RADIOCARBON IN GLOBAL TROPOSPHERIC CARBON DIOXIDE

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ABSTRACT. Since the 1950s, observations of radiocarbon (14C) in tropospheric carbon dioxide (CO2) have been conducted in both hemispheres, documenting the so-called nuclear “bomb spike” and its transfer into the oceans and the terrestrial biosphere, the two compartments permanently exchanging carbon with the atmosphere. Results from the Heidelberg global network of Δ14C-CO2 observations are revisited here with respect to the insights and quantitative constraints they provided on these carbon exchange fluxes. The recent development of global and hemispheric trends of Δ14C-CO2 are further discussed in regard to their suitability to continue providing constraints for 14C-free fossil CO2 emission changes on the global and regional scale.

KEYWORDS: anthropogenic 14CO2 perturbations, bomb radiocarbon, global carbon cycle, tropospheric 14CO2 data.

INTRODUCTION

Atmospheric nuclear weapon testing in the 1950s and 1960s in the Northern Hemisphere was a period of great anxiety, however, it had significant side effects for environmental sciences in many aspects. The artificial production of more than $6 \times 10^{28}$ atoms or about 1.4 tons of radiocarbon (14C) (Naegler and Levin 2006), lead to a doubling of the 14C/C ratio in tropospheric CO2 of the Northern Hemisphere with a prominent spike in 1963 (Nydal and Lövseth 1983; Levin et al. 1985). This “bomb spike” that reached the Southern Hemisphere with some delay (Manning et al. 1990), has been used as a transient tracer in all compartments of the fast carbon cycle (e.g., Broecker et al. 1985; Levin and Hesshaimer 2000; Trumbore 2009), but also to study atmospheric dynamics, such as inter-hemispheric (e.g., Münnich and Vogel 1958, 1963; Czeplak and Junge 1974) and stratosphere-troposphere air mass exchange (e.g., Telegadas 1971; Hesshainer and Levin 2000; Levin et al. 2010). As an indirect outcome of the weapon testing, a wealth of new insights into atmospheric and carbon cycle dynamics could be achieved in the decades following the nuclear test ban treaty in 1963.

Today, the transient bomb-radiocarbon signal has levelled off, and the anthropogenic input of radiocarbon-free fossil CO2 into the atmosphere has become the dominant driver of the decrease of the 14C/C ratio in global atmospheric CO2 (Levin et al. 2010; Graven 2015). This 14CO2-free anthropogenic CO2 flux from the burning of fossil fuels and cement production has increased globally by more than fourfold compared to the 1960s, and, together with ongoing land-use changes, has resulted in an increase of the atmospheric CO2 burden by more than 5 PgC (1 PgC = 10^{15} gC) or >0.5% per year in the last decade (Friedlingstein et al. 2020). The fossil CO2 emissions are not evenly distributed over the globe and because the biosphere and oceans today, in most regions, are sources of bomb

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14CO2, the distribution of tropospheric Δ14C-CO2 has undergone significant changes since the 2000s.

After a brief overview on the development in the last 60+ years of 14C in tropospheric CO2, this paper reviews the insights from earlier studies that used bomb 14C for carbon cycle budgeting. Further, we present our extended high-precision Δ14C-CO2 observations in background air at Alert, Jungfraujoch, Izana, Cape Grim, Macquarie Island and the German Antarctic Neumayer station (Figure 1), and discuss these data in view of their potential to serve as additional constraints for global carbon fluxes. Finally, we provide a perspective on how ongoing changes of the distribution and trends of Δ14C-CO2 in the background troposphere could potentially be used to constrain major components of the carbon cycle, including monitoring of anthropogenic CO2 emission and their reductions in view of the Paris agreement from the regional to the global scale.

