Improving the CO₂ storage measurements with a single profile system in a tall-dense-canopy temperate forest

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ABSTRACT

The CO₂ storage \( F_s \) measurement can contribute significantly to the estimation of the net ecosystem exchange of CO₂ (NEE) especially in tall-canopy forest ecosystems. The \( F_s \) is often measured with a profile of CO₂ concentration or with an eddy covariance system at the tower top, but few studies investigated potential errors in the \( F_s \) measurements. We assessed the errors in \( F_s \) relevant to the vertical distribution of sampling levels and window sizes of averaging time of CO₂ mixing ratio and their effects on NEE in a temperate deciduous forest site in Northeast China using the standardized major axis method. The CO₂ storage per unit height typically decreased with the height increasing, suggesting that the below-canopy layer need a higher spatial resolution. CO₂ storage could be underestimated by up to one third based only on the tower-top measurement. The uncertainty (standard deviation) of the \( F_s \) decreased with the length of the averaging time window increasing. However, taking time averaging of CO₂ mixing ratio caused significant underestimate of \( F_s \), and consequently led to significant underestimates of CO₂ uptake and release at a 30-min time scale. Our results highlight that appropriately combining spatial resolution and temporal resolution (response time for a whole sampling of all levels) is essential to improving the \( F_s \) estimates with the existing CO₂ profile systems in forest ecosystems. Since the systematic bias and random error in \( F_s \) estimate with a single profile system are irreconcilable, there is an urgent need to develop a fast response planar-averaging profile or measure the instantaneous vertical-mean concentration to improve the accuracy of \( F_s \) measurement.

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1. Introduction

Rapid development of the FLUXNET network (Baldochi et al., 2001) has greatly improved our understanding of carbon and water fluxes in terrestrial ecosystems in recent years (Papale et al., 2015). The eddy covariance (EC) method provides direct flux measurements of trace gas and energy across the atmosphere–ecosystem interface without disturbing the vegetation and the soil (Aubinet et al., 2012). There are more than 600 active sites globally (Papale et al., 2015), among which dozens have more than a decade of the duration of the time series (Baldochi, 2014). Generally, the net ecosystem exchange of CO₂ (NEE) can be estimated by summing the vertical eddy flux \( F_v \), the advection term, and the storage term \( F_s \) (Aubinet et al., 2005). The \( F_v \) can be a close estimation of NEE under ideal conditions (horizontal homogeneity, enough footprint, flat terrain or after appropriate axis choice, strong turbulent mixing, and steady state conditions) (Aubinet et al., 2012; Baldocihi, 2003). However, the conditions in field are rarely ideal, which leads to frequent underestimation of NEE due to ignoring considerable contributions of the advection and \( F_s \) especially in tall–canopy forest ecosystems (Papale et al., 2006; Yang et al., 2007).

Great efforts have been made on improving the accuracy of \( F_v \) and advection (Aubinet et al., 2012), but few studies investigated the errors of \( F_s \). The \( F_s \) can be significant at short time periods such as half-hours, particularly around sunrise, sunset, or at night (e.g., Dolman et al., 2002; Finnigan, 2006; Yang et al., 2007). Therefore, the \( F_s \) should be taken into account because of potential error propagation in the gap-filling processing for annual NEE estimation (Papale et al., 2006). As long as the \( F_s \) is properly tackled, potential underestimations of NEE on calm nights can be greatly reduced after the critical friction velocity filtering (Papale et al., 2006; Yang et al., 2007) or by using the data in the early evening period (van Gorsel et al., 2007).

The first objective of this study is to assess the effect of vertical configuration of a CO₂ profile system on \( F_s \) estimates. An essential issue of \( F_s \) is the uncertainty caused by the spatial and temporal

