1 **Evolution and variation of atmospheric carbon dioxide concentration**
2 **over terrestrial ecosystems as derived from eddy covariance**
3 **measurements**

4
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Abstract

Carbon dioxide (CO₂) is the most important anthropogenic greenhouse gas contributing to global climate change. Understanding the temporal and spatial variations of CO₂ concentration over terrestrial ecosystems provides additional insight into global atmospheric variability of CO₂ concentration. Using 355 site-years of CO₂ concentration observations at 104 eddy-covariance flux tower sites in Northern Hemisphere, we presented a comprehensive analysis of evolution and variation of atmospheric CO₂ 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 CO₂ observed in Mauna Loa in the United States (MLO). The seasonal variation of CO₂ 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 CO₂ were not significant at interannual scale, which implied that the inter-annual changing trends of atmospheric CO₂ in Northern Hemisphere were likely to depend more on anthropogenic CO₂ 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⁻¹, respectively. The uptrend of annual ACTE could be attributed to the dramatic global increase of CO₂ 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 CO₂ 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.

Keywords: CO₂ concentration; Terrestrial ecosystem; Carbon flux; Eddy covariance technique

1. Introduction

Carbon dioxide (CO₂) is the most important anthropogenic greenhouse gas that contributes most to global climate change (Hofmann et al., 2009; IPCC, 2013). Precise measurements of atmospheric CO₂ concentration over a wide geographical area are indispensable for understanding global carbon cycle (Zhang et al., 2008). Atmospheric CO₂ concentration has been systematically monitored for decades at many ground-based sites and networks all over the world (Keeling&Whorf, 2005; Zhang et al., 2007; Sasakawa et al., 2013). These observations indicate that, during the past decade, global atmospheric CO₂ concentration has been consistently increasing at a rate of 2 parts per million (ppm) yr⁻¹ (WMO, 2012), and passed 400 ppm in May 2013 (Bala, 2013; Williamson,2013). The consistent long-term increase in atmospheric CO₂ concentration makes it important to explore the temporal variations of atmospheric CO₂. Meanwhile, it is also important to examine the spatial variations since atmospheric CO₂ 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 CO₂ is generally assumed to be well-mixed in the boundary layer. Better knowledge of the temporal and spatial variations of CO₂ concentration in the surface layer of the atmosphere is needed to improve the forecast of future CO₂ concentration levels (Wu et al., 2012).

Terrestrial ecosystems play important role in controlling atmospheric CO₂ concentration (IPCC, 2013; Pan et al., 2011; Yu et al., 2014). It has been found that the varying CO₂ exchanges between atmosphere and the terrestrial biosphere is the main driver of the observed atmospheric CO₂ cycle, including its seasonal cycle and inter-annual variations (Friend et al., 2007; Arneth 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 CO₂ concentration because it has much greater land area and plant biomass than the Southern Hemisphere(Yu et al., 2014). CO₂ emissions from forest fires in the tropical and boreal region have also been found to contribute the inter annual variability in the CO₂ 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 CO₂ seasonal amplitude. However, the precise causal mechanisms behind the observed changes in atmospheric CO₂ seasonality remain unclear(Gray et al., 2014). Ambient information about CO₂ evolution over specific terrestrial ecosystems can provide important insight into atmospheric CO₂ variability and the land-atmospheric feedbacks (Wu et al., 2012).

The data from precise measurements of the atmospheric CO₂ 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 CO₂ 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 CO₂ over different regions. To better understand carbon cycle over terrestrial ecosystems, continuous in-situ measurements of land-atmosphere CO₂ 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 CO₂ 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 CO₂ variability.

In this paper, we illustrated the long-term atmospheric CO₂ 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 CO₂ concentration among different terrestrial ecosystems. The atmospheric CO₂ 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 CO₂.

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), and ChinaFLUX (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 CO₂ 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 CO₂ concentration, only original data gap less than 30% were selected. The latitudes range 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 CO₂ concentration measurements, spike screening was applied for all dataset. Any data outside ± 3 standard deviations within 6 hours (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 CO₂ observations rejected. After spike screening, the valid CO₂ data were 75 ± 11 % of the total observations for all site-years. In the end, small gaps (< 2 h) were linearly interpolated, while larger gaps in CO₂ records were filled with mean diurnal variation (MDV) method (Falge et al., 2001).

