with observational estimates (Key et al., 2004) along a transect through the Atlantic, Southern Ocean, and Pacific (Fig. 4). The mapped observational data are derived from samples taken in the 1980s in the Atlantic, while model output is from the year 1995, representative for the Pacific and Southern Ocean data. The different sampling periods call for caution when comparing model and gridded data in the North Atlantic. The model represents the main features with shallow penetration of the bomb signal around the equator, deep penetration in the regions of intermediate and mode water masses, and intermediate penetration in the Southern Ocean.

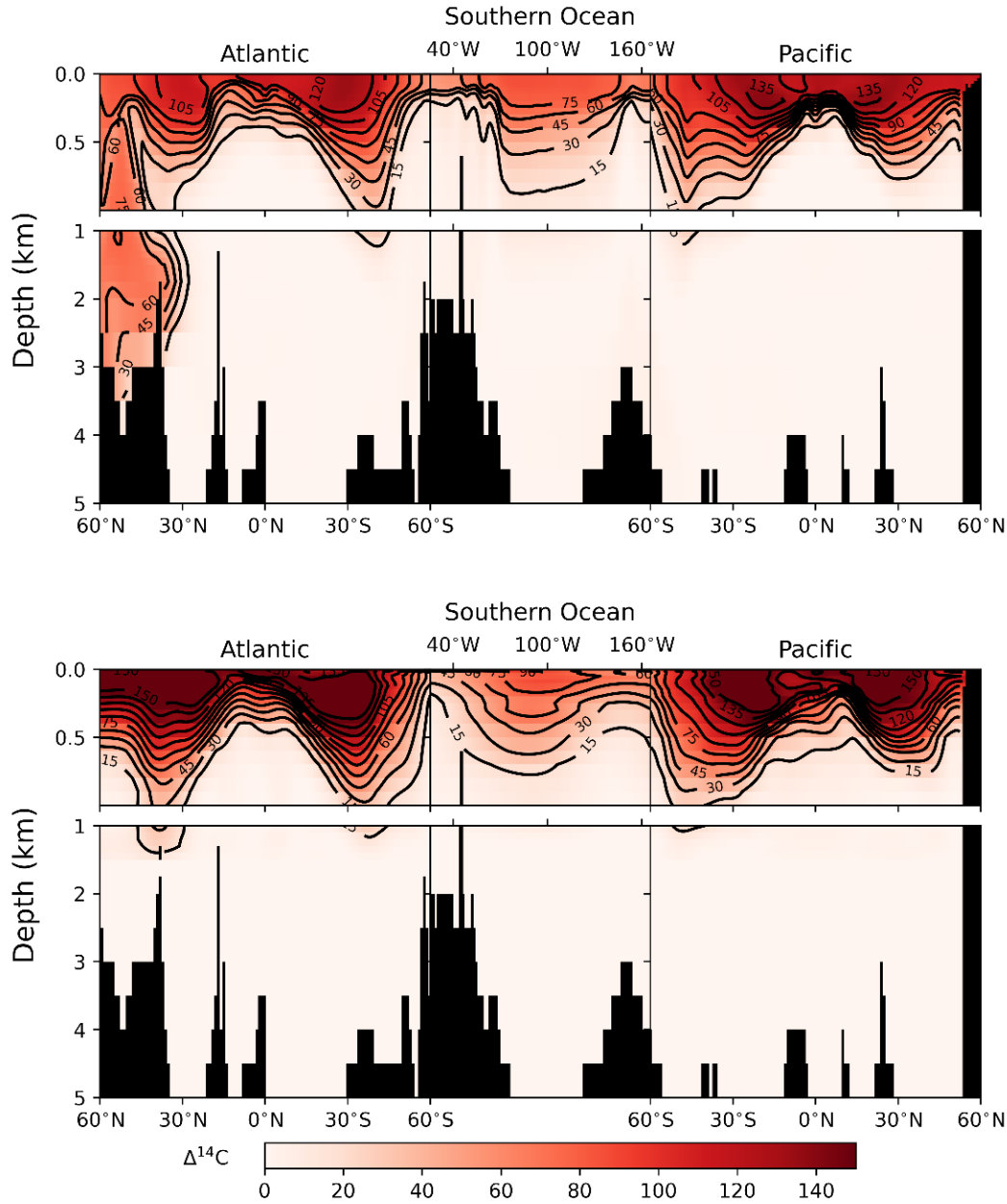


Figure 4: (a) Modeled versus (b) observation-based distribution of “bomb” $\Delta^{14}\text{C}$. Model results represent the difference in $\Delta^{14}\text{C}$ between the historical period simulation HIST_OC with Normal Year Forcing at 1995 CE and the corresponding year in the control simulation CTRL_OC. Observational data are from the gridded data product of the Global Ocean Data Analysis Project for Carbon (GLODAP) (Key et al., 2004). Results are shown for a section through the Atlantic at 25°W, through the Southern Ocean westward from 25°W to 175°W at 60°S, and through the Pacific at 175°W. Observational bomb ^{14}C maps were calculated only for the upper 1500 m by Key et al. Note that the mapping in the North Atlantic is based on samples taken between 1981 and 1983, and thus 12 to 14 years earlier than the model output.

Focusing on the 15‰ isoline, we find a good model-data agreement. The deepest occurrence of the 15‰ isoline in the South Atlantic is around 1300 m at 38°S in the GLODAP mapping and at 41°S in POP2. Its depth varies along 60°S between 160 m and 790 in GLODAP and between 160 m and 880 m in POP2. In the South Pacific, its deepest location is at 54°S and a depth of 1100 m (GLODAP) and 1300 m (POP2), respectively. The maximum and minimum depth of the 15‰ isoline is similarly matched in the North Pacific. In the equatorial Pacific, however, this isoline is simulated at 400 m in POP2 and about 200 m shallower than in GLODAP. This may point to too vigorous equatorial upwelling in POP2 in the Pacific.

