

Basis of Tracing Fossil Fuel CO₂ Using ¹⁴C

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Contributor: Ming-Yuan Yu , Yu-Chi Lin , Yan-Lin Zhang

Carbon dioxide (CO₂), the most important greenhouse gas, is a significant driver of global warming. Radiocarbon (¹⁴C), a widely used dating method in archaeology, geosciences, etc., is a direct tracer and a promising method to differentiate the emissions of fossil fuel and non-fossil fuel from atmospheric carbon.

radiocarbon

fossil fuel carbon dioxide

emission estimation

1. Introduction

Carbon dioxide (CO₂), the most important greenhouse gas, is a significant driver of global warming. In 2019, the annual average concentration of CO₂ reached 410 ppm, which was higher than any time in at least 2 million years [1]. The observed increase in CO₂ concentrations since the beginning of the industrial era is unequivocally caused by human activities, among which the combustion of fossil fuels is responsible for most of the total anthropogenic CO₂ emissions. Global warming has caused increases in the global temperature of the surface and upper ocean, increases in precipitation and sea level, weather, and climate extremes, and decreases in glaciers and sea ice [2][3][4][5]. Besides CO₂, fossil fuel combustion is also a primary contributor to air pollutants [6]. Thus, slowing down the increase in fossil fuel CO₂ (FFCO₂) concentration is of vital importance. According to the Paris Agreement and the sixth assessment report of IPCC (Intergovernmental Panel on Climate Change), CO₂ emissions need to be net negative to hold the global surface temperature lower than 1.5 °C or 2 °C at the end of this century (very low and low greenhouse gas emission scenarios, according to IPCC, 2021). This means that the anthropogenic removal of CO₂ exceed anthropogenic emissions. Under these circumstances, identifying the contribution of FFCO₂ to total atmospheric CO₂, as well as its atmospheric process interpretation and emission estimation, is a fundamental work for studies on its climatic and environmental impacts and on the evaluation of mitigation actions.

Multiple tracers that co-emitted with CO₂ have been used to quantify FFCO₂, including carbon monoxide (CO), sulfur hexafluoride (SF₆), tetrachloroethylene (C₂Cl₄) and even air pollutants, based on the ratio of each tracer to CO₂ [7][8][9][10][11][12][13][14]. However, there are large uncertainties due to the non-fossil emissions of the tracers [15]. Radiocarbon (¹⁴C), a widely used dating method in archaeology, geosciences, etc. [16], is a direct tracer and a promising method to differentiate the emissions of fossil fuel and non-fossil fuel from atmospheric carbon. The abundances of three naturally occurring carbon isotopes ¹²C, ¹³C and ¹⁴C are 98.89%, 1.11%, and $\sim 10^{-10}\%$, respectively [17]. The radiocarbon content of CO₂ is expressed as $\Delta^{14}\text{C}$ or $\Delta^{14}\text{CO}_2$ [18][19]:

$$\Delta^{14}\text{C} = \left[\frac{(^{14}\text{C}/^{12}\text{C})_{\text{SN}}}{(^{14}\text{C}/^{12}\text{C})_{\text{ABS}}} - 1 \right] \times 1000\%. \quad (1)$$

$(^{14}\text{C}/^{12}\text{C})_{\text{SN}}$ is the ¹⁴C to ¹²C ratio of the sample, and $(^{14}\text{C}/^{12}\text{C})_{\text{ABS}}$ is related to the commonly used primary measurement standard Oxalic Acid I. Radiocarbon is cosmogenic, and has a radioactive half-life of 5730 ± 40 years [20]. Thus, there are no ¹⁴C in fossil fuels because they are all depleted during long-term radioactive decay. Since fossil fuel CO₂ contains no ¹⁴C whereas CO₂ from other sources has similar ¹⁴C concentrations with the ambient air, the release of fossil fuel CO₂ will cause a decrease in the ¹⁴C/¹²C ratio in the atmosphere. This was first discovered by Hans Suess [21], and is called the “Suess effect”. With industrial development, atmospheric $\Delta^{14}\text{CO}_2$ decreased by 25‰ between 1890 and 1950 [22]. Then comes the nuclear testing period between the 1950s and the early 1960s, during which large-scale detonations of nuclear bombs produced ¹⁴C atoms in the Northern Hemisphere. Atmospheric $\Delta^{14}\text{CO}_2$ in the Northern Hemisphere increased swiftly and reached a peak value of nearly 1000‰ in 1963, and then decreased after the Limited Nuclear Test Ban Treaty [23][24].

