Abstract. The isotopic composition of carbon (Δ14C and δ13C) in atmospheric CO2 and in oceanic and terrestrial carbon reservoirs is influenced by anthropogenic emissions and by natural carbon exchanges, which can respond to and drive changes in climate. Simulations of 14C and 13C in the ocean and terrestrial components of Earth System Models (ESMs) present opportunities for model evaluation and for investigation of carbon cycling, including anthropogenic CO2 emissions and uptake. The use of carbon isotopes in novel evaluation of the ESMs’ component ocean and terrestrial biosphere models and in new analyses of historical changes may improve predictions of future changes in the carbon cycle and climate system. We compile existing data to produce records of Δ14C and δ13C in atmospheric CO2 for the historical period 1850-2015. The primary motivation for this compilation is to provide the atmospheric boundary condition for historical simulations in the Coupled Model Intercomparison Project 6 (CMIP6) for models simulating carbon isotopes in the ocean or terrestrial biosphere. The data may also be useful for other carbon cycle modelling activities.

1 Introduction

The isotopic composition of carbon in atmospheric, oceanic and terrestrial reservoirs has been strongly perturbed by human activities since the Industrial Revolution. Fossil fuel burning is diluting the proportion of the isotopes 14C and 13C relative to 12C in atmospheric CO2 by the addition of aged, plant-derived carbon that is partly depleted in 13C and entirely depleted in 14C. This process is referred to as the Suess Effect following the early observations of radiocarbon in tree rings by Hans Suess (Suess, 1955; Revelle and Suess, 1957). The term Suess Effect was also later adopted for 13C (Keeling, 1979). The magnitudes of the atmospheric 14C and 13C Suess Effects are determined not only by fossil fuel emissions, but also by carbon exchanges with the ocean and terrestrial reservoirs and the residence time of carbon in these reservoirs, which regulate the mixing of the fossil fuel signal out of the atmosphere (Stuiver and Quay, 1981; Keeling, 1979). In addition, some biological and physical processes cause isotopic fractionation and the associated fractionation factors can vary with environmental conditions. Land use changes can also influence carbon isotope composition (Scholze et al., 2008). Variations in 13C are reported as δ13C, which represents deviations in 13C/12C from a standard reference
material (VPDB). For $^{14}$C, the notation $\Delta^{14}$C is used, which represents deviations from the Modern standard $^{14}$C/$^{12}$C ratio and includes a correction for mass-dependent isotopic fractionation based on $\delta^{13}$C as well as a correction for $^{14}$C radioactive decay of the sample (Stuiver and Polach, 1977).

In addition to the perturbation from fossil fuel emissions, atmospheric $\Delta^{14}$C was also subject to a large, abrupt perturbation in the 1950s and 60s when a large amount of $^{14}$C was produced during atmospheric nuclear weapons testing. The introduction of this “bomb $^{14}$C” nearly doubled the amount of $^{14}$C in the Northern Hemisphere troposphere, where most of the tests took place (Rafter and Fergusson, 1957; Münnich and Vogel, 1958). Most testing stopped after 1962 due to the Partial Test Ban Treaty, after which tropospheric $\Delta^{14}$C decreased quasi-exponentially as bomb $^{14}$C mixed through the atmosphere and into carbon reservoirs in the ocean and terrestrial biosphere that exchange with the atmosphere on annual to decadal timescales (Levin and Hesshaimer, 2000).

Sustained, direct atmospheric measurements of $\Delta^{14}$C in $\text{CO}_2$ began in 1955 in Wellington, New Zealand, capturing the dramatic changes over the weapons testing period (Rafter and Fergusson, 1957; Manning et al., 1990; Currie et al., 2009). Observations of $\Delta^{14}$C at several more stations started in the late 1950s, with some ceasing operation by the 1970s (Nydal and Lövseth, 1983; Levin et al., 1985). For $\delta^{13}$C in $\text{CO}_2$, sustained flask sampling programs began in 1977-78 at the South Pole (Antarctica), Christmas Island, and La Jolla and Mauna Loa (USA) (Keeling et al., 1979; Keeling et al., 2001), and in 1978 at Cape Grim (Australia) (Francey and Goodman, 1986; Francey et al., 1996). A global network for $\Delta^{14}$C measurements is currently run by Heidelberg University (Levin et al., 2010), while global networks for $\delta^{13}$C measurements are run by Scripps Institution of Oceanography (SIO) (Keeling et al., 2005), the Commonwealth Scientific and Industrial Research Organisation (CSIRO) (Allison and Francey, 2007), and jointly by the University of Colorado Institute for Arctic and Alpine Research and the National Oceanic and Atmospheric Administration (referred to here as NOAA) (Vaughn et al., 2010). Several other groups are also conducting long-term isotopic $\text{CO}_2$ observations at individual sites or in regional networks.

Records of atmospheric $\Delta^{14}$C and $\delta^{13}$C have been extended into the past using measurements of tree rings and of $\text{CO}_2$ in air from ice sheets (ice cores and firn) respectively; recent examples include Reimer et al. (2013) and Rubino et al. (2013). Ice cores are generally not used to construct atmospheric $\Delta^{14}$C records due to in situ $^{14}$C production, and tree rings are generally not used to construct atmospheric $\delta^{13}$C records due to climatic and physiological influences on $^{13}$C discrimination. These records clearly show decreases in $\Delta^{14}$C and $\delta^{13}$C due to increased emissions of fossil-derived carbon following the Industrial Revolution and carbon from land use change. Ice cores, and tree ring and other proxy records (e.g. lake macrofossil, marine foraminifera, coral and speleothem records) additionally reveal decadal to millennial variations associated with climate and carbon cycle variability, and, for $^{14}$C, changes in solar activity and the geomagnetic field (Damon et al., 1978).