The bomb $\Delta^{14}\text{C}$ signal is simulated to reach near-bottom waters around 40°N to 60°N in the North Atlantic. However, Key et al. mapped bomb $\Delta^{14}\text{C}$ only to a depth of 1500 m and from samples taken in the early 1980s, while model results are displayed for 1995. The modeled deep penetration of the bomb ^{14}C signal in the high latitude Atlantic is consistent with the observed penetration of bomb-produced tritium, CFCs, and anthropogenic carbon in this area (Perez et al., 2018; Danabasoglu et al., 2009; Schlitzer, 2007; Sabine et al., 2004; Weiss and Roether, 1980), and linked to North Atlantic Deep Water formation. It is also qualitatively consistent with the deep penetration of bomb ^{14}C reconstructed by Broecker et al. (1995).

In summary, the overall good representation of the uptake and penetration of bomb ^{14}C by the POP2 model under NYF is in line with the good agreement between observed and simulated CFC-11 concentrations documented by Danabasoglu et al. (2009). The comparison shown in Fig. 4 implies that the time scales for the ventilation of the upper thermocline are reasonably well captured by POP2. The good agreement between simulated and observation-based global ocean bomb ^{14}C inventories at 1975 and 1995 suggests that the Earth system bomb ^{14}C budget imbalance (Fig. 1) is likely not caused by deficiencies in ocean ^{14}C uptake simulated by POP2 with NYF. Rather the Earth system budget imbalance may be linked to too low uptake by the land model CLM5.

3.3 Excess ^{14}C in the CLM5 land biosphere

In this section, we investigate the evolution of the globally integrated carbon and ^{14}C inventory of vegetation and soils from 1850 to 2010 in more detail (Fig. 5a,b) and compare the simulated land biosphere inventory of excess ^{14}C with the observational estimate of Naegler and Levin (2009a) (Fig. 5b, black line, and gray shading). We also briefly address the evolution of $\Delta^{14}\text{C}$ of

vegetation and soils (Fig. 5c), and heterotrophic respiration (Fig. 5d) as well as column inventories of bomb ^{14}C on land (Fig. 2).

CLM5.0 simulates a terrestrial carbon release of 71 GtC over the period from 1850 to 1970 (Fig. 5a). Land-use emissions are larger than sink fluxes in this period. Afterward, the model simulates a terrestrial carbon sink of 48 GtC until 2010. These changes are mainly driven by changes in vegetation carbon. Soil carbon (difference between black and green line in Fig. 5a) decreased by about 5 GtC until 1970 and increased by 16 GtC from 1970 to 2010.

The simulated global land biosphere turned from a ^{14}C source into a sink around 1960 (Fig. 5b). The CLM5 land biosphere released ^{14}C in the 19th and early 20th century due to the loss of carbon, discussed in the previous paragraph, and the slight decrease in atmospheric $\Delta^{14}\text{C}$ (Fig 1d, red lines) and gained ^{14}C , mainly in response to the atmospheric bomb $\Delta^{14}\text{C}$ peak (Fig 1d). Vegetation lost 5.8 kmol of ^{14}C between 1850 and 1960, gained about 10 kmol in the next 20 years and its ^{14}C inventory remained stable thereafter. The soil ^{14}C inventory changed little (-0.26 kmol) until 1960, increased by 8.1 kmol to peak around 1995, followed by a slight decline.

Most of the land uptake of excess ^{14}C is by forest ecosystems (Fig. 2). Typical bomb ^{14}C column inventories are around $1 \times 10^{-9} \text{ mol } ^{14}\text{C m}^{-2}$ in forested areas, and of similar magnitude as the column inventories in the northern North Atlantic. Deforestation caused local reductions in ^{14}C inventories over the bomb period in several regions, including Brazil, tropical Africa, and East Asia (Fig. 2).

Naegler and Levin (2009a) analyzed uncertainties in the bomb ^{14}C budget and constructed the best estimate and the allowable range for the land biosphere bomb ^{14}C inventory for the period after 1963 (Fig. 5b, grey line and shading, right y-axis). The bomb ^{14}C inventory, i.e., the inventory change since 1945 simulated by CLM5.0 is below the lower bound of the range constructed by Naegler and Levin before 1990 and 5.3 and 5.5 kmol lower than the reference scenario of Naegler and Levin in 1970 and 1985, respectively (Fig. 5b). These results imply that the cumulative uptake of bomb ^{14}C until 1970 is underestimated by CLM5 by about 25%. A 5 kmol higher bomb ^{14}C uptake over 1945 to 1970 by CLM5 would also reconcile the imbalance in the Earth system budget discussed in section 3.1 for the period after 1970.