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variability in CO2 concentration. The \( F_3 \) for a single flux tower design is often estimated from a single vertical profile of CO2 concentration (Finnigan, 2006). A literature review highlighted that there were substantial differences in sampling designs of CO2 concentration profile systems, e.g., the number and vertical distribution of sampling levels, time needed for a complete profile measurement, and the percentage of the effective measurement time (Table S1). The simplest method for estimating \( F_3 \) is based on the change in CO2 density (or molar mixing ratio) measured by the EC system (the tower-top method) if a profile is not available. Some researchers assume that estimates of \( F_3 \) based on tower-top and profile measurements are interchangeable (Carrara et al., 2003; Hollinger et al., 1994; Knohl et al., 2003), thus the tower-top method is widely used when profile measurements are not available. However, others report that the tower-top method substantially underestimates \( F_3 \) compared with the profile system (Gu et al., 2012; Iwata et al., 2005; Yang et al., 2007, 1999). Then how many measurement levels are required to accurately quantify the CO2 mixing ratio profile may depend on canopy complexity and height (Munger et al., 2012). For example, Yang et al. (2007) reported that more sampling levels were needed at the Missouri Ozark flux site in order to achieve the same level of accuracy as at a boreal aspen site (Yang et al., 1999), perhaps because of the difference in their canopy complexity (vertical complex vs. simple) (Yang et al., 2007). But it is not clear whether the difference in topography (e.g., the top of a ridge vs. flat terrain) contributes to it. In this study, we investigated the vertical configuration of an eight-level profile in a temperate forest at the Maershan flux tower site in Northeast China with vertical complex canopy structure over the toe slope in a small valley. We will examine the relative importance of the complexity in canopy structure and topography on determination of the number of sampling levels and their vertical distribution.

Our second objective is to quantify the uncertainty (random error) in \( F_3 \) due to discrete measurements for each level by a single infrared gas analyzer (IRGA). Many researchers use the design of single-IRGA with multiple inlets for CO2 concentration gradient to remove the systematic errors between IRGAs (Table S1; Munger et al., 2012). The air from each inlet (height level) is sequentially flowed through a manifold and measured with an IRGA (Munger et al., 2012; Xu et al., 1999). The actual measurement from each inlet is thus discrete, and the effective measurement time for each height is much shorter than the averaging period (varying from 8% to 80% for single profile systems, Table S1). Because of the wavelike fluctuation of CO2 concentration, the instantaneous or small time-window averaged CO2 concentration may introduce large uncertainty in \( F_3 \) (Heinesch et al., 2007; Marcolla et al., 2014; van Gorsel et al., 2011). van Gorsel et al. (2011) reported that the uncertainty caused by the wavelike motion of CO2 concentration and discrete measurement was approximately 0.9 μmol m⁻² s⁻¹ in an open-canopy forest, which was large compared to the \( F_3 \) of 0.3 μmol m⁻² s⁻¹. Compared to the open-canopy forest, we will quantify the uncertainty of \( F_3 \) measured with a profile system in our dense-canopy forest.

The third objective is to test whether time averaging of CO2 concentration underestimates the \( F_3 \) and consequently NEE. Theoretically, the \( F_3 \) is the change rate of the instantaneous CO2 concentration integrated in the control volume (Finnigan, 2006). For a single flux tower, it is the difference between instantaneous CO2 concentration profiles at the tower measured at the beginning and end of the flux-averaging period, divided by the length of this period (Finnigan, 2006; Xu et al., 1999). The instantaneous CO2 concentration profile for a single tower can be very noisy due to wind gusts and may not represent the average concentration of the whole control volume. To reduce the random error caused by insufficient spatial sampling, researchers adopt the strategy of replacing the spatial averaging by time-averaging (Finnigan, 2006). However, Finnigan (2006) inferred that the time-averaging procedure can underestimate \( F_3 \) by at least 50% in most conditions because of filtering high frequency information. Using the single-point high-frequency CO2 density measurement by the EC system, Yang et al. (2007) qualitatively verified the theoretical inference by Finnigan (2006). In contrast, Ohkubo et al. (2008) argued that the difference in \( F_3 \) calculated based on a profile system by different sizes of time-averaging window was negligible. Whether the discrepancy is caused by the difference between single-point and profile measurements needs further studies. We will explore how the time averaging of CO2 concentration influences the \( F_3 \) and NEE.

In this study, we extended the work conducted by Yang et al. (1999) and Yang et al. (2007), and conducted our study in a temperate broad leaved deciduous forest at the Maershan Forest Ecosystem Research Station in Northeast China. The fast cycling (one cycle of sequential measurements over the eight levels is completed in 2 min) eight-level profile (API100, Campbell Scientific Inc., Logan, UT, USA) (Table S1; Wang et al., 2013b) provides us an opportunity to quantify uncertainty due to the discrete measurement and time averaging of CO2 concentration. The hilly terrain and wind regime are common properties in forest ecosystems. A classic mountain-valley wind system over forest canopy was previously found at the Maershan site (Wang et al., 2015). The \( F_3 \) also had a typical diurnal variation in the forest ecosystem, with the negative peak occurring in the early morning and the positive maximum in the early evening (Wang et al., 2013b). Therefore, assessing the effects of sampling strategy and time averaging of the profile system on estimating \( F_3 \) at the Maershan site is of importance for assessing the NEE of forest ecosystems in Northeast China, and also has implications for other forest sites. We calculated