Studies using $\Delta^{14}$C and $\delta^{13}$C observations of carbon in the atmosphere, ocean and terrestrial biosphere together with simulated $^{14}$C and $^{13}$C dynamics in models can provide insights to key processes in the global carbon cycle including air-sea gas exchange, ocean mixing, water use efficiency in plants, and vegetation and soil carbon turnover rates. Ocean $\Delta^{14}$C observations have been separated into “natural” and “bomb” $^{14}$C components (Key et al., 2004) and combined with models to constrain the global air-sea gas exchange velocity (Naegler, 2009;
Sweeney et al., 2007), and to constrain or identify biases in ocean model transport and mixing (Oeschger et al., 1975; Matsumoto et al., 2004; Khatiwala et al., 2009). Observations of δ¹³C in ocean dissolved inorganic carbon have been used to investigate anthropogenic CO₂ uptake (Quay et al., 2003) and to evaluate ocean models that include marine ecosystem dynamics (Tagliabue and Bopp, 2008; Schmittner et al., 2013). With terrestrial biosphere models, simulations of the response of plants and photosynthesis to rising atmospheric CO₂ and changing water availability can be evaluated with δ¹³C observations in atmospheric CO₂ or in leaves or tree rings, because a close relationship exists between processes controlling leaf-level isotopic discrimination and water-use efficiency (Randerson et al., 2002; Scholze et al., 2008; Ballantyne et al., 2011; Keller et al., 2017; Keeling et al., 2017). Additionally, observations of Δ¹⁴C can be used to constrain models of carbon turnover rates in vegetation and soil carbon at plot-level and global scales (Trumbore, 2000; Naegler and Levin, 2009).

The Coupled Model Intercomparison Project phase 6 (CMIP6, Eyring et al., 2016) is leading the coordination of current global earth system modelling activities. CMIP6 follows the previous phase CMIP5 that contributed to the Intergovernmental Panel on Climate Change’s Fifth Assessment Report (IPCC, 2013). CMIP6 is organizing common standards for reporting of model output and protocols for a set of core experiments including historical simulations and for several additional specialized experiments. The specialized experiments focus on individual processes or time periods and they are referred to as CMIP6-endorsed MIPs, which are organized by separate committees. Ocean MIP (OMIP) focuses on historical ocean physics and biogeochemistry and provides a separate set of simulation protocols including climatic forcing provided by atmospheric reanalyses (Griffies et al., 2016; Orr et al., 2017). The Coupled Climate–Carbon Cycle MIP (C4MIP) encompasses historical, future, and idealized biogeochemical simulations in both the ocean and the terrestrial biosphere, using climatic forcing from coupled ESMs as opposed to observations (Jones et al., 2016). For any historical simulations in CMIP6 that use observed atmospheric greenhouse gas concentrations to drive the Earth system models, compiled records of atmospheric CO₂ and other greenhouse gas concentrations are provided by Meinshausen et al. (2017). Here we describe a compilation of historical data for carbon isotopes in atmospheric CO₂ to support the inclusion of carbon isotope modelling in CMIP6. The carbon isotope datasets are provided in Table S1 and available at input4MIPs (Graven et al. 2017ab): https://esgf-node.llnl.gov/search/input4mips/.

By providing atmospheric datasets for Δ¹⁴C and δ¹³C in CO₂ as part of CMIP6, we hope to stimulate more activity in carbon isotope modelling. So far, the inclusion of carbon isotopes in large-scale models and model intercomparisons has been limited. Carbon isotopes were not included in CMIP5, the previous phase of coupled model intercomparison. One study, the Ocean Carbon Cycle Model Intercomparison Project 2 (OCMIP2), used simulations of ocean ¹⁴C to evaluate modelled ocean circulation and its effects on simulated anthropogenic CO₂ uptake and marine biogeochemistry (Orr et al., 2001; Matsumoto et al., 2004). Model intercomparisons for ¹³C in the ocean, and for both ¹³C and ¹⁴C in the terrestrial biosphere have not been performed. This may be partly a result of the small number of carbon cycle models presently simulating carbon isotopes, although some simulations with global models that don’t explicitly include carbon isotopes have been possible by using off-line isotope models (Joos et al., 1996; Thompson and Randerson, 1999; Graven et al., 2012a; He et al., 2016).

In this paper, we first review how carbon isotopes are being included in the protocols for CMIP6, which are described in more detail in Orr et al. (2017) and Jones et al. (2016). We then describe the historical atmospheric
datasets we compiled for $\delta^{13}C$ and $\Delta^{14}C$ in CO$_2$. We refer to the compiled datasets as “forcing datasets” to emphasize that (i) they are intended for model input data, (ii) atmospheric observations have been used to calculate annual and spatial averages, and (iii) some observations have been adjusted as described below.

Because of these modifications, the forcing data are not intended to be used in atmospheric inversions. For that purpose we direct modellers to the original atmospheric observations used to produce the forcing datasets, available through data repositories listed in Table 1. The firm and ice core data, updated from Rubino et al. (2013), are available in Table S2.