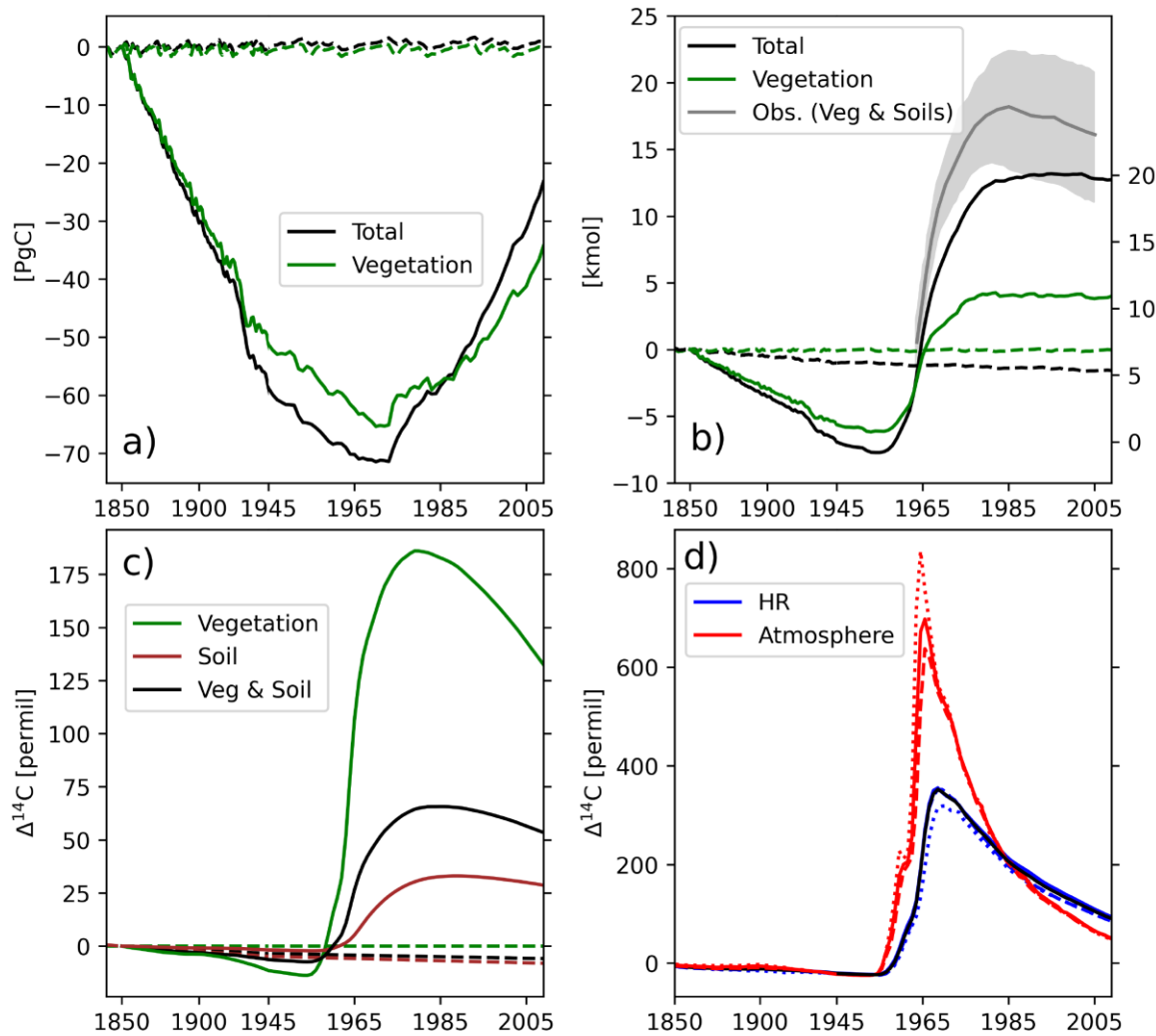


Figure 5: Changes in the global land biosphere simulated by CLM5. (a) Carbon inventory, (b) ^{14}C inventory, (c) mean $\Delta^{14}\text{C}$ of vegetation (green), soils (red), and vegetation and soils (black), (d) mean $\Delta^{14}\text{C}$ of heterotrophic respiration ('HR'; blue) and tropospheric CO_2 ('Atmosphere'; red) for 3 latitudinal belts (dotted: $>30^\circ\text{N}$; solid: 30°S - 30°N ; dash $<30^\circ\text{S}$). The right y-axis in panel b) refers to changes in the total (soil and vegetation) inventories of ^{14}C in kmol relative to 1945 and permits one to compare the ^{14}C inventories simulated by CLM5 (black) with the observation-inferred estimate of Naegler and Levin (2009a) (grey solid: their reference; shading: range spanned by their upper- and lowermost estimates). Dashed lines in (a), (b), and (c) show changes in the control simulation as used to correct for model drift. Note different x-axis scaling before and after 1945.

Soil ^{14}C data suggest that modeled ^{14}C uptake by soils is high. Shi et al. (2020) report less negative $\Delta^{14}\text{C}$ and younger soil ages in CLM5 than measured in modern soils (Shi et al., 2020; He et al., 2016). In surface soils and for 2000 CE, over 60% of carbon had positive $\Delta^{14}\text{C}$ values compared with only about 14% of carbon in the gridded dataset (Shi et al., 2020). Positive $\Delta^{14}\text{C}$

values must originate from the uptake of bomb ^{14}C . In other words, the higher than observed $\Delta^{14}\text{C}$ in surface soils suggests that the surface soils of CLM5 absorb too much bomb ^{14}C . Bringing simulated soil $\Delta^{14}\text{C}$ in agreement with soil $\Delta^{14}\text{C}$ observations, would probably lower bomb ^{14}C uptake by soils and thus widen the discrepancy between the bomb ^{14}C uptake simulated by CLM5 and the observational range given by Naegler and Levin (2009a). These considerations imply that the uptake of bomb ^{14}C by vegetation is too low.

A change in the inventory of carbon causes a corresponding change in the ^{14}C inventory. The release of carbon in the 19th and early 20th century and the carbon uptake in the late 20th century is in line with results from a deconvolution of the atmospheric CO_2 record (Joos et al., 1999; Friedlingstein et al., 2019). For the period 1970 to 2010, CLM5.0 yields a global land sink of about 48 GtC, while the Global Carbon Budget suggests a sink of 41 GtC. This difference translates into a small difference in the ^{14}C inventory of $\sim 1 \text{ kmol } ^{14}\text{C}$. However, independent evidence (Joos et al., 1999; Friedlingstein et al., 2019) suggests that the land biosphere turned into a sink already around 1940 and not only around 1970 as in CLM5. The land biosphere absorbed $6 \pm 6 \text{ GtC}$ and 10 GtC during the period 1945 to 1970 according to the observation-based estimate of Joos et al. (1999) and the results from the Global Carbon Budget (Friedlingstein et al., 2019), respectively. In contrast, CLM5.0 suggests a release of about 13 GtC in the same period. This difference between modeled and estimated carbon uptake of 19 to 23 GtC transfers in an additional uptake of $\sim 2 \text{ kmol } ^{14}\text{C}$ during the period 1945 to 1970. This additional ^{14}C would contribute to lower the discrepancy between model-based ^{14}C inventory and production estimates as discussed in sections 3.1 and would bring CLM5 results in better agreement with the reference scenario for the land inventory of Naegler and Levin (2009a) (Fig. 5b).