Fig. 1. Geographical distribution of the study sites

Table 1. Distribution of sites(site-years) with respect to climate and vegetation classes. Climate was defined according to aggregated Koppen-Geiger classification (Rubel& Kottek, 2010). Vegetation classes were from IGBP definitions: 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). The numbers in the brackets represented the site-years.

In addition, to examine the contribution of carbon fluxes over terrestrial ecosystems to the trend of atmospheric CO₂, the half-hour net ecosystem production(NEP) and gross primary production(GPP) fluxes at each site-year were also collected. For each site, carbon fluxes were determined by measuring the covariance between fluctuations in vertical velocity and the mixing ratio of CO₂ (Aubinet et al., 2000). Positive NEP represented a loss of CO₂ from the atmosphere and a gain by the surface. To minimize the uncertainty derived from different data processing method, all data were processed and filled in a harmonized manner using techniques described in Reichstein et al.(2005), Papale et al.(2006), and Moffat et al.(2007). The goal of gap filling is the reproduction of the NEP time series and partitioned into GPP and ecosystem respiration(RE). After gap-filling, the CO₂ concentration and the corresponding carbon fluxes over terrestrial ecosystems were aggregated into the daily, monthly and annual scale.

2.2 Analysis of annual and seasonal variation of CO₂ concentration

To estimate the annual trends of CO₂ concentration accurately, the following harmonic model was used to fit the monthly CO₂ data (Wu et al., 2012; Perez et al.,

2007) :

$$y = a + bt + c \cos(\omega t - \theta_1) + d \sin(\omega t - \theta_2) \quad (1)$$

Where y is the monthly CO₂ concentration time-series at the site or sites which need to be studied, a is the mean initial value, b is the monthly trend of CO₂ concentration, t is time expressed as consecutive months (1, 2, ..., 120 in this study), and ω is the frequency ($2\pi/12$ in radians per month) that is applicable to a period of one year, c and θ_1 correspond the change rate and angular location associated with the annual maximum monthly CO₂ concentration, d is the change rate of annual amplitude, and θ_2 provides the difference of angular location between annual monthly maximum and minimum CO₂. In this study, these six parameters ($a, b, c, \theta_1, d, \theta_2$) in Eq.(1) were determined by a nonlinear regression fit with least square method.

3. Results and discussion

3.1 Seasonal variations of CO₂ concentration over terrestrial ecosystem (ACTE)

The averaged daily and monthly ACTE during the study period, from 1997 to 2006, were shown in Fig. 2. For comparison, we presented the CO₂ observations from Mauna Loa in the United States (MLO) which was generally considered representative of global mean CO₂ since there is very little local human and vegetation influence at this site (Conway et al., 1994; Bala, 2013). It was obvious from Fig.2 that the CO₂ concentration over terrestrial ecosystems and MLO increases with time, accompanying by clear seasonal cycle. ACTE decreased beginning in spring as a result of plant photosynthesis and rose again in autumn as fallen leaves decayed and released CO₂ into the atmosphere (Nevison et al., 2008; Wu et al., 2012). The seasonal cycle of ACTE has a significant correlation of 0.7 ($p < 0.01$) with that of MLO at a lag of 2 months (Fig 2b). For MLO, the minimum and maximum CO₂ concentration appeared in September and May, which was partly related to the long-distance transfer of atmospheric carbon dioxide in the horizontal direction (Gerbig et al., 2003). The difference between monthly ACTE and CO₂ observation at MLO was season-dependent. Specifically, the monthly ACTE during growing season

(i.e., May-August) was lower than that of MLO by 2.0-5.8 ppm. However, with the influence of local carbon sources from human activity and the deterioration of carbon uptake by terrestrial ecosystem, the ACTE was higher than that of MLO by ~1.5 ppm in April and September. In addition, local anthropic local emissions from fossil fuel combustions, mainly related to indoor energy use(including the heating and lighting requirements), and biomass-burning peaked in winter (Blasing et al.,2005; Szidat et al.,2006), which leading to a higher CO₂ concentration over terrestrial ecosystems in Northern Hemisphere than that observed in MLO by 4.0-9.4 ppm.