2 Historical Simulations of Carbon Isotopes in CMIP6

For CMIP6, carbon isotopes are included in historical biogeochemical simulations as part of OMIP (Ocean MIP (Orr et al., 2017)) and C4MIP (Coupled Climate–Carbon Cycle MIP (Jones et al., 2016)). Carbon isotopes will also be included in the simulation of future scenarios. In a separate paper, we will provide atmospheric $\Delta^{14}C$ and $\delta^{13}C$ for future scenarios in CMIP6 created with a simple carbon cycle model (Graven, 2015) and CO$_2$ emission and concentration scenarios from ScenarioMIP (O’Neill et al., 2016).

The CMIP6 simulation protocols for carbon isotopes are provided in detail in Orr et al. (2017) and Jones et al. (2016), so only a short summary is given here. The variables requested for CMIP6 are stocks and fluxes of $^{14}C$ and $^{13}C$ from any model including $^{14}C$ or $^{13}C$ in the land or ocean component. Stocks and fluxes of $^{14}C$ should be reported with a normalization factor of 1/Rs where Rs is the standard $^{14}C$/C ratio, 1.176×10$^{-12}$ (Karlen et al., 1965) whereas $^{13}C$ should be reported without normalization. For the ocean, the variables requested are the net air-sea fluxes of $^{14}C$ and $^{13}C$ and the dissolved inorganic $^{14}C$ and $^{13}C$ concentration (Jones et al., 2016; Orr et al., 2017). Models simulating dissolved inorganic $^{13}C$ concentration typically include $^{13}C$ in their marine ecosystem model (Tagliabue and Bopp, 2008; Schmittner et al., 2013) because the oceanic $\delta^{13}C$ distribution is strongly affected by marine productivity and organic matter remineralization. Models can simulate $^{14}C$ as an abiotic variable with corresponding abiotic carbonate chemistry (Orr et al., 2017) because the oceanic $\Delta^{14}C$ distribution is largely insensitive to biological activity (Fiadeiro, 1982), although some models might also include $^{14}C$ in their marine ecosystem model (Jahn et al., 2015). For the terrestrial biosphere, $^{14}C$ and $^{13}C$ fluxes associated with gross primary productivity, autotrophic respiration and heterotrophic respiration are requested. Stocks of $^{14}C$ and $^{13}C$ in vegetation, litter and soil should also be reported.

Expected uses for historical carbon isotope simulations in CMIP6 include the evaluation of modelled ocean CO$_2$ uptake and transport and carbon cycling in marine ecosystems, the evaluation of modelled carbon fluxes and stocks in terrestrial ecosystems and the ecosystem responses to higher CO$_2$ and ecohydrological changes, and the interpretation of atmospheric data. Including carbon isotopes in CMIP6 may also prepare for and motivate more activity in carbon isotope modelling in future work.

3 Historical atmospheric forcing dataset for $\Delta^{14}C$ in CO$_2$

We compiled historical data for $\Delta^{14}C$ in CO$_2$ from tree ring records and atmospheric measurements to produce the historical atmospheric forcing dataset. We use the data to estimate annual mean values for three zonal bands representing the Northern Hemisphere (north of 30°N), the Tropics (30°S-30°N) and the Southern Hemisphere...
(south of 30°S), shown in Figure 1. Here we describe the datasets used in the compilation, and a summary of datasets used in different time periods is given in Table S3.

For 1850-1940, we use estimates of Δ¹⁴C in CO₂ from previous compilations of tree rings and other records that define the calibration curves used for radiocarbon dating. Separate estimates have been made for the Northern Hemisphere, IntCal13 (Reimer et al., 2013), and for the Southern Hemisphere, SHCal13 (Hogg et al., 2013). Linear interpolation was used to estimate annual values from data with 5-year resolution provided by IntCal13 and SHCal13. We estimate Δ¹⁴C in the Tropics as the average of the Northern and Southern Hemispheres for 1850-1940. This estimate is consistent with annual tree ring measurements from 22°S in Brazil over 1927-1940 (Santos et al., 2015), which are 2.2 ± 2.5 ‰ lower than the average of Northern and Southern Hemisphere Δ¹⁴C. We did not find tropical tree ring data available for the period 1850-1927, but measurements from northern Thailand in an earlier period 1600-1800 were bracketed by measurements from New Zealand and USA (Hua et al., 2004), suggesting the average of Northern and Southern Hemisphere Δ¹⁴C is likely to provide a reasonable estimate of tropical Δ¹⁴C.

For the period between 1940 and 1954, we set Δ¹⁴C in the Southern Hemisphere and Tropics to be the same as the Northern Hemisphere Δ¹⁴C, where Northern Hemisphere Δ¹⁴C is given by IntCal13 over 1940-50 and by tree ring data from Stuiver and Quay (1981) over 1951-54. We therefore fix the spatial Δ¹⁴C gradients at 0 ‰ over this period 1940-55. This approach is motivated by differences between the Southern Hemisphere Δ¹⁴C in 1950 in SHCal13 and another compilation of tree ring data by Hua et al. (2013). In SHCal13, Southern Hemisphere Δ¹⁴C becomes 4 ‰ higher than Northern Hemisphere Δ¹⁴C over 1940-50, after being similar to Northern Hemisphere Δ¹⁴C over 1915-40. However in Hua et al. (2013), Southern Hemisphere Δ¹⁴C is only 1 ‰ higher than Northern Hemisphere Δ¹⁴C, north of 40°N, in 1950. Tree ring measurements from Brazil in 1940-1954 (Santos et al., 2015) are also consistent with Northern Hemisphere Δ¹⁴C in IntCal13. There is no significant difference between Northern Hemisphere Δ¹⁴C in IntCal13 and Hua et al. (2013) in 1950.