Simulated changes in the $\Delta^{14}\text{C}$ signatures of vegetation, soil, and heterotrophic respiration are briefly addressed. The $\Delta^{14}\text{C}$ signature of heterotrophic respiration, vegetation, and soils follows the atmospheric $\Delta^{14}\text{C}$ forcing with a delay and with muted amplitude (Fig. 5c). $\Delta^{14}\text{C}$ of heterotrophic respiration peaks at around 330‰ and a few years later than tropospheric $\Delta^{14}\text{C}$. It equals tropospheric $\Delta^{14}\text{C}$ around 1985 and remains above tropospheric $\Delta^{14}\text{C}$ thereafter. The global average $\Delta^{14}\text{C}$ of vegetation declined from -5.4 to -18.7‰ from 1850 to 1955, increased in the next 25 years to peak around +180‰ and declined by about 50‰ until 2010. The global-average bomb $\Delta^{14}\text{C}$ signals in soil carbon, including coarse woody debris and litter, is

substantially smaller than in vegetation carbon and peak $\Delta^{14}\text{C}$ is further delayed. $\Delta^{14}\text{C}$ in soil carbon increased from around -137‰ to -102‰ from 1955 to 1990. These responses in $\Delta^{14}\text{C}$ are consistent with an increase in mean age (Bolin and Rodhe, 1973) from heterotrophic respiration, to vegetation carbon, and to soil carbon.

3.4 The preindustrial distribution of $\Delta^{14}\text{C}$ in the ocean

The comparison of observed versus simulated $\Delta^{14}\text{C}(\text{DIC})$ in the surface ocean (Fig. 6 and 7) suggests that the air-sea gradient of $\Delta^{14}\text{C}$ is represented roughly in agreement with observational estimates. The global mean area-weighted surface ocean $\Delta^{14}\text{C}$ is 1.5‰ lower in the model (SPIN_OC; -67.94 ‰) than in the GLODAP data (-66.47 ‰). The root mean square deviation between model and GLODAP data is 9‰. Surface $\Delta^{14}\text{C}$ is overestimated in parts of the Pacific and Indian sectors of the Southern Ocean and underestimated in the Atlantic sector (Fig. 7). Surface $\Delta^{14}\text{C}$ is also underestimated in the northern North Atlantic and Pacific (Fig. 7).

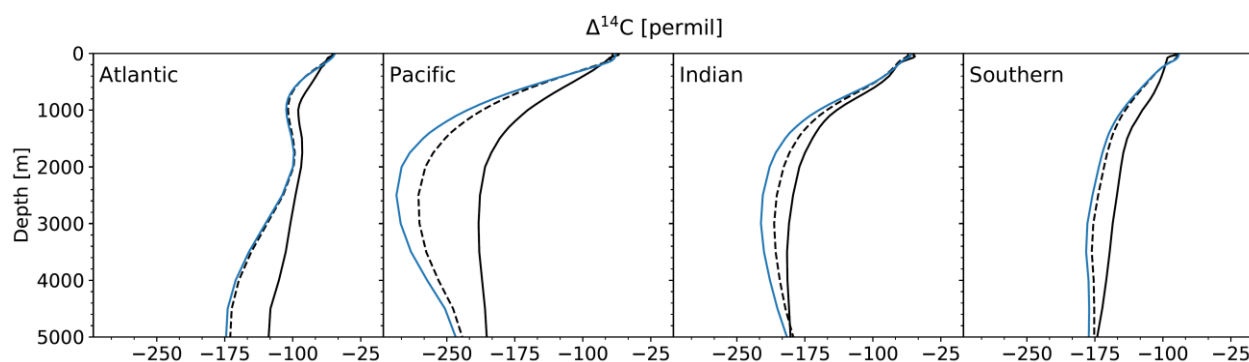


Figure 6: Simulated (black dash) versus observation-based (black solid) (Key et al., 2004) basin-mean $\Delta^{14}\text{C}(\text{DIC})$ profiles for the preindustrial period. The blue lines show the preindustrial model results for the abiotic radiocarbon tracer. The radiocarbon tracers still show some drift at the end of the spin-up which affects the simulated differences between these two tracers. The Southern Ocean includes the area south of 35°S and these areas are not included in the profiles for the Atlantic, Pacific, and Indian Ocean. The model is only sampled where GLODAP gridded data are available; the Arctic Ocean and some other smaller oceans are not covered by the GLODAP data.

In contrast to the surface, $\Delta^{14}\text{C}(\text{DIC})$ is consistently underestimated in the deep ocean by POP2 (Fig. 6, 7). Below the surface, radioactive decay of ^{14}C acts as a sink process, and $\Delta^{14}\text{C}$ decreases along the flow path of a water mass. Basin-average differences between model and observational data are about 5‰ in the upper Atlantic and grow to about 45‰ in the deepest waters. This

implies that the radiocarbon age of the deep Atlantic is overestimated by about 450 years. Even larger mean biases of around 70‰, or 770 years in age, are found at around 2000 m in the Pacific, while mean biases are smaller and less than ~25‰ (250 years) in the Indian and Southern Ocean. Ocean overturning and ventilation of the deep water masses are too sluggish in the POP2 model. This is a known model bias of POP2 as discussed by Jahn et al. (2015, and references therein)

3.5 Abiotic versus biotic ^{14}C tracers

Next, results for the abiotic versus biotic ^{14}C formulations are compared to test the validity of the simpler and less computing demanding abiotic formulation. We recall from the method section that fluxes between DIC and organic matter are neglected for the calculation of the abiotic tracer. The basin-mean profiles in the upper thermocline and the entire Atlantic show close agreement between abiotic and biotic $\Delta^{14}\text{C}$ (blue vs dashed line in Fig. 6). Mean $\Delta^{14}\text{C}$ differences in the Southern Ocean are relatively small, while substantial differences are simulated in the Indian, and in particular in the deep Pacific. $\Delta^{14}\text{C}$ in the biotic simulation is consistently less negative (younger) than in the abiotic simulations at depth. This may be expected as the remineralization of organic material adds DIC with a relatively high $\Delta^{14}\text{C}$ to the deep DIC pool. However, model drift in the deep ocean affects this comparison between the abiotic and biotic tracer at the end of the spin-up and calls for caution.