THE HISTORY OF ATMOSPHERIC Δ14C-CO2 OBSERVATIONS

Shortly after Anderson and Libby (1951) published their first measurements of natural radiocarbon in various compartments of the Earth system, a number of Radiocarbon Laboratories have been established world-wide. First measurements of Δ14C-CO2 in background air were conducted in 1954 in the Southern Hemisphere with sampling on the south-western coast of the Northern Island of New Zealand (NZ) close to Wellington (41.25°S, 174.69°E, 300 m asl; Rafter and Fergusson 1957). Münnich and Vogel (1963) collected their first atmospheric CO2 samples for 14C analysis in 1959 at a number of stations in the Northern and Southern Hemisphere. Quasi-continuous sampling was finally established at Vermunt in the Austrian Alps (47.07°N, 9.57°E, 1800 m asl; Levin et al. 1985). In the early years, all samples were collected by passive absorption of atmospheric CO2 in sodium hydroxide solution. CO2 was then extracted in the laboratory from this basic solution by acidification. After purification of the extracted CO2 the samples were analyzed by gas proportional counting (Kromer and Münnich 1992; Turnbull et al. 2017).
In the 1970s, advancements in both the sampling and analysis systems occurred, e.g. the Heidelberg sampling system at Vermunt station was changed to actively flushing air through a rotating absorption column, now allowing quantitative (less fractionated) sampling of CO2 over a well-defined time interval (Levin et al. 1980). At the NZ station, in addition to passive sampling, whole air flask samples started to be collected, and 14C analysis changed from gas counting to Accelerator Mass Spectrometric analysis (for details, see Turnbull et al. 2017, supplementary material). While in Heidelberg measurement of background air samples was and still is today by gas proportional counting, we changed our European background monitoring site in the Alps from Vermunt in Austria to Jungfraujoch in Switzerland (46.55°N, 7.98°E, 3450 m asl) in 1986. This seemed advantageous as the high elevation of Jungfraujoch provides better access to free tropospheric air. In addition, several continuous measurements of other atmospheric trace substances, including CO2, are conducted at this site (Forrer et al. 2000).

The two pioneer stations in New Zealand and in the Austrian Alps, which document the largest excursions of bomb 14CO2 in the Southern and the Northern Hemisphere troposphere, were supplemented by an increasing number of Δ14C-CO2 observations world-wide, including the tropics and sub-tropics. The most comprehensive records were published by Nydal and Lövseth (1983, 1996) and by Meijer et al. (1995). These globally distributed data sets provided the opportunity to track the bomb 14CO2 spike throughout the entire troposphere. Unfortunately, most of these sampling efforts were discontinued in the 1980s after bomb 14CO2 had become well-mixed in both hemispheres.

Starting in 1983, when the German Antarctic Neumayer station became operational in Dronning Maud Land at the Antarctic coast (70.65°S, 8.25°E, 17 m asl), the Heidelberg Radiocarbon laboratory was granted the opportunity to start CO2 sampling for 14C analysis at this remote Antarctic site. Soon after the first data became available, it was realized that the Δ14C-CO2 level at Neumayer station was significantly influenced by a 14CO2 disequilibrium flux with the Antarctic circumpolar surface water, originating from upwelling of about 800-year-old intermediate water of the Pacific Ocean, which, due to its age, is strongly depleted in (natural) 14C (Levin et al. 1987). This exciting feature of lower Δ14C-CO2 levels in atmospheric CO2 of Southern Hemisphere air when compared to the Northern Hemisphere had already been observed by Lerman et al. (1970) on tree rings. Their results were now confirmed by direct atmospheric observations. For us, this finding was the catalyst to re-start global monitoring of Δ14C-CO2 in the atmosphere, i.e. during a period when other research agencies were terminating their 14CO2 monitoring efforts. The potential of Δ14C-CO2 observations as an ongoing constraint of gross carbon exchange fluxes between atmosphere, ocean and biosphere seemed obvious. However, unlike the situation immediately after the test ban treaty in 1963, being 20 years later, the spatial gradients and temporal variations of Δ14C-CO2 in the atmosphere were more than two orders of magnitude smaller. To detect such small signals required increased precision in Δ14C-CO2 analysis (about 2‰), and could only be achieved by gas proportional counting at that time. In the 1980s, we begun establishing a new network of Δ14C-CO2 observations at a number of globally distributed stations, in cooperation with colleagues operating the Global Atmosphere Watch stations for continuous greenhouse gases observations (see map in Figure 1). Other investigations (Hesshaimer et al. 1994; Levin and Hesshaimer 2000), were also carried out using these data to close the atmospheric bomb 14C budget, and, in turn, determine the total input of anthropogenic 14C into the global carbon system (Naegler and Levin 2006).
In the 2000s, the Scripps Institution of Oceanography (SIO) started analyzing $\Delta^{14}C$ on CO$_2$ extracted from flask samples collected from their global network of stations along the Pacific Ocean, stretching from Barrow, Alaska, down to the South Pole (Graven et al. 2012). These samples were analyzed with high precision Accelerator Mass Spectrometry. The data were included in a global atmospheric transport model (TM3, Heimann and Korner 2003) to investigate the spatial $\Delta^{14}C$-CO$_2$ distribution and document its changes since the 1990s. Unfortunately, $\Delta^{14}C$-CO$_2$ results from the SIO network since 2007 have not been published. In the 2000s, the Institute of Arctic and Alpine Research (INSTAAR) began measuring $\Delta^{14}C$-CO$_2$ on flask samples from the Global Monitoring Laboratory (GML) of the National Oceanic and Atmospheric Administration (NOAA/GML), primarily from aircraft flights and ground-based locations over North America (Turnbull et al. 2006; Miller et al. 2012). The primary focus of the INSTAAR program was to use these data to partition the $^{14}C$-free fossil from the biogenic CO$_2$ component over the continent (Miller et al. 2012; Basu et al. 2020). $\Delta^{14}C$-CO$_2$ observations over continents have become the main focus of the carbon cycle community today, as $^{14}C$ is the most direct tracer to quantify the regional fossil CO$_2$ component (Levin et al. 2003; Turnbull et al. 2017). Also, in Europe, a new network of $\Delta^{14}C$-CO$_2$ monitoring stations was established as part of the atmospheric observational network of the Integrated Carbon Observation System (ICOS) Research Infrastructure (Levin et al. 2020). Since 2018 Jungfraujoch station has officially become part of this network, and the $\Delta^{14}C$-CO$_2$ analyses are conducted by the ICOS Central Radiocarbon Laboratory in Heidelberg (https://www.iup.uni-heidelberg.de/research/kk/icos).