| Nomenclature | Definition |
|--------------|------------|
| \( \chi_c \) | Molar mixing ratio of CO2 (μmol mol⁻¹) |
| \( c_c \) | Molar fraction of CO2 (μmol mol⁻¹) |
| \( c_v \) | Molar fraction of water vapor (mmol mol⁻¹) |
| \( \rho_a \) | Moist air molar density (mol m⁻³) |
| \( \rho_c \) | CO2 density (mg m⁻³) |
| \( \rho_d \) | Molar density of dry air at 36 m (mol m⁻³) |
| \( \Delta X_c / \Delta t \) | Time derivative of CO2 mixing ratio |
| \( \Delta t \) | Time interval between the two samplings (1800 s) |
| \( F_c \) | Vertical eddy flux (μmol m⁻² s⁻¹) |
| \( F_s \) | Storage term (μmol m⁻² s⁻¹ or μmol m⁻³ s⁻¹) |
| \( F_s / \rho \) | Storage term by the tower-top method (μmol m⁻² s⁻¹) |
| \( F_s / \rho \delta \) | Storage term by the top level of the profile system (μmol m⁻² s⁻¹) |
| \( H \) | Height of the EC system (36 m) |
| IRGA | Infrared gas analyzer |
| LAI | Leaf area index (m² m⁻²) |
| \( M \) | Number of time windows for averaging of CO2 mixing ratio in a flux averaging period (30 min) |
| \( M_c \) | Molar mass of CO2 |
| \( N \) | Number of sampling levels of a profile system |
| NEE | Net ecosystem exchange of CO2 (μmol m⁻² s⁻¹) |
| OLS | Ordinate least square |
| \( P \) | Length of the window (min) |
| SD | Standard deviation |
| SMA | Standardized major axis |
F\textsubscript{3} based on CO\textsubscript{2} mixing ratio instead of CO\textsubscript{2} density (Yang et al., 2007, 1999) based on previous studies (Gu et al., 2012; Lee and Massman, 2011; Leuning, 2007; Wang et al., 2013b). The difference in estimated F\textsubscript{3} between subprofile and the eight-level profile baseline was assessed by the standardized major axis (SMA) approach (Warton et al., 2006), rather than the ordinary least square (OLS) regression (Bjorkgren et al., 2015; Yang et al., 2007, 1999) that is inappropriate to assess the difference between two estimates. Our objectives are to: (1) explore the effect of vertical configuration on F\textsubscript{3} estimates and test if there is any systematic difference in estimates of F\textsubscript{3} between the tower-top method and the profile system, (2) assess the uncertainty of F\textsubscript{3} due to discrete sampling, and (3) quantify the impact of the length of averaging time window on F\textsubscript{3}.

2. Materials

2.1. Site description

The EC flux tower was established in 2007 at the Maershan Forest Ecosystem Research Station of Northeast Forestry University, Heilongjiang Province, Northeast China (45° 24' N, 127° 40' E, 400 m a.s.l.). The climate is a continental monsoon climate with a windy and dry spring, a warm and humid summer, and a dry and cold winter (Wang et al., 2013a). The mean (1989–2009) annual air temperature is 3.1 °C, and the mean January and July air temperatures are −18.5 °C and +22.0 °C, respectively. The mean annual precipitation is 629 mm, of which ~50% falls between June and August. The mean slope around the tower is ~9°, and the slope of the valley center is ~1° along the valley (Wang et al., 2015).

The forest around the EC flux tower was about 60-year-old, with a canopy height of 18–20 m. The dominant tree species include Ulmus japonica Sarg., Fraxinus mandshurica Rupr., Betula platyphylla Suk., Populus davidiana Dode, Juglans mandshurica Maxim., etc. Based on the inventory of 106 circular plots (with diameters of 20 m) within the fetch (1500 m × 400 m), the mean basal area and tree biomass density were 24.16 m\textsuperscript{2} ha\textsuperscript{-1} and 155.64 Mg ha\textsuperscript{-1}, respectively (Liu et al., 2016). The maximum leaf area (semi-surface leaf area) index (LAI) of the canopy, measured with the litter fall collection method, was about 5.7 m\textsuperscript{2} m\textsuperscript{-2} in eight 20 × 30 m permanent plots (Wang et al., 2015). A 1–8 m high and well-developed understory is dominated by Syringa reticulata var. mandshurica, which contributed 27% to the stand LAI (Wang et al., 2015).