2. The Basis of Tracing Fossil Fuel CO₂ Using ¹⁴C

2.1. The Theory of Quantifying Fossil Fuel CO₂ Using ¹⁴C

Observed CO₂ mole fraction (or concentration) is thought to be a mixing of many components, mainly including atmospheric background CO₂, fossil fuel CO₂, biospheric CO₂ and oceanic CO₂. The most commonly used method to constrain recently added FFCO₂ in the atmosphere with ¹⁴C is called the pseudo-Lagrangian method [25][26][27], in which a parcel of air with an initial CO₂ mixing ratio (CO₂_{bg}) and $\Delta^{14}\text{CO}_2$ value (Δ_{bg}) moves across a polluted region, and then CO₂ mixing ratio and $\Delta^{14}\text{CO}_2$ value are modified to CO₂_{obs} and Δ_{obs} by the addition of FFCO₂ and other sources or sinks of CO₂. If combining other sources (and sinks) together, the mixing ratio and the $\Delta^{14}\text{CO}_2$ value could be written as CO₂_{other} and Δ_{other} . Two balance equations for CO₂ mixing ratio and $\Delta^{14}\text{C}$ can be formulated as below.

$$\text{CO}_2_{\text{obs}} = \text{CO}_2_{\text{bg}} + \text{CO}_2_{\text{ff}} + \text{CO}_2_{\text{other}} \quad (2)$$

$$\Delta_{\text{obs}} \text{CO}_2_{\text{obs}} = \Delta_{\text{bg}} \text{CO}_2_{\text{bg}} + \Delta_{\text{ff}} \text{CO}_2_{\text{ff}} + \Delta_{\text{other}} \text{CO}_2_{\text{other}} \quad (3)$$

By combining Equations (2) and (3), CO₂_{ff} can be calculated as:

$$\text{CO}_{2\text{ff}} = \frac{\text{CO}_{2\text{obs}} (\Delta_{\text{obs}} - \Delta_{\text{bg}})}{\Delta_{\text{ff}} - \Delta_{\text{bg}}} - \frac{\text{CO}_{2\text{other}} (\Delta_{\text{other}} - \Delta_{\text{bg}})}{\Delta_{\text{ff}} - \Delta_{\text{bg}}}. \quad (4)$$

CO_{2obs}, Δ_{obs} are measured in collected samples at interested sites. Δ_{bg} is measured in samples from background sites in general, while free tropospheric measurements can also act as Δ_{bg} , too [13]. Δ_{ff} is known to be -1000% since CO_{2ff} is ¹⁴C-free.

The second term of Equation (4) is bias due to the effect of the others:

$$\beta = \frac{\text{CO}_{2\text{other}} (\Delta_{\text{other}} - \Delta_{\text{bg}})}{\Delta_{\text{ff}} - \Delta_{\text{bg}}}. \quad (5)$$

some researchers assume β to be zero, which means that all other sources have the same $\Delta^{14}\text{C}$ compared to those of the background atmosphere, $\Delta_{\text{other}} = \Delta_{\text{bg}}$ [26]. The main contributor to uncertainties in β would be heterotrophic respiration, which has large ¹⁴C disequilibrium. The ignorance of β would cause a systematic underestimation of CO_{2ff}, up to 0.5 ppm in summer and 0.2 ppm in winter [13][27]. There are two other factors that influence atmospheric $\Delta^{14}\text{CO}_2$, air-sea exchange in the oceans, and stratosphere-troposphere transport [28]. However, these exchanges are assumed to affect the background and observed samples equally; thus, normally, they will not be counted in the calculation of FFCO₂.