The good agreement in biotic and abiotic $\Delta^{14}\text{C}$ in the upper ocean and the Atlantic, as well as higher biotic than abiotic $\Delta^{14}\text{C}$ (Fig. 6), is in contrast to the findings of Jahn et al. (2015). These authors show biotic $\Delta^{14}\text{C}$ to be around 40‰ lower than abiotic $\Delta^{14}\text{C}$ at the surface and for 1990 CE (their Fig. 4). Biotic $\Delta^{14}\text{C}$ is also lower than abiotic $\Delta^{14}\text{C}$ throughout the water column and therefore in waters not affected by the bomb signal. It appears that the large difference between biotic and abiotic $\Delta^{14}\text{C}$ presented by Jahn et al. is explained by their atmospheric boundary condition for the biotic tracer. Too low biotic $\Delta^{14}\text{C}$ values are simulated when inadvertently prescribing atmospheric $\Delta^{14}\text{C}$ instead of atmospheric $\delta^{14}\text{C}$ to the biotic ^{14}C tracer. The difference between $\delta^{14}\text{C}$ and $\Delta^{14}\text{C}$ amounts to about 37‰ ($2 \cdot (\delta^{13}\text{C} + 25)$) for a preindustrial $\delta^{13}\text{C}$ of -6.379‰ as used by Jahn et al. (2015).

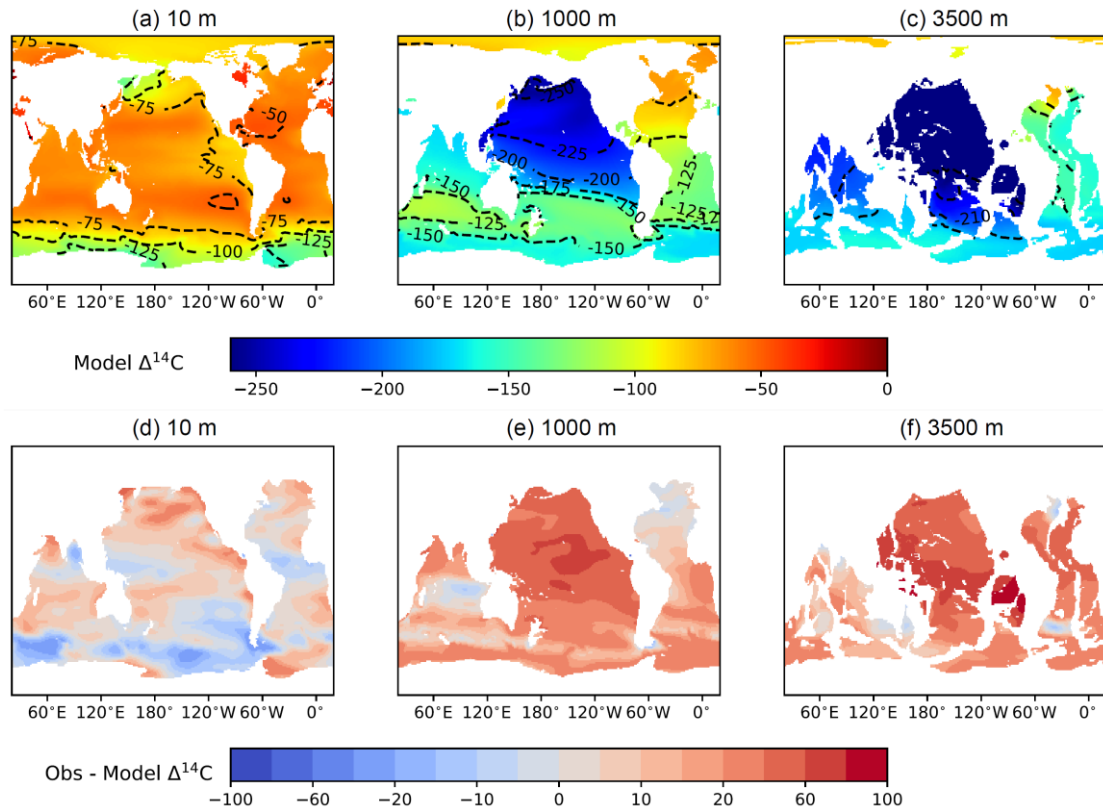


Figure 7: Simulated versus observation-based distribution of $\Delta^{14}\text{C}(\text{DIC})$ for the preindustrial period at the surface, 1000 m, and at 3500 m. Model results (top panels) are from the end of the spin-up simulation SPIN_OC. The lower panels show the difference of observation-based $\Delta^{14}\text{C}$ (Key et al., 2004) minus simulated $\Delta^{14}\text{C}$.

4 Discussion

The Earth system budget of “excess” or “bomb” ^{14}C is revisited in this study. The purpose is to evaluate the cycling of ^{14}C and underlying carbon cycle and physical and biological processes in the land (CLM5) and ocean (POP2) components of the Community Earth System Model (CESM2). We performed simulations with POP2 and CLM5 in stand-alone mode and forced with prescribed atmospheric CO_2 and $^{14}\text{CO}_2$ under preindustrial conditions and over the historical period. In addition to the standard Normal Year Forcing (NYF) of Large and Yeager (2009), we also performed a historical period simulation using the Japanese Reanalysis Data (JRA55), which implies different winds and therefore different air-sea gas exchange piston velocities and bomb ^{14}C transfer than under NYF. Imbalances in the Earth system budget of bomb ^{14}C are diagnosed by comparing the cumulative bomb ^{14}C production from bomb-test statistics and the observation-based stratospheric and tropospheric and modeled ocean and terrestrial bomb ^{14}C inventories.