2.2. Instrumentation and data collection

An open-path eddy covariance system (LI-7500, Li-Cor Inc, Lincoln, NE, USA; CAST3, Campbell Scientific Inc, USA) was installed at the 36 m height. The CO\textsubscript{2}/H\textsubscript{2}O concentrations were measured at 0.5, 2.0, 4.0, 8.0, 16.0, 20.0, 28.0, and 36.0 m above the ground surface using the AP100 (Campbell Scientific Inc, USA). The AP100 was designed similar to the system by Xu et al. (1999). The profiling system included the following components: an air sampling system, a calibration and flow control system, a closed-path IRGA (LI-840, Li-Cor Inc, USA), and a datalogger (CR1000, Campbell Scientific Inc, USA).

Each intake had a coarse filter with a rain diverter and a 7 μm filter. A precision orifice in each line was used to set the flow rate and the pressure in the system. And a 0.5 W heater was used to prevent condensation. The tubings had an inner diameter of 1/4 inch, and were made of high-density polyethylene bonded to an overlapped aluminum tape with an ethylene copolymer coating. The polyethylene liner reduced the interaction of water vapor with the walls of the tubing.

The pressure in the tubing and IRGA was maintained approximately at half of the local atmospheric pressure to further prevent condensation and reduce the interaction of the water vapor with the walls of the tubing, which reduced the settling time after switching to the new level. Air was drawn continuously from eight heights through tubings of equal length and diameter, each connected to a three-way solenoid valve mounted on a manifold. The flow from each inlet was directed either to the analyzer (only when the level is selected) or the pump, so that the system was constantly flushed and followed the ambient concentration changes when the corresponding level was not selected. The flow from only one intake line went through the analyzer at one time, which was further filtered with a 1 μm pore size PTFE filter (Acro50, Pall Corporation, MI, USA). The valves were switched sequentially every 15 s; the first 7 s reading was removed to purge the tubings and the path of IRGA to getting an equilibration; and the reading in the next 8 s was recorded with a datalogger. The 7 s’ delay before logging the data was based on the apparatus equilibration time (less than 5 s), which allowed the system to complete one cycle of sequential measurements over the eight heights levels within a short interval (a 2-min cycle) and 15 cycles in a flux-averaging period (30 min). The datalogger recorded the CO\textsubscript{2} and H\textsubscript{2}O concentrations (molar fraction) at 2-min and 30-min time intervals. The calibration system, controlled by the datalogger, compensated for any deviations in the calibration of the IRGA with the gas sources of known concentrations (zero and span of CO\textsubscript{2}). The LI-840 IRGA was automatically calibrated for zero and span of CO\textsubscript{2} once a day at 10:00 a.m., and also manually calibrated once a month to remove potential long-term drifts.

The temperature/relative humidity profile was composed of five HMP45C probes (Vaisala, Helsinki, Finland) with aspirated radiation shield (0768, Met One Instruments Inc., USA) at heights of 2.0, 16.0, 28.0, 36.0 and 48.0 m. Air temperature and relative humidity were measured once every two seconds for all heights and recorded the 30-min averages. The barometric pressure was measured at 28 m above the ground surface.

3. Data processing

The CO\textsubscript{2} molar fraction is constrained in the ranges between 300 μmol mol\textsuperscript{-1} and 900 μmol mol\textsuperscript{-1} within the canopy (below 20 m above the ground), and between 300 μmol mol\textsuperscript{-1} and 600 μmol mol\textsuperscript{-1} above the canopy (Jiao et al., 2011). The F\textsubscript{3} out of the range (between −45 μmol m s\textsuperscript{-1} and 45 μmol m s\textsuperscript{-1}) was also excluded in further analysis. The CO\textsubscript{2} profile and F\textsubscript{3} data during precipitation were included because the closed IRGA was not influenced by precipitation (Wang et al., 2013b), while the LI-7500 data during precipitation were excluded. The data coverage rates were 95% for the profile and 80% for the LI-7500 after the quality control. Data gapfilling was not implemented because we were focusing on the flux density and avoiding other uncertainties caused by gapfilling (Wang et al., 2013b). To explore the effect of canopy density on F\textsubscript{3}, we divided the calendar year into two seasons: the leaf-on season (11 May to 5 October) and leaf-off season (the other times) (Wang et al., 2015) based on the canopy phenology (Liu et al., 2015).