For Southern Hemisphere Δ¹⁴C over 1955-2015, we use direct measurements of atmospheric Δ¹⁴C, primarily the measurements conducted by Heidelberg University (Levin et al., 2010, Levin et al. unpublished). However, we use data from 1955 through 1983 made by the Rafter Radiocarbon Laboratory at Wellington, New Zealand, to specify Δ¹⁴C in the Southern Hemisphere for 1955-83. A correction of -4 ‰ is added to the Wellington data, as reported in Manning and Melhuish (1994), to account for a systematic difference between the Wellington and Heidelberg laboratories. For 1984-2014, data from Heidelberg University (Levin et al., 2010) from Neumayer (Antarctica), Cape Grim (Australia) and Macquarie Island (Australia) are averaged, if available, and where there is missing data the following procedure is used. If Macquarie Island data are missing, averages from Neumayer and Cape Grim are adjusted by -1.2‰, the average difference in mean Δ¹⁴C across the three sites when available Macquarie Island data are included or not. This adjustment takes into account that Δ¹⁴C observed at Macquarie Island is lower than the stations further north and south, resulting from gas exchange over the Southern Ocean (Levin and Hesshaimer, 2000). Macquarie Island data are available for 1993-1999, 2003, 2007-09 and 2011. For 2015, only Neumayer data were available, so the annual mean at Neumayer was adjusted by -2‰, which is the mean difference between Neumayer and the calculated Southern Hemisphere average for 2010-14.
For the Northern Hemisphere, Δ14C for 1955 to 1958 is based on tropospheric data compiled by Tans (1981). From 1959-1984, Δ14C observations from Vermunt, Austria are used (Levin et al., 1985). For the years 1985-86, only a few observations from Vermunt are available. Observations by Heidelberg University from Izaña began in 1984 and from Jungfraujoch, Switzerland in 1986. Sampling at Alert, Canada started in 1989. Even though Izaña is located at 28°N, slightly south of the 30°N bound, we use data from Izaña to specify Northern Hemisphere Δ14C for the period 1985-88 when very little data are available, after correcting for the mean difference between Izaña and the average of Izaña, Jungfraujoch, and Alert over 1989-97. For 1989-97, the average of Izaña, Jungfraujoch, and Alert is used. For 1997-2010, the average of Jungfraujoch, Alert and Mace Head, Ireland is used. From 2011-2015, only Jungfraujoch data are available (Hammer and Levin, 2017), which were used here but adjusted by +0.4‰, taking into account that Jungfraujoch is influenced slightly more by fossil fuel CO2 than Alert and Mace Head further north (see also Levin and Hesshaimer, 2000).

Observations in the tropics were made by the Heidelberg laboratory for the period 1991-97 at Mérida, Venezuela (8°N), and the annual averages at Mérida are used to specify tropical Δ14C for 1991-97. Measurements at Mérida were 2.9 ‰ higher than the Northern Hemisphere Δ14C over 1991-97, on average, and the difference (2.9 ‰) was applied to Northern Hemisphere Δ14C to estimate tropical Δ14C for the periods 1985-1991 and 1998-2015. To estimate tropical Δ14C before 1985, an atmospheric box model was used together with Northern and Southern Hemisphere Δ14C data. The atmospheric box model is part of a carbon cycle model which also simulates other atmospheric species and radioisotopes, and the model exchange parameters were optimized to match atmospheric data including Δ14C, SF6 and 10Be/7Be (Naegler and Levin, 2006). The model includes six tropospheric boxes separated at the Equator and 30° and 60° in each hemisphere. For each hemisphere, we calculated the ratio between simulated annual Δ14C averaged in the tropical boxes and simulated annual Δ14C averaged in the polar boxes. The annual average tropical-to-polar ratio was then multiplied by the observed average Northern and Southern Hemisphere Δ14C for each year to yield the values for the tropics before 1985.

A preliminary version of the Δ14C data compilation (version 1.0) was released in early 2017 via email to C4MIP and OMIP researchers and at input4MIPs (https://esgf-node.llnl.gov/search/input4mips/). The version we describe here (version 2.0, Graven et al. 2017b) incorporates new and updated Δ14C data from Heidelberg University (Hammer and Levin, 2017; Levin et al. unpublished data), whereas version 1.0 was based on fewer data and on extrapolated values for the last few years. For the Northern Hemisphere, Δ14C for 2011-15 was updated in version 2.0 but Δ14C for 2010 and earlier is the same as in version 1. For the Southern Hemisphere, Δ14C for 2000-2015 was updated. For the tropics, Δ14C for 1998-2015 was updated. Differences between version 1.0 and version 2.0 are smaller than 3.5 ‰ for individual years, and average -0.2 ‰ for the Northern Hemisphere (2011-2015), 0.0 ‰ for the tropics (1998-2015) and -1.3 ‰ for the Southern Hemisphere (2000-2015). Both versions are available at input4MIPs.

Here we make some comparisons with other reported atmospheric Δ14CO2 measurements. Organized comparisons between laboratories conducting atmospheric Δ14CO2 measurements using reference air were initiated around 2005, and results to date indicate that most laboratories are currently compatible within 2-3 ‰ (Miller et al., 2013; Hammer et al., 2016), which is similar to current measurement uncertainty but not within
the current goal of <1‰ from the World Meteorological Organization (WMO/IAEA, 2016). Whereas the compilation we report here primarily uses the measurements of the global network of Heidelberg University, continued efforts to compare Δ¹⁴C measurements from different laboratories could help to incorporate more laboratories in future data compilations.