Fig. 2. Evolutions of the daily (a) and monthly (b) CO₂ means over the entire valid FLUXNET sites and Mauna Loa (MLO) from 1997 to 2006. The black circles depicted the observed CO₂ daily means (in upper panel) and monthly means (in the lower panel) from FLUXNET. The blue circles depicted the monthly observed CO₂ (in the lower panel) from MLO. The red and blue lines in lower panel denote the simulated monthly CO₂ values for EC sites and MLO, respectively, using Eq.(1).

The averaged seasonal amplitude (peak-trough difference) of ACTE (14.8 ppm) was approximately threefold that observed in MLO (5.7 ppm). The intra-annual of ACTE were strongly affected by regional vegetation canopy in summer, and the strong uptake by local vegetation obviously decrease the atmospheric CO₂, leading to monthly CO₂ concentration lower than that at MLO by 2.1 ± 1.5 ppm. In the other seasons, ACTE were obvious higher than MLO. Especially in winter, when plants were dormant, huge amount of CO₂ was returned back to atmosphere, with a higher value by 9.0 ± 1.8 ppm. As shown in Fig.2b, the harmonic model (Eq.(1)) performed well to capture the long term evolution of ACTE and CO₂ observation in MLO, with $R^2 = 0.84$ and 0.99 for ACTE and MLO, respectively. It was estimated, by fitting a harmonic model to monthly-mean ACTE, seasonal amplitude of ACTE increased over the 10-year period at rates of 0.60 ppm yr^{-1} . The increase in amplitude has been attributed to increasing assimilation and release of CO₂ by terrestrial ecosystem and the advance of spring drawdown to climate warming(Keeling et al., 1996; Piao et al., 2008), suggested that terrestrial ecosystems in Northern Hemisphere were experiencing greater CO₂ uptake during the growing season and greater CO₂ release during period outside the growing season(Sitch et al., 2015).

To provide a quantitative interpretation of the primary fluxes from terrestrial ecosystems on annual cycle of CO₂ concentration, seasonal patterns of ACTE and carbon fluxes(i.e., NEP and GPP) were analyzed further. As shown in Fig. 3, the seasonal variation of CO₂ were mostly dominant by terrestrial carbon fluxes with correlation coefficient(*r*) were -0.55 and -0.60 for NEP and GPP, respectively. It was also should be note that the influence of GPP on CO₂ level was higher than that of NEP. This was probably because NEP is the difference between ecosystem respiration RE and GPP (NEP = GPP-RE)(Falge et al., 2001), a fairly small value compared to large GPP and RE. A change in the seasonal cycle of either of these one way fluxes, whether in phase, magnitude, or form, will alter the seasonal cycle of NEP. On the other hand, the observed technique and the post-field data processing approach might cause biases in the NEP fluxes in a few sources, including systematic and random errors(Hollinger and Richardson, 2005; Lasslop et al., 2008; He et al., 2010), the uncertainty caused by quality control and gap-filling method (Papale et al., 2006; Liu et al., 2012). In addition to carbon fluxes from terrestrial ecosystems, surface fluxes from fossil fuel combustion, biomass burning and ocean exchange also contribute to the seasonal cycle of atmospheric CO₂ (Randerson et al.,1997; Navison et al., 2008). Systematic changes in the magnitude and seasonal distribution of these sources and sinks, as well as changes in atmospheric transport, may also drive trends in the seasonal cycle of CO₂(Randerson et al.,1997).

Fig. 3. Monthly carbon flux from terrestrial ecosystems to the atmosphere from 1997 to 2006(labeled on the left in g CO₂ m⁻² month⁻¹), compared to CO₂ concentration over terrestrial ecosystems(labeled on the right). The correlation between carbon flux and CO₂ level was 0.55 and 0.60(*p*<0.01) for NEP and GPP, respectively.