3.1. Calculation of CO\textsubscript{2} storage

The dry molar fraction of CO\textsubscript{2} (molar mixing ratio relative to the dry air) was adopted to calculate the F\textsubscript{3} (Gu et al., 2012; Kowalski, 2008; Leuning, 2007), because it was conservative to expansion and contraction of the air column below the measurement height (Kowalski and Serrano-Ortiz, 2007; Lee and Massman, 2011). At the Maershan site, Wang et al. (2013b) found using CO\textsubscript{2} density to calculate the storage could overestimate 8.5%, but the error of using mixing ratio was only 0.1%. For the tower-top measurement, the output of CO\textsubscript{2} concentration of the LI-7500 was gas density,
which was firstly converted to CO2 molar mixing ratio ($\chi_c$, \(\mu\text{mol mol}^{-1}\)) as Wang et al. (2013b),

$$\chi_c = \frac{\rho_c \times 10^3}{M_c \rho_a (1 - c_v \times 10^{-3})}$$  \hspace{1cm} (1)

where $\rho_c$ is CO2 density (mg m\(^{-3}\)), $M_c$ is molar mass of CO2 (44.01 g mol\(^{-1}\)), $\rho_a$ is moist air molar density (mol m\(^{-3}\)), and $c_v$ is molar fraction of water vapor (mmol mol\(^{-1}\)). Then the $F_s$ estimated by the tower-top method ($F_{s,EC}$, \(\mu\text{mol m}^{-2} \text{s}^{-1}\)) was calculated as,

$$F_{s,EC} = \frac{\Delta \chi_c}{\rho_a} \Delta z \Delta t$$  \hspace{1cm} (2)

where $\rho_a$ is the molar density of dry air at the 36 m (mol m\(^{-3}\)), $\Delta \chi_c$ is the change in the molar mixing ratio of CO2 at the EC height (\(\mu\text{mol mol}^{-1}\)), $\Delta t$ is the time interval between the two samplings (1800s), and $h$ stands for the height of the EC system (36 m). For the profile system, the molar fraction of CO2 by the LI-840 was also transformed to molar mixing ratio as (Leuning, 2007; Montagnani et al., 2009),

$$\chi_c = \frac{c_c \times 10^{-6}}{1 - c_v \times 10^{-3}}$$  \hspace{1cm} (3)

where $c_c$ is molar fraction of CO2 (\(\mu\text{mol mol}^{-1}\)), and $c_v$ is molar fraction of water vapor (mmol mol\(^{-1}\)). And the $F_{s}$ estimated by the eight-level profile systems ($F_{s,p}$, \(\mu\text{mol m}^{-2} \text{s}^{-1}\)) was calculated as (Gu et al., 2012; Lee and Massman, 2011; Leuning, 2007),

$$F_{s,p} = \frac{\partial \chi_c}{\partial t} \int_0^h \frac{\partial \chi_c}{\partial t} dz = \rho_a \sum_i^8 \frac{\Delta \chi_c}{\Delta t} h_i$$  \hspace{1cm} (4)

where $\chi_c$ represents the molar mixing ratio of CO2 at each sampling height section (\(\mu\text{mol mol}^{-1}\)). $\Delta \chi_c/\Delta t$ between two adjacent levels were averaged to get a more representative for the layer. At the bottom level, the measurement right at the ground surface was linearly extrapolated based on the low two layers (0.5 and 2 m). The CO2 measurements at the levels where air temperature was not measured were obtained by applying linear interpolation to the two adjacent measurements (Yang et al., 2007).