In comparisons of the Southern Hemisphere Δ¹⁴C forcing data with observations at the South Pole, differences are less than 2 ‰ with data from SIO and Lawrence Livermore National Laboratory (LLNL) over 2000-07 and differences are approx. 5 ‰ with University of Groningen data from 1987-89 (Meijer et al., 2006; Graven et al., 2012c). Observations from Wellington, New Zealand from 1983 to 2014 also show similar trends (Turnbull et al., 2016). Differences are less than 3.5 ‰ in comparisons of the Northern Hemisphere Δ¹⁴C forcing data with annual mean Δ¹⁴C from SIO/LLNL observations at Point Barrow and La Jolla for 2002-07, and with University of Groningen observations at Point Barrow for 1987-89. The Northern Hemisphere Δ¹⁴C forcing data also compares well with observations at Niwot Ridge, Colorado, where trends of -4 to -6 ‰ yr⁻¹ have been observed since 2004 (Turnbull et al., 2007; Lehman et al., 2016). Estimated tropical Δ¹⁴C shows good agreement with observations of Δ¹⁴C at Hawaii and Samoa for 2002-07 made by SIO/LLNL (Graven et al., 2012c), with differences less than 1.5 ‰ compared to the annual averages of Mauna Loa, Kumukahi and Samoa. A limited amount of data available for Mauna Loa, Kumukahi and Samoa from 2014-15 is also consistent with the estimated tropical Δ¹⁴C. Our estimate for tropical Δ¹⁴C in the 1960s lies between observations from Ethiopia and Madagascar made at the Trondheim laboratory (Nydal and Lövseth, 1983). The Trondheim data were not used directly since no comparison between Heidelberg University and the Trondheim laboratory took place, however future studies could potentially incorporate the Trondheim data, which are available at the Carbon Dioxide Information Analysis Center (see Table 1).

A comparison with the data compilation covering 1950-2010 including tree ring and atmospheric Δ¹⁴C data by Hua et al. (2013) is shown in Fig. 2. The range in tree ring Δ¹⁴C data overlaps the Northern and Southern Hemisphere Δ¹⁴C forcing data in all periods, showing good consistency between the records. Hua et al. (2013) separate data from tropical regions over the period 1950-1972, which also overlap the CMIP6 tropical Δ¹⁴C forcing data for 1950-1972 (not shown in Fig. 2).

The data compilation shows the trends and spatial gradients in Δ¹⁴C of CO₂ over the historical period since 1850, as reported in previous studies. Between 1850 and 1952, atmospheric Δ¹⁴C decreased from approximately -4 ‰ to a minimum value around -25 ‰ as emissions from fossil fuel combustion increased after the Industrial Revolution. In the preindustrial and early industrial period to 1915, Δ¹⁴C was 3-6 ‰ lower in the Southern Hemisphere than the Northern Hemisphere due to the negative influence of CO₂ exchange with aged, ¹⁴C-depleted waters upwelling in the Southern Ocean (Braziunas et al., 1995; Rodgers et al., 2011; Lerman et al., 1970; Levin et al., 1987). Between 1915 and 1955, the interhemispheric gradient decreased due to the growth in fossil fuel emissions, which are concentrated in the Northern Hemisphere (McCormac et al., 1998). After 1955, Δ¹⁴C increased rapidly as a result of nuclear weapons testing, reaching a maximum of 836 ‰ in the Northern Hemisphere and 637 ‰ in the Southern Hemisphere, where the values 836 ‰ and 637 ‰ are the maxima in the forcing data. Δ¹⁴C was even higher in the stratosphere and some Northern Hemisphere sites (Hesshaimer and Levin, 2000). After 1963-64, tropospheric Δ¹⁴C decreased quasi-exponentially as the bomb ¹⁴C mixed with
oceanic and biospheric carbon reservoirs while growing fossil fuel emissions continued to dilute atmospheric
14CO2. Differences between the Northern and Southern Hemisphere reduced rapidly and were close to zero for
the 1980s-90s (Meijer et al., 2006; Levin et al., 2010; Levin and Hesshaimer, 2000), until the mid-2000s when a
Northern deficit in Δ14C emerged (Levin et al., 2010; Graven et al., 2012c). The Northern deficit in Δ14C has
been linked to a growing dominance of fossil fuel emissions in the Northern Hemisphere as air-sea exchange
with 14C-depleted ocean water in the Southern Hemisphere weakened with decreasing atmospheric Δ14C (Levin
et al., 2010; Graven et al., 2012c). Δ14C in background air has exhibited an average trend of about -5 ‰ yr⁻¹
since the 1990s (Graven et al., 2012b; Levin et al., 2013) and Δ14C in background air was 14-20 ‰ in 2015.

The CMIP6 forcing data for Δ14C are similar to the forcing data used in the ocean carbon cycle model
intercomparison OCMIP2 (Fig. 2), with a few notable differences. The zonal bands are defined slightly
differently: for CMIP6 we use boundaries of 30°N and 30°S whereas OCMIP2 used 20°N and 20°S. The
OCMIP2 forcing data include spatial differences over the period 1955-1968 only, for all other periods the same
Δ14C is used for all three zonal bands. Over the peak Δ14C period 1962-64 the OCMIP2 forcing data for the
tropics are 50-125 ‰ higher than CMIP6, whereas differences for the Northern and Southern Hemispheres are
smaller, less than 50 ‰. Larger differences in Δ14C in the tropics may be partly due to a lack of data in the
southern tropics over 1962-64. Measurements by Nydal and Lövseth (1983) in Madagascar (21°S) only started
in late 1964 and, compared to their observation sites in the northern tropics, the data from Madagascar show
tropical gradients up to 100‰ in 1965.