3.2 Annual mean value and long-term trend of variation of ACTE

In addition to the trend and the annual cycle, ACTE also shows interannual variability. An increasing trend, consistent with the observation sites in MLO and other measurement stations of the World Meteorological Organization's Global Atmosphere Watch program (WMO/GAW) (Keeling et al., 1996; WMO, 2012), could be identified from the observations and the best-fitting curves(Fig.2b). Regarding the

parameters presented in Eq.(1), the annual growth rate of ACTE, as indicated by parameter b , was estimated to be 2.04 ppm yr^{-1} , which was basically equal the average growth rate of global CO_2 concentration in the past decade ($\sim 2.0 \text{ ppm yr}^{-1}$) (Hofmann et al., 2009; WMO, 2012), primarily because of emissions from combustion of fossil fuels, cement production and land-used change in Northern America and Europe(carbon emissions were 7.8 to 9.5 Gt C yr^{-1} during 1997-2006, according to <http://www.globalcarbonproject.org>, see Table 2). This value was slighter higher than other reported results in natural vegetation, e.g., 1.7 ppm yr^{-1} for a tall forest in China (Wu et al.,2012); 1.8 ppm yr^{-1} for a temperate deciduous forest at the southernmost part of Japan (Murayama et al., 2003; Zhang et al., 2007); and 1.7 ppm yr^{-1} at two European coastal stations(Sirignano et al., 2010). But this was a relatively low value when compared to three regional measurements stations of WMO/GAW in China's economically developed region with large carbon emission, where CO_2 concentration typically increased at a rate of 3.3 ppm yr^{-1} from 2009 to 2011(Fang et al., 2014).

Table 2 Global averaged CO_2 concentration, global carbon sink/source emission and their mean annual growth rate from 1997 to 2006.

It should be noted that the annual average ACTE was higher than that in MLO by 2.5 ppm yr^{-1} . The most likely reason for the higher CO_2 concentration at EC sites was that most studied sites were located in the regions with strong human activities. Among the 104 studied sites, $\sim 85\%$ of the sites located in European Union (EU28, 63 sites) and America(USA, 31 sites), which were the dominated region for global CO_2 emissions. According to the results reported by Global Carbon Project (<http://www.globalcarbonproject.org>), the global cumulative CO_2 emissions from fossil-fuel and cement production during 1870-2013 were mainly distributed in USA(26%) and EU28(23%), covering 49% of the total share. Model results indicated that, during the 1990s, terrestrial ecosystems accounted for about 25-35% of the total emissions (land plus fossil fuels)(Ballantyne et al., 2012; Le Quéré et al.,2009). About 40% to 45% of total CO_2 emissions have remained in the atmosphere. In addition, there was no significant influence of carbon fluxes(i.e., NEP and GPP) on atmospheric CO_2 at interannual scale in this study. These results showed that although terrestrial act as a sink for fossil-fuel emissions, and thereby slowing the rise of

atmospheric carbon dioxide concentrations (Poulter et al., 2014), the trends in atmospheric CO₂ in Northern Hemisphere were likely to depend more on fossil fuel emissions than on ecosystem change.

3.3 Evolution and variation of CO₂ concentrations at individual ecosystem

Since the number of valid observation sites and ecosystem types were not consistent among different years, we chose the CO₂ measurements in 2004 and 2005(with 132 sites from 19 countries) to show the variability of CO₂ concentrations at individual ecosystem. The CO₂ concentration showed variation among ecosystems(Fig.4). The high value appeared in DBF, EBF and CRO. As DBF mainly located in mid-latitude and high-latitude regions, the probable reason for the high CO₂ concentration appeared in DBF might related to the limited growing season and the added demands for heating in winter(Rotty,1987). For CRO, which has a shorter and more intense carbon uptake periods (Zeng et al.,2014), are more affected by human activities such as land use change, fertilizer use, rice production, biomass burning(IPCC,2007), and other non-energy activities, which then leading to release carbon to the atmosphere(Gray et al., 2014). The differences among the forest, grassland and savanna were most likely due to the regional carbon emissions (Zhou et al., 2005; Wu et al., 2012). Most forest measurement sites in this study located in Europe and Northern America, in which intensive human activities and large amount of carbon emissions were reported (Le Quéré et al.,2009; Qu et al., 2013). This result could be interpreted by the comparison of atmospheric CO₂ over terrestrial ecosystems and urban area, which were responsible for the largest proportion of anthropogenic emissions in the world (Churkina, 2008). The difference between intensive human activity areas and natural ecosystems was similar to the increase in CO₂ concentration found in Phoenix, USA, where an increase of 111-185 ppm was reported from a pristine rural site to the city center (Idso et al., 1998, 2001).