4.1 A low bias in the simulated uptake of bomb ^{14}C by trees indicates biases in carbon allocation and overturning time scales in CLM5

Imbalances in the Earth system bomb ^{14}C budget accumulate mainly in the 1960s, while the production records and inventory changes are consistent after 1970 (Fig. 1, black versus magenta lines). These imbalances suggest a too-low simulated uptake of ^{14}C from the atmosphere during the 1960s. The bomb ^{14}C inventory simulated by POP2 under NYF for the GEOSCES and WOCE era is well within the range of central observation-based estimates from a range of studies (Fig.1 solid blue curve and symbols). In contrast, the inventory of bomb ^{14}C in CLM5 remains below the lower uncertainty bound of the observational estimate of Naegler and Levin (2009a) for the period 1963 to 1990 (Fig. 5b). Naegler and Levin considered uncertainties in ocean uptake, the stratospheric and tropospheric inventory, and the production rates of bomb ^{14}C to derive their uncertainty bound for the land biosphere inventory. This estimate and the Earth system budget of bomb ^{14}C both rely on the same bomb ^{14}C production records and atmospheric inventory data and therefore share some common uncertainties. The results suggest that CLM5 simulates a too-low uptake of bomb ^{14}C , mainly in the 1960s, and that this low uptake explains, at least partly, the imbalances in the Earth system budget and the deviation between simulated and observation-derived land biosphere inventory of bomb ^{14}C .

The combined inventory of bomb ^{14}C in the atmosphere and modeled by POP2 and CLM5 declined by about 10% from 1964 to 1967 in the absence of any relevant other sinks (Fig. 1, magenta). This implies that either the observation-inferred decline in the stratospheric and tropospheric inventories is too larger and/or modeled ocean and land uptakes are too low. This decline is too large to be explained by uncertainties of a single factor, but can likely only be removed by a downward revision of the atmospheric inventory decline in combination with an upward revision of CLM5, and, perhaps, POP2 bomb ^{14}C uptake.

CLM5 underestimates the net carbon sink on land during the period 1945 to 1970 and this translates into a deficit in ^{14}C uptake of about 2 kmol. Thus, a better representation of the net carbon sink in CLM5 would potentially tend to narrow the gap between the modeled and observation-based bomb ^{14}C inventory of the land biosphere.

The CLM5 results show that forests are the largest sink of bomb ^{14}C in the land biosphere (Fig. 2). This carbon reservoir is of high importance for the land sink of anthropogenic carbon (Arora et al., 2020). We argue that too low bomb ^{14}C sequestration by wood in CLM5 is mainly responsible for the low bias of the simulated global land biosphere bomb ^{14}C inventory. Grasses and shrub ecosystems have a small above-ground carbon inventory. In turn, the vegetation of these systems cannot sequester much bomb ^{14}C . Similarly, the short-turnover time and the low carbon inventory of leaves and needles and fine roots do restrict their potential for bomb ^{14}C sequestration. Soils could potentially sequester a lot of bomb ^{14}C given their large carbon stocks and slow overturning. However, $\Delta^{14}\text{C}$ in modern soils is consistently overestimated in CLM5 (Shi et al., 2020), suggesting rather a too large and not a too low uptake of bomb ^{14}C by soils in CLM5. Taken together, we conclude that the uptake of ^{14}C by forest vegetation and stem and branches is biased low. In turn, the carbon fluxes allocated to wood and/or the overturning time scales of wood carbon in forests are probably biased low in CLM5. This could potentially also be associated with a low bias in NPP of forest ecosystems

4.2 Biotic and abiotic ^{14}C in the POP2 ocean: a too slow deep ocean circulation

The POP2 ocean model simulates the highest bomb ^{14}C column inventories in the well-ventilated water masses of the North Atlantic and Antarctic Intermediate and Mode waters. The modeled penetration of bomb $\Delta^{14}\text{C}$ compares reasonably well with the gridded GLODAP data suggesting that ventilation time scales are reasonable for the global thermocline in agreement with earlier findings (Danabasoglu et al., 2009). A more thorough comparison of ocean model results with station data and along repeat sections is beyond the scope of this study.

The increase in $\Delta^{14}\text{C}$ over the bomb test period as recorded by corals and bivalves in the North Atlantic and the low latitude Indian and Pacific oceans is generally underestimated by POP2. It is unclear whether a too low piston velocity, a too rapid surface-to-deep transfer at these proxy locations, or a sampling bias is the cause of this mismatch and further studies are needed to clarify this.

Preindustrial, natural $\Delta^{14}\text{C}$ is biased low in particular in the deep ocean with the largest basin-mean age biases of order 450 years below 3000 m in the Atlantic and 770 years at 2000 m in the Pacific, similar to the results obtained by Jahn et al. (2015) with a lower resolution version of

POP2. This points to a too slow overturning circulation of the deep ocean in POP2. This large bias in deep ocean ventilation likely affects projections of atmospheric CO₂ and global warming, ocean heat uptake and sea-level rise, and ocean acidification and deoxygenation.

POP2 includes two radiocarbon tracers. For the biotic tracer, ¹⁴C fluxes related to the marine biological cycle and fractionation are considered, whereas these processes are neglected for the abiotic tracer. We find close agreement between simulated biotic and abiotic Δ¹⁴C in the upper ocean at preindustrial after accounting for differences in the implementation of atmospheric boundary conditions in POP2. This confirms that results from the simpler, abiotic Δ¹⁴C tracer are useful to evaluate ocean ventilation time scales. Larger deviations are simulated in the deep Indo-Pacific, with less negative Δ¹⁴C values for the biotic than abiotic tracer. It is not clear whether these deviations are due to the addition of ¹⁴C rich carbon by organic matter remineralization to the biotic ¹⁴C tracer or due to model drift at these deep locations.