3.2. Effect of vertical configuration of a profile system on CO2 storage estimates

The data set used in this section covered the whole year of 2009. The SMA line-fitting approach (Warton et al., 2006) was used to test the consistency between $F_s$ estimates by continuous (tower-top measurement by LI-7500, $F_{s,EC}$) and by discrete (36.0 m height of the profile, $F_{s,p,36m}$) samplings, and by tower-top ($F_{s,EC}$) and by profile ($F_{s,p}$) measurement. The effect of $F_s$ calculations based on $F_{s,EC}$ and $F_{s,p}$ on NEE was also assessed by the SMA approach. The SMA test is used because it is suitable for consistency test between two methods, while the OLS regression is often used for predictive purposes (Warton et al., 2006). We found that the OLS approach substantially underestimated the slopes of the fitted lines compared with those by the SMA approach (Supplementary material S1, giving an example of time-averaging effect on $F_s$). Similarly, Wilson et al. (2002) also found that the OLS tended to underestimate the slope for the energy balance, and the underestimation increased with $R^2$ decreasing.

The NEE was calculated as,

$$\text{NEE} = F_c + F_s$$  \hspace{1cm} (5)

where $F_c$ and $F_s$ are eddy and storage fluxes of CO2 (\(\mu\text{mol m}^{-2} \text{s}^{-1}\)), respectively. The advection term of NEE is omitted as did all single-tower designs (Aubinet et al., 2012). The $F_c$ data were processed with the flux measurement standard procedures, including despiking, time-lag removing, planar-fit tilt correction, frequency response correction, WPL correction, and quality control (Aubinet et al., 2012).

To examine the effect of vertical configuration of a profile on $F_s$, we used the eight-level profile system as a benchmark to test subsets of the full profile system as did Yang et al. (2007). The subset profiles were grouped based on the total number of measurement levels in each profile. Within a group, all subset profiles contained the same number of measurement levels but different combinations of measurement heights. The topmost level (36 m) was always included as the top boundary in all subset profiles except the one-level measurement. Again, the SMA method was used to test the possible biases of the subset profiles based on the slopes and intercepts of the fitted lines (Warton et al., 2006). Because the intercepts of $F_s$ for most combinations against the $F_{s,p}$ were within $[-0.04, 0.04 \mu\text{mol m}^{-2} \text{s}^{-1}]$ (except the 0.5 m height for one-level of $-0.08 \mu\text{mol m}^{-2} \text{s}^{-1}$), the slopes could be used to correct the biases of subset profile combinations.

3.3. Uncertainty of CO2 storage estimates due to discrete sampling

The data set in this section derived from the 2-min averaged CO2 molar fraction measured by the profile from 10 June 2008 to 31 December 2009 (the data from 2 July to 16 August of 2009 were missing). Based on the fast response profile system of AP100 (2-min for one cycle of all eight-level), we calculated $F_s$ for 15 time windows varying from 2 min to 30 min, which increased with a time step of 2 min. The number of measurements ($M$) of time windows for averaging of CO2 mixing ratio in a flux averaging period (30 min) depended on the length of the time window $[M=(30 - P)/2 + 1$, where $P$ is the length of the window (min)], e.g., 15 moving independent 2-min time windows, or 14 moving 4-min windows (allowing partly overlap between moving windows). We noted that calculating the $F_s$ for each flux-averaging period by averaging all independent or moving windows is inappropriate, because performing such averaging is identical to doing averaging CO2 concentration for all independent windows in the flux averaging period (Supplementary material S2). Therefore, we used the mean CO2 mixing ratio of the time window in the middle of a flux averaging period.

According to previous studies on uncertainty analysis (Heinesch et al., 2007; Marcolli et al., 2014), the standard deviation (SD) of the estimates for all windows in a given flux period was adopted as a measure of uncertainty of $F_s$ induced from discrete sampling. We performed no interpolation between cycles because any interpolation would introduce additional uncertainties (Bjorkgren et al., 2015).

3.4. Effect of time-averaging on CO2 storage estimates

The data set for this section was the same as that in Section 3.3. The recordings of CO2 concentrations for each cycle of a whole sampling of all levels gave us an opportunity to analyze the effect of time-averaging of CO2 concentrations on $F_s$ estimates using profile systems. The fast profile data had an advantage in spatial representation than the tower-top measurement of CO2 density from LI-7500 by Yang et al. (2007). The slopes and intercepts of the SMA fitted lines were used to assess the systematic bias of the magnitude of $F_s$. We also compared the NEE based on the 2-min-window $F_s$ and the 30-min-window $F_s$ using the SMA method.
depleted after sunrise. The $\chi_c$ was always higher below the canopy than above the canopy, with the greatest gradient of $\chi_c$ occurring at night in the leaf-on season. The $\chi_c$ rapidly decreased after sunrise. Because the depletion of $\chi_c$ was more dramatic near the ground surface due to turbulent mixing, the vertical gradient declined to its minimum around noon. The $\chi_c$ started to increase at the forest floor before sunset, and then extended to the whole canopy till the early evening. The diurnal evolution of $\chi_c$ was much less in the leaf-off season than the leaf-on season (Fig. 1b). Such diurnal variation in $\chi_c$ was similar to those in boreal (Yang et al., 1999), temperate (Leuning et al., 2008; Ohkubo and Kosugi, 2008; Yang et al., 2007) and tropical (Araújo et al., 2008; Hutyra et al., 2008; Ohkubo et al., 2008; Yao et al., 2012) forests, suggesting that the Maoershan flux site shared a common feature with many forest sites.