Two other periods where the CMIP6 forcing data noticeably deviate from the OCMIP2 forcing data are 1976-
82, when the OCMIP2 forcing data are 10-30 ‰ higher than the CMIP6 forcing data, and 1992-95, the OCMIP2
forcing data are approx. 10 ‰ lower. For these periods, the OCMIP2 forcing data appears to also be inconsistent
with the Hua et al. (2013) compilation (Fig. 2). The OCMIP2 forcing data ends in 1995, but the 1995 value (107
‰) is also appended for the year 2000 in the OCMIP2 forcing data, which is approx. 15 ‰ higher than the
observed Δ14C in 2000.

4 Historical atmospheric forcing dataset for δ13C in CO2:
We compiled historical data for δ13C in atmospheric CO2 from ice core and firn records and from flask
measurements to produce the historical atmospheric forcing dataset. We use the data to estimate annual mean,
global mean values for δ13C (Fig. 1, Fig. 3) as described below. We combine the most extensive dataset for firm
and ice core δ13C measurements for the period after 1850 (Rubino et al., 2013) with recent flask data from three
laboratories, including the earliest flask data currently available. This approach provides a consistent data set
that incorporates ice core data spanning the historical period and higher temporal resolution in the flask data
available starting in 1978-80.

The ice core and firm δ13C records are from Law Dome and South Pole, comprising 154 individual samples
measured by CSIRO (Rubino et al., 2013). Due to the high snow accumulation rate at Law Dome, the firm and
ice core record has high time resolution, air age distribution (68% width) of 8 years or less (Rubino et al., 2016),
and overlaps the atmospheric record. The datasets published in Rubino et al. (2013) were updated to the latest
calibration scale at CSIRO in September 2016. The calibration scale is revised from that presented in Allison &
Francey (2007) with updated corrections for cross contamination effects in the isotope ratio mass spectrometer ion source and ion correction procedures for $^{17}$O interference (Brand et al., 2010). The calibration procedure also uses a link to the VPDB reference scale established by the World Meteorological Organization Central Calibration Laboratory (CCL) for stable isotopes in CO$_2$ (Max Planck Institute for Biogeochemistry, Jena, Germany). The revised procedure ensures that all corrections are consistently applied to all samples measured at CSIRO since 1990, including all ice-core, firn air and flask measurements. We do not include measurements from 24 South Pole firn air samples conducted at NOAA reported in Rubino et al. (2013), because we could not easily quantify any NOAA laboratory offset for these measurements, relative to the current CSIRO calibration.

We use atmospheric $\delta^{13}$C measured by flask sampling from CSIRO (Allison and Francey, 2007), NOAA (Vaughn et al., 2004) and SIO (Keeling et al., 2001). Observations from SIO between 1977 and 1992 were made in collaboration with the University of Groningen, using the analytical facilities at the University of Groningen for $\delta^{13}$C measurement. After 1992, the SIO measurements were conducted solely at SIO (Guenther et al., 2001). Observations of $\delta^{13}$C by CSIRO began at Cape Grim in 1978 and expanded to a global network in the 1980s. CSIRO $\delta^{13}$C data prior to the early 1990s is not as well calibrated and therefore not publicly available except for Cape Grim, which has data available from the early 1980s. Observations of $\delta^{13}$C by NOAA and INSTAAR started in the early 1990s. Atmospheric $\delta^{13}$C data were downloaded in July 2016 from CSIRO, NOAA and SIO. Websites for data access are listed in Table 1.

Here we use atmospheric $\delta^{13}$C data from two sites, South Pole (SPO) and Mauna Loa (MLO), in order to capture interhemispheric differences in $\delta^{13}$C in defining a global mean value. These two sites are measured by all three laboratories. In order to compile data from the three laboratories, we used a third station, Alert, Canada, to assess inter-laboratory offsets, also referred to as “scale offsets”. There is ongoing work in the community to incorporate best practices for preparing and measuring reference materials, and to use CCL CO2-in-air reference materials to evaluate and resolve scale offsets in atmospheric $\delta^{13}$C data (Wendeberg et al., 2013; WMO/IAEA, 2016), however, not all currently reported data consistently account for any scale offsets between laboratories.

We adjust SIO and NOAA $\delta^{13}$C data to be consistent with CSIRO $\delta^{13}$C data using scale offsets identified in measurements from Alert. Average differences between annual mean $\delta^{13}$C observations made at Alert for 2005-2015 by CSIRO and by the Max Planck Institute for Biogeochemistry (the CCL) are less than 0.01 ‰ (WMO/IAEA, 2016), so the compiled $\delta^{13}$C record we present here can also be regarded to be consistent with the VPDB scale established by the CCL.