2.2. Air Sampling and Measurement

Atmospheric $\Delta^{14}\text{CO}_2$ can be measured with direct air sampling. Whole air samples are normally collected using flasks or bags. Short-period and integrated samples can be collected by pump and acid solution, respectively. CO₂ samples can be collected by static absorption using CO₂-free sodium hydroxide (NaOH) or barium hydroxide (BaOH) solutions in flasks [25][29][30]. The primary collection method is the static absorption of CO₂ using CO₂-free sodium hydroxide (NaOH) or barium hydroxide (BaOH) solutions in discrete glass flasks [25][29]. The flasks are exposed to air for collection of integrated samples. Besides ground sites, tall towers, aircrafts, balloons, and even kites are all effective platforms to collect CO₂ samples [13][31][32][33][34].

Air samples reflect near real-time atmospheric $\Delta^{14}\text{CO}_2$, can be used to characterize the FFCO₂ temporal variations with high resolution effectively. However, the representativeness of the air samples is limited to those of the sampling region and period, while little information (spatial and temporal distribution) is known beyond that. In addition, the sample collecting process and/or the site maintenance is labor and cost intensive. Direct sampling of air is not the only way to analyze atmospheric $\Delta^{14}\text{CO}_2$. Plants fix CO₂ from the atmosphere via photosynthesis, offering a unique complementary analysis method.

For plants, their carbon isotopic composition can be used to reflect the mean atmosphere $\Delta^{14}\text{CO}_2$ isotopic composition of their growing period. By collecting plant samples in different regions and analyzing ¹⁴C, FFCO₂ spatial distribution on a large scale can be mapped out. Compared to air samples, collecting plant materials is more convenient and relatively cheap. Tree rings and annual leaves (grasses) are two main types used to reveal the spatiotemporal distribution of FFCO₂ [30][35][36][37][38][39][40]. Each plant species may have its own advantages in addition to those illustrated above. Maize is grown in many countries, so it is convenient to map out the large-scale spatial distribution of fossil fuel influences using corn leaves. Gingko is a perennial and deciduous tree that is widely planted in East Asian countries, urban areas, and rural areas. Thus, it is feasible to separate samples of clean sites from samples of polluted sites. Wine ethanol is a unique plant material that can represent previous sampling years, since the harvest year and region are all written on the label of the wine bottle. Tree rings, a unique plant material, help in the reconstruction of annual atmospheric $\Delta^{14}\text{CO}_2$ for decades or hundreds of years. In practice, however, the sampling of tree rings may be more difficult than that of annual plants since it is difficult to separate one annual ring from the others.

Comparisons of $\Delta^{14}\text{CO}_2$ and/or FFCO₂ between plant materials and air samples show nearly consistent results [30][41][42][43], which verifies the usage of plant materials. Xiong et al. [44] found a significant difference in $\Delta^{14}\text{CO}_2$ between respired CO₂ and bulk organic matter from 21 plant species, suggesting that bias associated with dark respiration should be considered when use ¹⁴C in plants to quantify atmospheric FFCO₂. It should be noted that biomass accumulated by plants only represents daytime $\Delta^{14}\text{CO}_2$ (when photosynthesis occurs), and the sampling should be well planned for different plant species considering their growing period and local climate.

Before the analysis of ¹⁴C, the preparations of air samples included extracting the CO₂ (purification), and the reduction of CO₂ to graphite. The extraction of CO₂ is to remove water cryogenically, freeze CO₂ completely together with N₂O (non-interfering), and without freeze O₂ or CH₄ [13][45]. Graphite is produced by adding hydrogen gas to CO₂ over an iron catalyst [46][47]. The atom counting of each graphite sample is then performed by an accelerator mass spectrometer (AMS). The preparation of annual leaves is a little different from air samples: plant samples need to (1) be cleaned by pure water and then dried, (2) be combusted to CO₂ and then reduced to graphite [35].

Direct atom-counting of ¹⁴C using AMS is a great progress of ¹⁴C analysis methods. Before that, the conventional methods were decay counting, solid carbon using a Geiger–Muller counter, and liquid scintillation counting [48]. The sensitivity was improved around 10⁶ times by AMS over the decay counting methods [49]. With the attempts to reduce sample size and to increase precision, the detection limits have been reduced to ~5 µg of carbon [50][51], and the reported precisions have reached 1% [17].

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