



# Evolution and variation of atmospheric carbon dioxide concentration over terrestrial ecosystems as derived from eddy covariance measurements



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## HIGHLIGHTS

- Atmospheric CO<sub>2</sub> concentration over terrestrial ecosystems (ACTE) was analyzed.
- ACTE was higher by 9.0 ppm in winter and lower 2.1 ppm in summer than global means.
- Annual mean and seasonal amplitude of ACTE increased with 2.04 and 0.60 ppm yr<sup>-1</sup>.
- The annual CO<sub>2</sub> concentration showed large variation among ecosystems.

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## ABSTRACT

Carbon dioxide (CO<sub>2</sub>) is the most important anthropogenic greenhouse gas contributing to global climate change. Understanding the temporal and spatial variations of CO<sub>2</sub> concentration over terrestrial ecosystems provides additional insight into global atmospheric variability of CO<sub>2</sub> concentration. Using 355 site-years of CO<sub>2</sub> concentration observations at 104 eddy-covariance flux tower sites in Northern Hemisphere, we presented a comprehensive analysis of evolution and variation of atmospheric CO<sub>2</sub> concentration over terrestrial ecosystem (ACTE) for the period of 1997–2006. Our results showed that ACTE exhibited a strong seasonal variations, with an average seasonal amplitude (peak-trough difference) of 14.8 ppm, which was approximately threefold that global mean CO<sub>2</sub> observed in Mauna Loa in the United States (MLO). The seasonal variation of CO<sub>2</sub> were mostly dominant by terrestrial carbon fluxes, i.e., net ecosystem production (NEP) and gross primary production (GPP), with correlation coefficient ( $r$ ) were  $-0.55$  and  $-0.60$  for NEP and GPP, respectively. However, the influence of carbon fluxes on CO<sub>2</sub> were not significant at interannual scale, which implied that the inter-annual changing trends of atmospheric CO<sub>2</sub> in Northern Hemisphere were likely to depend more on anthropogenic CO<sub>2</sub> emissions sources than on ecosystem change. It was estimated, by fitting a harmonic model to monthly-mean ACTE, that both annual mean and seasonal amplitude of ACTE increased over the 10-year period at rates of 2.04 and 0.60 ppm yr<sup>-1</sup>, respectively. The uptrend of annual ACTE could be attributed to the dramatic global increase of CO<sub>2</sub> emissions during the study period, whereas the increasing amplitude could be related to the increases in Northern Hemisphere biospheric activity. This study also found that the annual CO<sub>2</sub> concentration showed large variation among ecosystems, with the high value appeared in deciduous broadleaf forest, evergreen broadleaf forest and cropland. We attribute these discrepancies to both differential local anthropogenic impacts and carbon sequestration abilities across ecosystem types.

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## 1. Introduction

Carbon dioxide (CO<sub>2</sub>) is the most important anthropogenic greenhouse gas that contributes most to global climate change (Hofmann et al., 2009; IPCC, 2013). Precise measurements of atmospheric CO<sub>2</sub> concentration over a wide geographical area are indispensable for understanding global carbon cycle (Zhang et al., 2008). Atmospheric CO<sub>2</sub> concentration has been systematically monitored for decades at many ground-based sites and networks all over the world (Keeling and Whorf, 2005; Zhang et al., 2007; Sasakawa et al., 2013). These observations indicate that, during the past decade, global atmospheric CO<sub>2</sub> concentration has been consistently increasing at a rate of 2 parts per million (ppm) yr<sup>-1</sup> (WMO, 2012), and passed 400 ppm in May 2013 (Bala, 2013; Williamson, 2013). The consistent long-term increase in atmospheric CO<sub>2</sub> concentration makes it important to explore the temporal variations of atmospheric CO<sub>2</sub>. Meanwhile, it is also important to examine the spatial variations since atmospheric CO<sub>2</sub> concentration in the surface layer of the atmosphere varies significantly over different land covers (George et al., 2007; Zhang et al., 2008; Jacobson, 2010), although CO<sub>2</sub> is generally assumed to be well-mixed in the boundary layer. Better knowledge of the temporal and spatial variations of CO<sub>2</sub> concentration in the surface layer of the atmosphere is needed to improve the forecast of future CO<sub>2</sub> concentration levels (Wu et al., 2012).

Terrestrial ecosystems play important role in controlling atmospheric CO<sub>2</sub> concentration (IPCC, 2013; Pan et al., 2011; Yu et al., 2014). It has been found that the varying CO<sub>2</sub> exchanges between atmosphere and the terrestrial biosphere is the main driver of the observed atmospheric CO<sub>2</sub> cycle, including its seasonal cycle and inter-annual variations (Friend et al., 2007; Arneeth et al., 2010). This effect has been linked to changes in temperate, boreal and arctic ecosystem properties and processes such as enhanced photosynthesis, increased heterotrophic respiration, and expansion of woody vegetation (Piao et al., 2008; Barichivich et al., 2013). The Northern Hemisphere dominates the annual cycle of CO<sub>2</sub> concentration because it has much greater land area and plant biomass than the Southern Hemisphere (Yu et al., 2014). CO<sub>2</sub> emissions from forest fires in the tropical and boreal region have also been found to contribute the interannual variability in the CO<sub>2</sub> growth rate (Langenfelds et al., 2002; Patra et al., 2005; Zhang et al., 2007). It is also interesting to note that Gray et al. (2014) and Zeng et al. (2014) have shown that the intensification of agriculture may have been a key contributor to the increase in atmospheric CO<sub>2</sub> seasonal amplitude. However, the precise causal mechanisms behind the observed changes in atmospheric CO<sub>2</sub> seasonality remain unclear (Gray et al., 2014). Ambient information about CO<sub>2</sub> evolution over specific terrestrial ecosystems can provide important insight into atmospheric CO<sub>2</sub> variability and the land-atmospheric feedbacks (Wu et al., 2012).