Fig. 4. Box diagram of annual CO₂ concentration during 2004-2005 for each vegetation type. Black horizontal bars correspond to the lower and upper quartile values. The red lines and dot showed the median and means, respectively.

In order to capture the seasonal and annual evolutions of CO₂ concentration at

individual ecosystem, the harmonic model was optimized with observation data of each ecosystem over the whole study period, with the model performance and fitted parameters shown in Fig.5 and Table 3. There were long-term and continuous measurement in forest ecosystems (with n=120,120,84,72 for DBF, ENF, MF, EBF), from which the variation tendency of CO₂ could be well explored, with coefficient of determination(r^2) ranging from 0.43 to 0.83. However, the r^2 for WSA was low ($r^2=0.40$), mainly due to the limited observation sites at this biome in Northern Hemisphere(i.e., the number of observation sites for WSA was 5 in this study). As illustrated in Table 3, positive b indicated upward trend of annual CO₂ concentration were estimated for almost all studied ecosystems. The maximum b of 0.41 appeared in OSH, followed by DBF, WET and CRO, with the lowest b appeared in EBF and GRA. These results implied atmospheric CO₂ trend over ecosystems in dry, cold or high-elevation zones might be more vulnerable to the global climate change than that in better hydrothermal conditions. It was noting that the simulated seasonal cycle of some ecosystems, especially OSH, was different compared to the other ones. This was partly related to the limited observation sites and the inconsistent observation period among tower sites, which led to the poor fit with harmonic model. The seasonal cycle and evolution CO₂ concentration over these ecosystems still need further study with more CO₂ observations.

Fig. 5. Evolutions of monthly CO₂ means over different ecosystems during the study period. The black circles depicted the observed monthly CO₂ means. The solid red lines denoted the simulated monthly CO₂ values through harmonic model showed in Eq.(1).

Table 3. The optimized parameters of harmonized model for each vegetation type. The coefficient of determination(r^2), root mean square error (RMSE), and sample numbers (n) were also shown.

The last harmonic component yielded increase in the seasonal cycle of 0.36-2.04 ppm yr⁻¹ for different ecosystems. The seasonal CO₂ amplitude increase was higher in forest ecosystems(MF, EBF and EBF), whereas there is no distinguishable amplitude change in WSA and GRA. The same pattern appeared in Graven et al.(2013), with aircraft-based observation of CO₂. Their results showed that an increase of 30% to

60% in the seasonal exchange of CO₂ by northern extratropical land ecosystems, focused on boreal forests. It was noting that the increase in seasonal amplitude CO₂ may be closely related the the sink of CO₂ in northern extratropical land ecosystems(Pan et al., 2011).The relationship depends on how the sink has changed over time, which are very likely linked to the El Nino-Sourthern Oscillation(ENSO) (Artuso et al.,2009; Fang et al.,2014). Strong ENSO could be led to higher winter temperatures and an advanced spring season. This lead to increased respiration during winter and increased photosynthesis during spring(Gillett et al.,2002; Zeng et al.,2005).