**TEMPORAL DEVELOPMENT OF $\Delta^{14}C$-CO$_2$ IN THE GLOBAL TROPOSPHERE**

The long-term development of $\Delta^{14}C$ in tropospheric CO$_2$ in both hemispheres is displayed in Figure 2. The increase in the Southern Hemisphere (SH) since 1954 is documented by data from the New Zealand site close to Wellington (Turnbull et al. 2017). From 1983 onwards, we extended this record with data from the Neumayer station, Antarctica (Levin and Hammer.
In the Northern Hemisphere (NH) atmospheric, background observations are only available from 1959 onwards. In Figure 2 we extend the NH record back to the 1940s using annual mean values for 30°–90°N from the compilation by Graven et al. (2017), based on tree ring analyses. The Vermont record ended in 1985 when our Alpine background sampling site was moved to the Jungfraujoch station. For all stations, the results from individual atmospheric samples are shown in Figure 2. The data are available at the ICOS-ERIC Carbon Portal (Levin and Hammer 2021).

In the NH record, regular seasonal variations are clearly visible in the years of 1963–1968. This seasonality is caused by spring-time intrusion of stratospheric air into the troposphere (Telegadas 1971). During these initial years after the nuclear test ban treaty in 1963, a large share of bomb-produced 14CO2 continued to reside in the stratosphere, which, in the course of the following years, was transported into the troposphere, mainly during spring (Tans 1981; Hesshaimer and Levin 2000). The subsequent transport of bomb 14CO2 from the NH troposphere into the SH then caused a decrease of Δ14C-CO2 in late summer. However, the approximately 1.5 years shift between Northern and Southern Hemisphere Δ14C-CO2 curves provides the most direct measure of the interhemispheric exchange time. This atmospheric Δ14C-CO2 dynamic during and shortly after the bomb tests has been used to calibrate or evaluate the validity of transport in atmospheric circulation models (e.g., Johnston 1989; Kjellström et al. 2000; Levin et al. 2010).

The inlay of Figure 2 shows the year-to-year change of global mean Δ14C-CO2 values, calculated from the global compilation of Graven et al. (2017), extended with the most recent Heidelberg measurements from the NH and the SH. Unfortunately, no recent data from the tropics are available to calculate annual global means. We therefore extrapolated the tropical record with the annual mean trends observed in 2015–2020 for the extratropical latitudes. After the steep increase of tropospheric Δ14C-CO2, reaching a maximum in the NH in 1963 and in the SH about 1.5 years later, we observe a fast decrease in both hemispheres. This decrease is caused by equilibration of the atmospheric bomb 14C disturbance with reservoirs exchanging CO2 with the atmosphere, namely the ocean and the terrestrial biosphere. According to Levin et al. (2010: Figure 7) the largest net uptake of bomb 14C by the world oceans occurred in the 1970s and was about twice as high when compared to the net uptake by the biosphere. While the terrestrial biosphere acted as a net sink of anthropogenic 14C only until the 1980s (Naegler and Levin 2009), the oceans continued to be a sink of bomb 14C until around 2010.