Comparing annual mean $\delta^{13}$C observed at Alert over 1992-2014 shows that data reported by NOAA were 0.031 ‰ higher than CSIRO, and data reported by SIO were 0.046 ‰ higher than CSIRO, on average. The standard deviation in NOAA-CSIRO differences was 0.018 ‰, and 0.020 ‰ for SIO-CSIRO differences, with standard error of 0.004 ‰ for both. Similar offsets were found in comparisons of monthly mean rather than annual mean $\delta^{13}$C at Alert, with larger standard deviations of 0.031 ‰ for monthly NOAA-CSIRO differences and 0.044 ‰ for monthly SIO-CSIRO differences, but smaller standard errors of 0.002 ‰ for both. The offset 0.031 ‰ was subtracted from NOAA data at South Pole and Mauna Loa, and 0.046 ‰ was subtracted from SIO data at South Pole and Mauna Loa. Then the monthly values from the three laboratories were averaged, and used to calculate combined annual means. As a result of varying data availability, annual means for 1977-90 at SPO and 1980-89
at MLO are based only on SIO data (with the offset applied), and the annual mean for 2015 is based only on CSIRO data.

The atmospheric data show a gradient of -0.043 ‰ between Mauna Loa and South Pole in 1980-84, growing to -0.095 ‰ in 2010-14 (Fig 3). Keeling et al. (2011) suggest that the preindustrial Northern Hemisphere – Southern Hemisphere δ¹³C gradient was +0.09 ‰, opposite in sign to the present interhemispheric gradient, using a regression of SIO δ¹³C data with global total fossil fuel emissions. They further demonstrate that the inferred preindustrial gradient is consistent with a model of spatial variation in equilibrium fractionation during air-sea gas exchange. A similar preindustrial atmospheric δ¹³C gradient was simulated by Murnane and Sarmiento (2000) using a global ocean model, where they also attributed the primary driver of the gradient to equilibrium fractionation. δ¹³C data from Greenland ice cores and possibly deep firm are compromised by in situ CO₂ production so it is not possible to discern a precise preindustrial or pre-1980 δ¹³C gradient directly from observations (Anklin et al., 1995; Francey et al., 1999; Tschumi and Stauffer, 2000; Jenk et al., 2016). We account for the possibility that δ¹³C measured in ice core and firm in Antarctica is slightly different from the global mean by using the regression from Keeling et al. (2011) to estimate δ¹³C at MLO. Then to estimate global δ¹³C we average the observed Antarctic ice core and firm δ¹³C and the estimated δ¹³C for MLO. Previous studies have adjusted Antarctic ice core and firm δ¹³C to estimate global levels by assuming that the preindustrial gradient was zero (Rubino et al., 2013).

To calculate a smoothed, global average time series for δ¹³C in CO₂ over 1850-2015, we first average replicate measurements of ice core and firm samples from Rubino et al. (2013) and then calculate annual averages for any year that includes an ice core or firm measurement. Using the annual averages, we estimate a corresponding δ¹³C at MLO from the ice core and firm data using the regression from Keeling et al. (2011). We then append the ice core and firm data before 1977 to the annual average record at SPO beginning in 1978, omitting any ice core and firm data after 1978. We similarly append the δ¹³C at MLO estimated from the ice core and firm data to the annual average record at MLO beginning in 1980, omitting any ice core and firm- based estimates after 1980 (Fig. 3). Smoothed curves were then calculated for SPO and MLO with stronger weighting on the recent flask-based data to account for the coarser time resolution of the ice core versus flask data due to diffusive smoothing in the firm. Then these curves were averaged and evaluated in the middle of each year 1850-2015 to produce the atmospheric forcing data (Fig 1).

Ice core and firm data after 1977 or 1980 were not used directly to produce the forcing data, but they are included in Fig. 3 for comparison. The differences between SPO annual means and ice core and firm data after 1977 are less than 0.05 ‰ for 14 samples, and less than 0.09 ‰ for the two other samples. The differences between MLO annual means and estimates of MLO δ¹³C from ice core and firm data after 1980 are less than 0.03 ‰ for 8 samples, and less than 0.09 ‰ for the one other sample. In addition, applying the regression to the annual mean SPO δ¹³C flask data is consistent with MLO observations to within 0.05 ‰, suggesting the regression from Keeling et al. (2011) using 1979-2003 SIO data is also consistent with combined means including the NOAA and CSIRO data, and with the longer period encompassing 2004-2015.

We also compare the global δ¹³C forcing data with the global monthly mean Marine Boundary Layer δ¹³C estimated from NOAA’s larger network of stations, which is available for 1993-2015 (Fig. 3,
www.esrl.noaa.gov/gmd/ccgg/mbl/mbl.html) (Masarie and Tans, 1995). Here the NOAA-CSIRO offset has been applied (0.031 ‰). The differences for 1993-96 are -0.04±0.01 ‰, when the NOAA Marine BL global mean is close to MLO. Differences after 1996 are smaller, -0.02±0.01 ‰. Slightly lower values in NOAA Marine BL global mean indicates low- δ13C air from the Northern Hemisphere is slightly underrepresented in the MLO-SPO average. However, the long-term trends are similar: both decreased by 0.6 ‰ between 1993 and 2015.

5 Discussion and Conclusions

We have produced a compilation of atmospheric datasets for Δ14C and δ13C in CO2 over the historical period 1850-2015 with the aim of providing a standard atmospheric boundary condition for ocean and terrestrial biosphere models simulating 14C and 13C in CMIP6. The data can be accessed in Table S1 and at input4MIPs (Graven et al., 2017ab): https://esgf-node.llnl.gov/search/input4mips/.

In compiling these atmospheric forcing datasets for Δ14C and δ13C in CO2, our primary objective was to accurately and consistently compile the data available. We also aimed to provide datasets that are simple to use, particularly as δ13C has not been included previously in a large model intercomparison. Δ14C was included previously in OCMIP2 using a similar approach and atmospheric forcing dataset (Orr et al., 1999), but has been updated and improved in this version.