4.3 Implications for the air-sea gas transfer piston velocity

The magnitude of the piston velocity is key to correctly simulate the ocean uptake of bomb ¹⁴C. In this study, the piston velocity is computed using a quadratic wind speed relationship (Eq. 2) and the value (0.251 cm h⁻¹ m⁻² s⁻²) of the scaling factor *a* by Wanninkhof (2014) in combination with the NYF of Large and Yeager (2009) for the spin-up and the historical period and with the JRA55 wind product for a sensitivity simulation over the historical period. The observation-based bomb ¹⁴C inventories at the time of the GEOSECS and WOCE surveys are well-matched by the POP2 simulation with NYF, lending support to use the NYF wind data together with the scaling of Wanninkhof (2014). Jahn et al. (2015) applied a 24% larger coefficient (0.31 cm h⁻¹ m⁻² s⁻²) than used here. Correspondingly, the simulated bomb ¹⁴C inventory is at the upper end of the observational range (Fig. 1, black x versus blue symbol with error bar).

The modeled ocean inventories are in the lower observational range for JRA55 and Earth system ¹⁴C budget imbalances become larger when applying the JRA55 instead of NYF wind data. The low global ocean bomb inventories and large budget imbalances suggest that a larger value of *a* should be applied than used here for the JRA55 product to simulate ocean bomb ¹⁴C uptake. We estimate a correction to *a* for use with JRA55 winds as follows. An upward correction of 4.4 kmol is required to bring the bomb ¹⁴C inventories simulated with JRA55 to agree with the

central bomb ^{14}C inventory estimate of 40.1 kmol for 1975 and of 56.7 kmol for 1995 from Naegler and Levin (2006). This would also reduce the Earth system budget imbalance after 1970 (Fig. 1, magenta dashed curve) by the same amount. We apply the sensitivities, i.e., the relative change in inventory simulated by the Bern3D model to the relative change in piston velocity (Müller et al. (2008); their Tab. 3). An increase of 16.5% and 14.3% in a is required to increase the ocean bomb ^{14}C inventory by 4.4 kmol at 1975 and 1995. This yields an estimate for a of about $2.9 \text{ cm h}^{-1} \text{ m}^{-2} \text{ s}^{-2}$. This correction is within the overall uncertainty range for the piston velocity of 20% given by Wanninkhof (2014) but larger than the uncertainty of 5% to 10% suggested by Woolf et al. (2019). Iida et al. (2020) adjusted the scaling factor a of Eq. 2 for the JRA55 wind fields by 3% from 0.251 to 0.259 based on a comparison of the Cross Calibrated Multi-Platform (CCMP) wind product used by Wanninkhof (2014) and JRA55 winds. The correction proposed here is larger, possibly because spatio-temporal variations in the air-sea $^{14}\text{CO}_2$ gradient and air-sea ^{14}C fluxes are explicitly considered in the POP2 simulation.

The correction affects the estimates of the net air-to-sea tracer fluxes derived from observations of the atmosphere-surface ocean partial pressure (or fugacity) differences. The recent global air-sea CO_2 flux estimate of $2 \pm 0.5 \text{ GtC yr}^{-1}$ for the period from 1993 to 2008 by Iida et al. (2020) is 12% higher when accounting for the higher value of a as estimated in this study. This correction brings the central estimate of the average air-sea flux by Iida et al. closer to the estimate of Gruber et al (2019) for the annual mean change in ocean carbon inventory ($2.4 \pm 0.3 \text{ PgC yr}^{-1}$) as derived from ocean interior data and for 1994 to 2008.

Watson et al. (2020) calculated air-sea CO_2 fluxes globally from the Surface Ocean Carbon Dioxide Atlas (SOCAT) and corrected for temperature differences between the depth of CO_2 sampling and the ocean surface and assuming a fixed temperature and salinity gradients across the ocean skin ($\sim 100 \mu\text{m}$). Such corrections are implicitly accounted for when the piston velocity is scaled to bring modeled and observation-derived ocean bomb ^{14}C inventories to agree.