Starting in the mid-1990s, fossil CO2 emissions into the global atmosphere became the dominant contribution to the decreasing global Δ14C-CO2 trend. Levin et al. (2010) attempted to use the global Δ14C-CO2 trend of the last decades as an independent constraint for the global fossil CO2 emissions into the atmosphere. However, the uncertainty estimates were determined to be too large (25–30%) to be useful, due to the large uncertainties in the partitioned contributions from ocean and biosphere exchange fluxes to the Δ14C-CO2 trend, compared to the global bottom-up fossil emission estimates, which were estimated to better than ±10% (Gilfillan and Marland 2021). Today though, with improved information on ocean and biosphere 14CO2 fluxes, it may be a useful endeavor to repeat this exercise. However, for this purpose we would first need more representative high precision Δ14C-CO2 observations at background stations that are compatible to better than ±0.5‰, in order to determine reliable global trends and hemispheric gradients, which today are only a few ‰ (see Figure 3). Better knowledge on
compatibility can only be achieved through frequent intercomparisons between those labs that contribute to this global Δ14C-CO2 background monitoring network. An earlier intercomparison between our low-level counting laboratory in Heidelberg (ICOS CRL) and 8 AMS laboratories yielded an overall agreement of better than ±0.5‰, but it was not conclusive concerning the compatibility within the AMS laboratories themselves (Hammer et al. 2017). More frequent regular intercomparisons are therefore urgently needed to achieve an overall compatibility of better than ±0.5‰. Then 14C-based ffCO2 estimates could provide an excellent independent top-down check on the global stocktake of the Paris Climate Accord (UNFCCC 2015).

SPATIAL VARIATION OF Δ14C-CO2 IN THE TROPOSPHERE

Because the bulk of the fossil CO2 emissions are released in the NH, observed CO2 concentrations in the Northern are higher than in the Southern Hemisphere (Dlugokencky et al. 2019). However, until the end of the last century, the north-south difference of Δ14C-CO2 was counterintuitively positive, with higher Δ14C-CO2 being observed in the North than in the South (Levin and Hesshaimer 2000). This latter north-south difference was caused by a strong 14CO2 disequilibrium flux with the Δ14C-depleted surface water of the Southern Ocean around Antarctica (Levin et al. 1987). The sign of the observed north-south difference, however, changed in the mid-2000s, with lower Δ14C-CO2 being observed at high northern latitudes such as at Alert in the Arctic (82.45°N, 62.52°W, 185 m asl)
when compared to Neumayer, Antarctica. We calculated smoothed fitted curves (Nakazawa et al. 1997) through the individual data from the two polar stations and plot the long-term trends together with the individual data in Figure 3a. The difference between the two curves, fitting Alert and Neumayer $\Delta^{14}$C-CO$_2$ data, is shown in Figure 3b. It can be clearly seen that while the $\Delta^{14}$C-CO$_2$ difference was positive until the end of the 1990s, it decreased to about zero at the turn of the century and around 2003, it changed to negative values and continues in this manner until today. At the same time, the CO$_2$ concentration difference between the two polar stations increased from ca. 3.5 ppm in the 1990s to ca. 4–5 ppm in the last decade (Figure 3c).

The observed change in the sign in the north-south $\Delta^{14}$C-CO$_2$ difference has been previously reported by Graven et al. (2012). There are two reasons that may have caused this reversal: (1) the increase of the north-south gradient of CO$_2$ due to increasing fossil CO$_2$ emissions being more dominant in the NH, and in turn, causing an increasing dilution of the $^{14}$C/C ratio in NH CO$_2$, and (2) the decrease of the $\Delta^{14}$C-CO$_2$ disequilibrium between the atmosphere and surface waters in the circum Antarctic ocean (Graven et al. 2012). While the difference between atmospheric and surface ocean $\Delta^{14}$C-CO$_2$ in the late 1980 and 1990s was still about 200–300‰, it decreased by about 150‰ in the following decade, because atmospheric $\Delta^{14}$C-CO$_2$ decreased by this amount (Figure 2). If we assume that the increase of CO$_2$ difference from about 1994 to 2009 between Northern and Southern Hemisphere of about 1.5 ppm was caused by $^{14}$C-free fossil CO$_2$ alone, this would have caused a change in the $\Delta^{14}$C-CO$_2$ north-south difference of approximately 3‰. The remaining difference of more than 2‰ must therefore be due to other reasons. The right panels of Figure 3 show the mean meridional distributions of CO$_2$ concentration (d) and $\Delta^{14}$C-CO$_2$ (e) in the period of 1993–1995 compared to the 15 years later period of 2008–2010. The change in $\Delta^{14}$C-CO$_2$ relative to Neumayer (NMY) station and CO$_2$ concentration relative to South Pole is observed throughout the Northern Hemispheric sites. CO$_2$ and $\Delta^{14}$C-CO$_2$ are rather homogeneously distributed from polar regions (Alert) to the subtropics at Izaña (Tenerife Island, 28.3°N, 16.48°W, 2373 m asl) with variations smaller than 1.5‰ in $\Delta^{14}$C-CO$_2$ and 1 ppm in CO$_2$. In contrast, corresponding latitudes of the Southern Hemisphere show a significant $\Delta^{14}$C-CO$_2$ dip at Macquarie Island (54.5°S, 158.97°E, 6 m asl), which is not accompanied by a significant variation in CO$_2$ concentration. In this region of the Southern Ocean around Macquarie Island (MQA) with depleted $\Delta^{14}$C the $^{14}$CO$_2$ disequilibrium flux is largest, also because large wind velocities enhance air-sea gas exchange.