Several applications for simulations of 14C and 13C may require or benefit from the use of oceanic, terrestrial and/or other atmospheric data, including atmospheric data with higher temporal or spatial resolution. Available global-scale databases are listed in Table 1, and we encourage modellers to collaborate with data providers on model-data integration studies. In particular, data users should take care to account for any δ13C scale offsets between laboratories, as described above. The compilation of various datasets for Δ14C and δ13C in CO2 for modelling and model-data integration studies in the future will benefit from ongoing efforts to compare and harmonize Δ14C and δ13C measurements from different laboratories (Miller et al., 2013; Hammer et al., 2016; Wendeberg et al., 2013; WMO/IAEA, 2016), and from additional support for such efforts.

We note that ice core and firn δ13C data updated from Rubino et al. (2013) that were used to produce the δ13C forcing data are included in Table S2. The ice core and firn CO2 data are also included in Table S2. These data could be joined with CSIRO observations at SPO, available from the World Data Centre for Greenhouse Gases (Table 1), to provide Antarctic records of CO2 concentration and δ13C measured in the same air sample by the same laboratory, which may be advantageous for some applications.

Data and code availability. The atmospheric forcing datasets for Δ14C and δ13C in CO2 can be accessed in Table S1 and at input4MIPs (Graven et al., 2017ab): https://esgf-node.llnl.gov/search/input4mips/. Original atmospheric data are available from the websites listed in Table 1, and ice core and firn δ13C data updated from Rubino et al. (2013) are included in Table S2. Interpolation and smoothing were conducted with standard routines in Matlab; further details are available from the lead author on request. A table summarizing the data sources for the Δ14C compilation is given in Table S3.
Acknowledgments. We thank the staff of the atmospheric monitoring stations for their long-term commitment to the flask sampling activities. The teams of the Law Dome and South Pole drilling expeditions provided the firm air and ice core samples. CSIRO GASLAB and ICHELAB personnel supported the measurements of air from the CSIRO monitoring network and firm and ice core samples. Logistic support was provided by the Australian Antarctic Division (Macquarie Island, Law Dome) and the Bureau of Meteorology (Cape Grim, Macquarie Island). The Australian Climate Change Science Program contributed to funding of the CSIRO measurements. Measurements at SIO were supported by the US National Science Foundation, Department of Energy, and NASA under grants 1304270, DE-SC0012167, and NNX17AE74G, by The Eric and Wendy Schmidt Fund for Strategic Innovation, and by NOAA for collection of air samples. Measurements at NOAA and INSTAAR were supported by the NOAA Climate Program Office. Measurements at Heidelberg University were partly funded by a number of agencies in Germany and Europe, namely the Heidelberg Academy of Sciences, the Ministry of Education and Science, Baden-Wurttemberg, Germany; the German Science Foundation, the German Minister of Environment; the German Minister of Science and Technology; the German Umweltbundesamt and the European Commission, Brussels, as well as national funding agencies in Australia, Canada and Spain. H. Graven received support from the European Commission through a Marie Curie Career Integration Grant.

References


Table 1. Available global-scale databases of Δ^{14}C and δ^{13}C in atmospheric CO₂, terrestrial and ocean carbon, and fossil fuel emissions.

<table>
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<td>World Data Centre for Greenhouse Gases (Including CSIRO data)</td>
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<td>Carbon Dioxide Information Analysis Center (CDIAC)</td>
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<td>Soil Carbon Database</td>
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<td><a href="https://github.com/powellcenter-soilcarbon">https://github.com/powellcenter-soilcarbon</a></td>
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Figure 1. Historical atmospheric forcing datasets for $\Delta^{14}$C in CO$_2$ (top) and $\delta^{13}$C in CO$_2$ (bottom) compiled for CMIP6. Annual mean values of $\Delta^{14}$C are provided for three zonal bands representing the Northern Hemisphere (30°N-90°N), the Tropics (30°S-30°N) and the Southern Hemisphere (30°S-90°S). Annual mean, global mean values are provided for $\delta^{13}$C. Tabulated data are provided in Table S1.
Figure 2. The $\Delta^{14}C$ atmospheric forcing data for CMIP6 compared to the forcing data used in OCMIP2 and the Northern and Southern Hemisphere tree ring and atmospheric data compilations of Hua et al. (2013) over the time period 1950-2015 (top) and over the recent period 1975-2015 (bottom). Zones NH1 and SH1-2 from Hua et al. (2013) are used, to correspond to the Northern Hemisphere north of 30°N and the Southern Hemisphere south of 30°S. Tropical data from Hua et al. (2013) are not shown, for clarity.
Figure 3. Observations of $\delta^{13}C$ and the $\delta^{13}C$ atmospheric forcing data for CMIP6 over the full time period 1850-2015 (top) and over the recent period 1975-2015 (bottom). The $\delta^{13}C$ atmospheric forcing data for CMIP6 are shown in black, as in Fig. 1. Data from SPO and ice core and firn samples are shown in blue, and data from MLO and estimated data for MLO based on ice core and firn samples and the regression from Keeling et al. (2011) are shown in purple. Measurements conducted at different laboratories are shown with different symbols. Data from NOAA and SIO have been adjusted with their average laboratory offset from CSIRO. The global mean $\delta^{13}C$ estimated by NOAA based on a larger network of flask sampling stations over 1993-2015 is shown in light blue (NOAA Marine Boundary Layer).