Basis of Tracing Fossil Fuel CO_2 Using ^{14}C

Subjects: Meteorology & Atmospheric Sciences

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Carbon dioxide (CO_2), the most important greenhouse gas, is a significant driver of global warming. Radiocarbon (^{14}C), 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.

Keywords: radiocarbon ; fossil fuel carbon dioxide ; emission estimation

1. Introduction

Carbon dioxide (CO_2), the most important greenhouse gas, is a significant driver of global warming. In 2019, the annual average concentration of CO_2 reached 410 ppm, which was higher than any time in at least 2 million years [1]. The observed increase in CO_2 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 CO_2 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 CO_2, fossil fuel combustion is also a primary contributor to air pollutants [6]. Thus, slowing down the increase in fossil fuel CO_2 ($FFCO_2$) concentration is of vital importance. According to the Paris Agreement and the sixth assessment report of IPCC (Intergovernmental Panel on Climate Change), CO_2 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 CO_2 exceed anthropogenic emissions. Under these circumstances, identifying the contribution of $FFCO_2$ to total atmospheric CO_2, 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 CO_2 have been used to quantify $FFCO_2$, including carbon monoxide (CO), sulfur hexafluoride (SF_6), tetrachloroethylene (C_2Cl_4) and even air pollutants, based on the ratio of each tracer to CO_2 [7][8][9][10][11][12][13][14]. However, there are large uncertainties due to the non-fossil emissions of the tracers [15]. Radiocarbon (^{14}C), 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 ^{12}C, ^{13}C and ^{14}C are 98.89%, 1.11%, and ~10^{-10}%, respectively [17]. The radiocarbon content of CO_2 is expressed as $\Delta^{14}C$ or $\Delta^{14}CO_2$ [18][19]:

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

$(^{14}C/^{12}C)_{SN}$ is the ^{14}C to ^{12}C ratio of the sample, and $(^{14}C/^{12}C)_{ABS}$ 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 ^{14}C in fossil fuels because they are all depleted during long-term radioactive decay. Since fossil fuel CO_2 contains no ^{14}C whereas CO_2 from other sources has similar ^{14}C concentrations with the ambient air, the release of fossil fuel CO_2 will cause a decrease in the $^{14}C/^{12}C$ ratio in the atmosphere. This was first discovered by Hans Suess [21], and is called the “Suess effect”. With industrial development, atmospheric $\Delta^{14}CO_2$ 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 ^{14}C atoms in the Northern Hemisphere. Atmospheric $\Delta^{14}CO_2$ 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 CO_2 Using ^{14}C

2.1. The Theory of Quantifying Fossil Fuel CO_2 Using ^{14}C

Observed CO_2 mole fraction (or concentration) is thought to be a mixing of many components, mainly including atmospheric background CO_2, fossil fuel CO_2, biospheric CO_2 and oceanic CO_2. The most commonly used method to constrain recently added $FFCO_2$ in the atmosphere with ^{14}C is called the pseudo-Lagrangian method [25][26][27], in which a parcel of air with an initial CO_2 mixing ratio (CO_{2bg}) and $\Delta^{14}CO_2$ value (Δ_{bg}) moves across a polluted region, and then CO_2 mixing ratio and $\Delta^{14}CO_2$ value are modified to CO_{2obs} and Δ_{obs} by the addition of $FFCO_2$ and other sources or sinks of CO_2. If combining other sources (and sinks) together, the mixing ratio and the $\Delta^{14}CO_2$ value could be written as CO_{2other} and Δ_{other}. Two balance equations for CO_2 mixing ratio and $\Delta^{14}C$ can be formulated as below.

$$\mathrm{CO_{2\,obs}} = \mathrm{CO_{2\,bg}} + \mathrm{CO_{2\,ff}} + \mathrm{CO_{2\,other}} \tag{2}$$

$$\Delta_{\mathrm{obs}}\mathrm{CO_{2\,obs}} = \Delta_{\mathrm{bg}}\mathrm{CO_{2\,bg}} + \Delta_{\mathrm{ff}}\mathrm{CO_{2\,ff}} + \Delta_{\mathrm{other}}\mathrm{CO_{2\,other}} \tag{3}$$

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

$$\mathrm{CO_{2\,ff}} = \frac{\mathrm{CO_{2\,obs}}\left(\Delta_{\mathrm{obs}} - \Delta_{\mathrm{bg}}\right)}{\Delta_{\mathrm{ff}} - \Delta_{\mathrm{bg}}} - \frac{\mathrm{CO_{2\,other}}\left(\Delta_{\mathrm{other}} - \Delta_{\mathrm{bg}}\right)}{\Delta_{\mathrm{ff}} - \Delta_{\mathrm{bg}}}. \tag{4}$$

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 ^{14}C-free.

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

$$\beta = \frac{\mathrm{CO_{2\,other}}\left(\Delta_{\mathrm{other}} - \Delta_{\mathrm{bg}}\right)}{\Delta_{\mathrm{ff}} - \Delta_{\mathrm{bg}}}. \tag{5}$$

some researchers assume β to be zero, which means that all other sources have the same $\Delta^{14}C$ compared to those of the background atmosphere, $\Delta_{other} = \Delta_{bg}$ [26]. The main contributor to uncertainties in β would be heterotrophic respiration, which has large ^{14}C 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 $\Delta^{14}CO_2$, 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 $FFCO_2$.

2.2. Air Sampling and Measurement

Atmospheric $\Delta^{14}CO_2$ 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. CO_2 samples can be collected by static absorption using CO_2-free sodium hydroxide (NaOH) or barium hydroxide (BaOH) solutions in flasks [25][29][30]. The primary collection method is the static absorption of CO_2 using CO_2-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 CO_2 samples [13][31][32][33][34].

Air samples reflect near real-time atmospheric $\Delta^{14}CO_2$, can be used to characterize the $FFCO_2$ 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 $\Delta^{14}CO_2$. Plants fix CO_2 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 $\Delta^{14}CO_2$ isotopic composition of their growing period. By collecting plant samples in different regions and analyzing ^{14}C, $FFCO_2$ 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 $FFCO_2$ [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 $\Delta^{14}CO_2$ 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 $\Delta^{14}CO_2$ and/or $FFCO_2$ 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 $\Delta^{14}CO_2$ between respired CO_2 and bulk organic matter from 21 plant species, suggesting that bias associated with dark respiration should be considered when use ^{14}C in plants to quantify atmospheric $FFCO_2$. It should be noted that biomass accumulated by plants only represents daytime $\Delta^{14}CO_2$ (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 ^{14}C, the preparations of air samples included extracting the CO_2 (purification), and the reduction of CO_2 to graphite. The extraction of CO_2 is to remove water cryogenically, freeze CO_2 completely together with N_2O (non-interfering), and without freeze O_2 or CH_4 [13][45]. Graphite is produced by adding hydrogen gas to CO_2 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 CO_2 and then reduced to graphite [35].

Direct atom-counting of ^{14}C using AMS is a great progress of ^{14}C 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 10^6 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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