As is the case for the long-term trend, changes in the north-south $\Delta^{14}$C-CO$_2$ difference can also provide a constraint on the predominantly Northern Hemispheric fossil CO$_2$ emissions. But keeping in mind that there are uncertainties in the other components contributing to the meridional $\Delta^{14}$C-CO$_2$ difference as well as uncertainties associated in atmospheric model transport limits the uncertainty of the fossil emissions constraint to around 25–30% (Levin et al. 2010).

A special feature can be seen in the data of the tropical station Llano del Hato (Venezuela, 8.78°N 70.87°W, 3600 m asl). Here we observed a regional $\Delta^{14}$C-CO$_2$ maximum in the 1990s, which we attributed to a signal of net bomb $^{14}$CO$_2$ released by heterotrophic respiration from the tropical biosphere. Naegler and Levin (2009) calculated from their global bomb-$^{14}$C budget that the global biosphere switched from a net sink of anthropogenic $^{14}$CO$_2$ to a net source to the atmosphere around the 1980s. Small fossil CO$_2$ emissions or atmosphere ocean disequilibrium fluxes could be the reason why this
bomb $^{14}$C signal from the biosphere is visible at these latitudes. Unfortunately, due to logistics problems, sampling at this important tropical site was terminated in 1997. Graven et al. (2012) also found about 3–6‰ higher $\Delta ^{14}$C-CO$_2$ in 2005–2007 at two tropical stations in the Pacific Ocean when compared to mid-latitude sites in the NH. Continuing observations in the tropics could possibly provide independent constraints on the turnover times of carbon in the terrestrial biosphere, an important parameter to assess the sustainability of the currently observed net uptake of anthropogenic CO$_2$ emissions by the terrestrial biosphere.

**CONCLUSIONS AND PERSPECTIVES**

$\Delta ^{14}$C-CO$_2$ in the troposphere over the last 60+ years serves as an excellent transient tracer to study atmospheric and global carbon cycle dynamics. However, despite the wealth of information that is buried in the observed global distribution and trend of $\Delta ^{14}$C-CO$_2$, only the first few decades of the existing records have yet been used to constrain global and regional exchange fluxes between the main compartments of the carbon cycle. This may be due to the sparsity of representative observations, for example, in the very heterogeneous terrestrial biosphere. But also for the much more homogenous global oceans, representative continuous monitoring of $^{14}$C is incomplete. Further, only in the last decade, has $^{14}$C been fully implemented in global models such as in the National Center for Atmospheric Research Community Earth System model (Koven et al. 2013; Jahn et al. 2015). Concerning atmospheric tracer transport modeling of CO$_2$, including $^{14}$C in global and regional inversions is still at its infancy (e.g., Turnbull et al. 2009; Basu et al. 2020). Here, not only do the ocean and biosphere “boundary conditions” need to be well defined, but reliable atmospheric modeling of $^{14}$CO$_2$ also requires stratosphere-troposphere air mass exchange to be represented correctly in the model. Contrary to the stable isotopes in CO$_2$, where the tropospheric variability is governed by sources and sinks located at the Earth surface, the $^{14}$CO$_2$ cycle has a natural production source in the upper troposphere and lower stratosphere. All these challenges need to be accounted for in order to disentangle the small gradients and trends, particularly those observed in the last decade. However, most important are long-term high-precision measurements at key stations themselves. Models can always be improved in the future, but the atmosphere at present can only be sampled today.

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DATA AVAILABILITY

Individual $\Delta^{14}$C-CO$_2$ data from Alert, Jungfraujoch, and Neumayer stations are available at the ICOS-ERC Carbon Portal (Levin and Hammer 2021).

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