4. Conclusion

Long-term continuous atmospheric CO₂ concentration obtained with eddy covaraince method provided additional information for better understanding the temporal and spatial dynamics of atmospheric CO₂ variability. Using the 10-year atmospheric CO₂ observations from FLUXNET and ChinaFLUX, this study demonstrated that atmospheric CO₂ over terrestrial ecosystems exhibited a strong seasonal variations, with an average seaonsal amplitude (peak-trough difference) of 14.8 ppm, which was approximately threefold that global mean CO₂ observed in Mauna Loa in the United States. The influence of carbon fluxes(i.e., NEP and GPP) over terrestiral ecosystems was apparent. However, the influence of carbon fluxes on CO₂ were not significant at interannual scale, implying the inter-annual changing trends of atmospheric CO₂ in Northern Hemisphere were likely to depend more on anthropogenic CO₂ emissions sources than on ecosystem change.

The application of a harmonic model fits the monthly observed CO₂ data. For the whole study period, both annual mean and seasonal amplitude of ACTE increased at rates of 2.04 and 0.6 ppm yr⁻¹, respectively. The uptrend of annual ACTE could be attributed to the dramatic global increase of CO₂ 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 CO₂ concentration showed large variation among ecosystems.

Due to data restrictions, our analysis centered mainly on atmospheric CO₂ concentration in Northern Hemisphere, the uneven distribution of observation sites

among terrestrial ecosystems, climatic zones and different years partly limited our analysis of the seasonal variations and annual evolutions. A more extensive and unified CO₂ observation network and long-term measurements will be necessary to better understand short-term variability and long-term evolution of atmospheric CO₂ over terrestrial ecosystems.

Acknowledgments

We are very grateful for two anonymous reviewers in providing valuable and helpful suggestions for this paper. This study was supported by the National Science Foundation of China (41201092, 41471076). We thank all the data provider. This work used eddy covariance data acquired by the FLUXNET community and in particular by the following networks: AmeriFlux (U.S. Department of Energy, Biological and Environmental Research, Terrestrial Carbon Program (DE - FG02 - 04ER63917 and DE - FG02 - 04ER63911)), AfriFlux, AsiaFlux, CarboAfrica, CarboEuropeIP, CarboItaly, CarboMont, ChinaFlux, Fluxnet - Canada (supported by CFCAS, NSERC, BIOCAP, Environment Canada, and NRCAN), GreenGrass, KoFlux, LBA, NECC, OzFlux, TCOS - Siberia, USCCC. We acknowledge the financial support to the eddy covariance data harmonization provided by CarboEuropeIP, FAO - GTOS - TCO, iLEAPS, Max Planck Institute for Biogeochemistry, National Science Foundation, University of Tuscia, Université Laval and Environment Canada and US Department of Energy and the database development and technical support from Berkeley Water Center, Lawrence Berkeley National Laboratory, Microsoft Research eScience, Oak Ridge National Laboratory, University of California - Berkeley, University of Virginia.

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Table 1. Distribution of sites(site-years) with respect to climate and vegetation classes. Climate was defined according to aggregated Koppen-Geiger classification (Rubel& Kottek, 2010), Vegetation classes (top line) are from IGBP definitions: 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). The numbers in the brackets represented the site-years.

Number of sites(year) available	CRO	DBF	EBF	ENF	GRA	MF	OSH	WET	WSA	Total
Equatorial	0	0	1(2)	0	0	0	0	0	0	1(2)
Arid	0	0	0	1(6)	1(2)	0	0	0	0	2(8)
Warm temperate	11(20)	14(52)	6(19)	17(71)	20(57)	3(13)	2(3)	1(2)	2(6)	76(243)
Snow	3(8)	5(19)	0	12(58)	0	2(6)	2(6)	1(5)	0	25(102)
Total	14(28)	19(71)	7(21)	30(135)	21(59)	5(19)	4(9)	2(7)	2(6)	104(355)

Table 2. Global averaged CO₂ concentration, global carbon sink/source emission and their mean annual growth rate from 1997 to 2006.