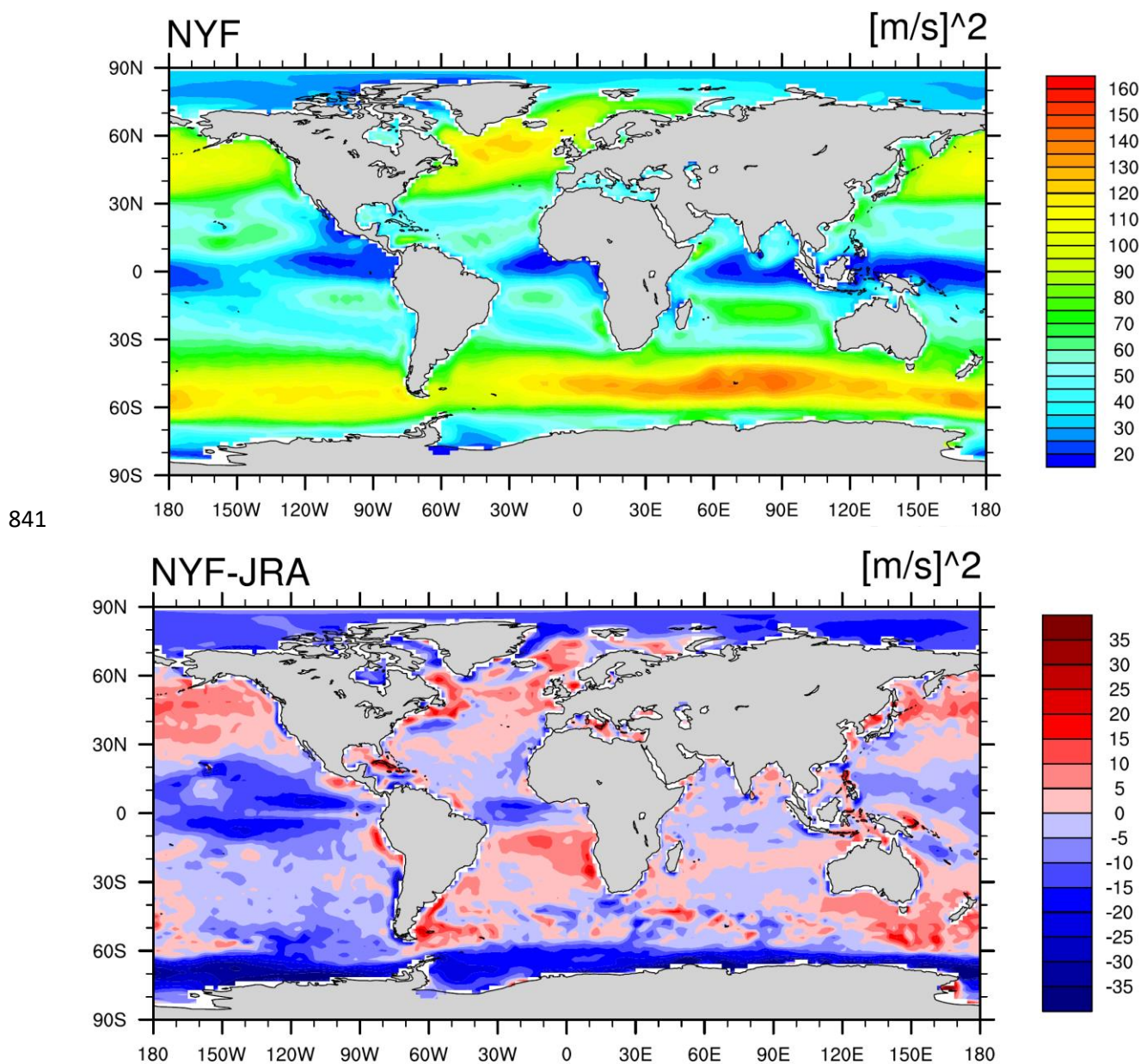
Interestingly, ocean-mean wind speed and piston velocity are higher for the JRA55 than the NYF data, despite that bomb ^{14}C inventories are lower under JRA55 forcing. This is in contrast to the expectation that a higher ocean-mean piston velocity yields also higher bomb inventories, pointing to the potential importance of spatio-temporal patterns. The global mean squared wind speed over the ocean is 8% higher for JRA55 (1958 to 1977: $67.65 \text{ m}^2 \text{ s}^{-2}$; range of annual values:

66.69 to 68.78 $\text{m}^2 \text{s}^{-2}$) than NYF (62.70 $\text{m}^2 \text{s}^{-2}$). Wind speeds of JRA55 are higher than of NYF in particular over ice-covered areas of the Arctic and the Southern Ocean, but also over large parts of the low latitude ocean and the southern Pacific (Appendix Fig. A.1). On the other hand, wind speeds of NYF are generally higher in mid-latitude areas. The global mean piston velocity normalized to a Schmidt number of 660 is 16.2 cm h^{-1} for NYF and 17.5 cm h^{-1} for the JRA55 product after applying the 3% upward correction for the ice-free ocean suggested by Naegler (2009). These values are lower but within the range of Naegler (2009) who proposes a global mean normalized piston velocity of $18.2 \pm 3.6 \text{ cm h}^{-1}$ over the ice-free ocean by summarizing estimates from four different studies (Müller et al., 2008; Sweeney et al., 2007; Krakauer et al., 2006; Naegler et al., 2006). Specifically, the piston velocities from NYF, yielding a match of simulated bomb inventories with observational estimates, are on a global average 11% lower than the central estimate of Naegler (2009). This finding suggests that the overall uncertainty in the piston velocity is larger than 10% and lower uncertainty ranges for the piston velocity may be too optimistic. The POP2 results show that differences in the spatio-temporal patterns of wind speed and, hence, of the piston velocity are, in addition to differences in global mean values, important and influence modeled global bomb ^{14}C inventories.

5. Conclusions

The analysis of bomb- and naturally-produced ^{14}C provides insight into the timescales and processes governing the global carbon cycle, atmospheric CO_2 , and climate. Our results suggest that the carbon flux allocated to wood and/or the overturning time scales of wood carbon in forests are biased low in the land biosphere model CLM5 and that the deep ocean ventilation is too slow in the POP2 ocean model. Modeled global ocean bomb ^{14}C inventories are consistent with observational estimates for the air-sea gas exchange piston velocity parameterization of Wanninkhof (2014) combined with NYF wind data. In contrast, the simulated ocean inventories are low when using the JRA55 instead NYF winds and we suggest an upward revision of the parameterization by 15% for the JRA55 product. Future efforts may be directed to adapt CLM5 and POP2 formulations to lower the identified biases in CLM5 and POP2. Another task is to simulate ^{14}C in fully coupled Earth system models with natural (Masarik and Beer, 2009; Kovaltsov et al., 2012) and anthropogenic ^{14}C sources prescribed in the atmosphere. This may enable further testing of transport time scales of Earth system models used for future projections of atmospheric CO_2 and climate.

840 Appendix A: Wind speed



842

843 Fig. A1: Annual mean of the wind speed squared at 10 m as used to compute the air-sea gas
 844 exchange piston velocity for (top) the Normal Year Forcing (NYF) and (bottom) the difference
 845 between NYF minus Japanese Reanalysis (JRA55) wind data. JRA55 winds are averaged over
 846 the period from 1958 to 1977; NYF data cover one year.

847

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The model code for CESM2 is publicly available:

https://www.cesm.ucar.edu/models/cesm2/release_download.html. ^{14}C records are available on the NOAA data server: <https://www1.ncdc.noaa.gov/pub/data/paleo/archive/>. The data used to produce the figures will be made available on a repository.

FJ wrote the paper and designed the study with input from all authors and based on the master thesis of TF. TF and AE carried out the simulations. TF prepared the figures.

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