The data from precise measurements of the atmospheric CO<sub>2</sub> concentration over a wide geographical area have been used to constrain the global carbon budget (Gurney et al., 2002; Patra et al., 2005). The present CO<sub>2</sub> measurements with nearly undisturbed air is good to represent global mean concentration (Keeling, 1998; Zhang et al., 2008), but it cannot be used as a real-time indicator of carbon exchanges that usually affected by both human activities and vegetation dynamics, or used to interpret the heterogeneity of atmospheric CO<sub>2</sub> over different regions. To better understand carbon cycle over terrestrial ecosystems, continuous in-situ measurements of land-atmosphere CO<sub>2</sub> exchanges have been made with the eddy covariance (EC) technique since 1990s. Consequently, many EC observation sites have been established, leading to the development of regional networks, such as AmeriFLUX, AsiaFLUX and ChinaFLUX, and then the global network FLUXNET, A

"network of regional networks" (Baldocchi et al., 2001). By now, EC technique has been widely applied for a wide range of ecosystem types, including forests, grasslands, and croplands etc., and has therefore become an indispensable tool for understanding and monitoring global carbon cycle (Friend et al., 2007). These measurements are being widely used in global carbon cycle and related modeling studies (Friend et al., 2007; Stockli et al., 2008; Williams et al., 2009). Furthermore, the long-term continuous observations of atmospheric CO<sub>2</sub> concentration and its fluxes at multiple temporal resolutions (from seconds to years), for different terrestrial ecosystems, provide additional information for better understanding the temporal and spatial dynamics of atmospheric CO<sub>2</sub> variability.

In this paper, we illustrated the long-term atmospheric CO<sub>2</sub> concentration over terrestrial ecosystems (ACTE) based from micrometeorological tower sites with EC technique. This study aims to reveal the evolution and variation of ACTE with two objectives: (1) to quantify the temporal variation and annual trends of ACTE; and (2) to examine the variability of CO<sub>2</sub> concentration among different terrestrial ecosystems. The atmospheric CO<sub>2</sub> concentration over terrestrial ecosystems presented in this study would provide additional insight for evaluating the combined effects of human activities and vegetation dynamics on atmospheric CO<sub>2</sub>.

## 2. Materials and methods

### 2.1. Data sources and site information

Our study was based on 355 site-years of data at 104 sites from 23 countries in the Northern Hemisphere. These data were collected from FLUXNET ([www.fluxdata.org](http://www.fluxdata.org)), and ChinaFLUX ([www.chinaflux.org](http://www.chinaflux.org)) during 1997–2006 (Fig. 1). At each individual site, EC technique was used to measure mass and energy exchange across a horizontal plane between vegetation and the free atmosphere. The original half-hour CO<sub>2</sub> concentrations are measured with open or closed path infrared gas analyzers at constant flux layer, where vertical carbon exchange varies little with altitude (Aubinet et al., 2000; Baldocchi et al., 2001; Yu et al., 2006). To examine the seasonal and interannual variability in CO<sub>2</sub> concentration, only original data gap less than 30% were selected. The latitudes ranges from 2°N to 69°N, longitudes are from 121°W to 128°E, with majority of sites are located within the region of mid-high latitude and temperate climates (Table S1). The ecosystem types included cropland (CRO), deciduous broadleaf forest (DBF), evergreen broadleaf forest (EBF), evergreen needleleaf forest (ENF), grassland (GRA), mixed forest (MF), open shrub lands (OSH), wetlands (WET), and woody savanna (WSA) (Table 1). Due to the availability of EC datasets, among the 104 studied sites, ~85% of the sites located in European Union (EU28, 63 sites) and America (USA, 31 sites). Within North America or Europe, EC tower sites are reasonably well distributed among the major biomes.

Due to the existing of outliers in the CO<sub>2</sub> concentration measurements, spike screening was applied for all dataset. Any data outside  $\pm 3$  standard deviations within 6 h (12 points) were regarded as outliers and excluded from the records (Zhang et al., 2007). This procedure was repeated until no outlier was identified, with ~6.75% of the CO<sub>2</sub> observations rejected. After spike screening, the valid CO<sub>2</sub> data were  $75 \pm 11\%$  of the total observations for all site-years. In the end, small gaps (<2 h) were linearly interpolated, while larger gaps in CO<sub>2</sub> records were filled with mean diurnal variation (MDV) method (Falge et al., 2001).

In addition, to examine the contribution of carbon fluxes over terrestrial ecosystems to the trend of atmospheric CO<sub>2</sub>, the half-hour net ecosystem production (NEP) and gross primary production (GPP) fluxes at each site-year were also collected. For each site,

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