Year	CO ₂ concentration (ppm) ^a			Global Carbon Budget (Gt C yr ⁻¹) ^b			
	FLUXNET	Marine sites ^c	Mauna Loa	Carbon emissions	Atmospheric Growth	Ocean Sink	Land sink
1997	365.2	362.9	363.7	8.8	4.2	2.2	2.5
1998	367.0	365.6	366.7	8.2	6.0	2.3	-0.1
1999	368.8	367.6	368.3	7.8	2.8	2.2	2.8
2000	375.7	368.9	369.5	7.9	2.6	2.1	3.1
2001	376.2	370.4	371.1	7.8	3.8	1.9	2.1
2002	378.5	372.4	373.2	8.0	5.1	2.5	0.5
2003	378.4	375.0	375.8	8.3	4.7	2.4	1.1
2004	377.0	376.8	377.5	8.8	3.4	2.3	3.1
2005	379.5	378.8	379.8	9.2	5.1	2.4	1.6
2006	385.8	380.9	381.9	9.5	3.7	2.5	3.3

^aThe annual mean CO₂ concentration of Marine sites and Mauna Loa was based on globally averaged marine surface data and observation in Mauna Loa provided by Ed Dlugokencky and Pieter Tans, NOAA/ESRL (www.esrl.noaa.gov/gmd/ccgg/trends/).

^bGlobal Carbon Budget was provided by Global carbon project(2013) (<http://www.globalcarbonproject.org>), with global carbon emissions from fossil fuels, cement production and land-use change.

^cThe CO₂ concentration record for marine sites was measured by Global Monitoring Division of NOAA/Earth System Research Laboratory from a globally distributed network of air sampling sites described in Conway et al.(1994) (<http://www.esrl.noaa.gov/gmd/ccgg/trends/global.html>).

Table 3. The optimized parameters of harmonized model for each vegetation type. The coefficient of determination(r^2), root mean square error (RMSE), and sample numbers (n) were also shown.

Ecosystem	a	b	c	θ_1	d	θ_2	r^2	RMSE	n
CRO	362.78	0.21	2.66	0.16	0.04	0.83	0.67	3.97	60
DBF	366.38	0.24	5.35	-0.33	0.08	1.30	0.74	5.82	120
EBF	385.56	0.02	-9.52	0.57	0.17	0.45	0.43	5.19	72
ENF	364.11	0.16	5.37	0.47	0.03	1.42	0.83	3.30	120
GRA	383.00	0.00	7.81	0.69	0.02	-0.32	0.48	6.45	72
MF	378.82	0.00	3.79	3.86	0.17	0.69	0.56	6.29	84
OSH	339.69	0.41	17.56	-0.57	-0.18	-0.73	0.76	4.28	60
WET	360.08	0.22	-1.44	-20.21	0.06	0.80	0.54	6.40	71
WSA	362.57	0.19	6.21	0.70	0.01	-0.05	0.40	7.00	57
ALL	365.45	0.17	4.54	0.17	0.05	1.19	0.84	3.47	120
MLO	362.88	0.16	-2.75	-0.97	0.00	-1.19	0.99	0.77	120

Fig. 1. Geographical distribution of the study sites

Fig.2. Evolutions of the daily (a) and monthly (b) CO₂ means over the entire valid FLUXNET sites and Mauna Loa (MLO) from 1997 to 2006. The black circles depicted the observed CO₂ daily means (in upper panel) and monthly means (in the lower panel) from FLUXNET. The blue circles depicted the monthly observed CO₂ (in the lower panel) from MLO. The red and blue lines in lower panel denote the simulated monthly CO₂ values for EC sites and MLO, respectively, using Eq.(1).

Fig. 3. Monthly carbon flux from terrestrial ecosystems to the atmosphere from 1997 to 2006 (labeled on the left in g CO₂ m⁻² month⁻¹), compared to CO₂ concentration over terrestrial ecosystems (labeled on the right). The correlation between carbon flux and CO₂ level was 0.55 and 0.60 ($p < 0.01$) for NEP and GPP, respectively.

Fig. 4. Box diagram of annual CO₂ concentration during 2004-2005 for each vegetation type. Black horizontal bars correspond to the lower and upper quartile values. The red lines and dot showed the median and means, respectively.

Fig.5. Evolutions of monthly CO_2 means over different ecosystems during the study period. The black circles depicted the observed monthly CO_2 means. The solid red lines denoted the simulated monthly CO_2 values through harmonic model showed in Eq.(1).