The magnitude of the time derivative of CO$_2$ mixing ratio ($\Delta \chi_c / \Delta t$) decreased from the forest floor to the 36 m (the EC system height; Fig. 2a and b). In the leaf-on season, the $\Delta \chi_c / \Delta t$ during the period of nocturnal stable boundary layer (21:30–5:30) was slightly positive (weakly accumulating CO$_2$); that in the morning transition phase (6:00–8:30) was strongly negative (strongly releasing CO$_2$); that in the period of daytime convective boundary layer (09:30–16:00) was slightly negative or near neutral; and that in the evening transition phase (16:30–21:00) was strongly positive (strongly accumulating CO$_2$) but smaller in magnitude than that in the morning transition phase. Although the magnitude decreased by one order, the daily change of the vertical profile was similar in the leaf-on season to that in the leaf-on season. The standard deviation (SD) of $\Delta \chi_c / \Delta t$ also decreased with height increasing (Fig. 2c and d), and its vertical gradient was more dramatic at night and in the morning transition phase. Vertically, both magnitudes of mean and SD of $\Delta \chi_c / \Delta t$ were higher below 8 m, intermediate between 8–20 m, and lower above 20 m. Therefore we separated the whole air column below the EC height into three vertical sections to evaluate the contributions of different layers to the total $F_{30}$: below-canopy layer (0–8 m), canopy layer (8–20 m) and above-canopy layer (20–36 m).

4. Results and discussion

4.1. Vertical distributions of CO$_2$ mixing ratio and storage

The seasonal averaged daily courses of CO$_2$ molar mixing ratio ($\chi_c$) profile (Fig. 1) showed that the CO$_2$ accumulated at night and

![Fig. 1. Diurnal variations in vertical contours of seasonal means of CO$_2$ molar mixing ratio (µmol mol$^{-1}$) during the leaf-on (a) and leaf-off seasons (b).](image)

![Fig. 2. Vertical profiles of the mean and standard deviation (SD) of the time derivative of CO$_2$ molar mixing ratio ($\Delta \chi_c / \Delta t$) during the leaf-on (a,c) and leaf-off seasons (b,d).](image)
The $F_\alpha$ per unit height also varied with vertical sections (Fig. 3) as did the mean $\Delta x / \Delta t$ (Fig. 2). The $F_\alpha$ per unit height in the below-canopy layer was higher than those above for both leaf-on and leaf-off seasons, particular at dusk and night. In the morning transition phase (6:00–8:30), the peak of below-canopy $F_\alpha$ was slightly lagged those in the upper layers (Fig. 3a). In the evening transition phase (16:30–21:00), the peak of $F_\alpha$ occurred earlier in the below-canopy layer than did those aloft. Volume percentages of the below-canopy, canopy, and above-canopy layers (22%, 33% and 44%, respectively) also influenced the contribution of specific vertical sections to the total $F_\alpha$. For example, during the night in the leaf-on season, the $F_\alpha$ per unit height increased from below-canopy, canopy, to above-canopy layer, with the corresponding contributions of 48.3%, 27.3%, and 24.4%, respectively (Table 1). Although the magnitude of $F_\alpha$ was much lower in the leaf-off season, the diurnal courses and relative magnitudes of $F_\alpha$ per unit height from the three sections were similar to those in the leaf-on season (Fig. 3b). Note that the data of leaf-off-night were within the typical measurement errors, thus the relative contribution of particular layer varied considerably than those of the other periods. During the night, the direction of the below-canopy $F_\alpha$ sometimes differed from those in the other two layers (Table 1), mainly due to the near neutral values in the dormant season (Wang et al., 2015). These results illustrated that the layer near the ground surface tended to contribute disproportionately higher to its height percentage than did the upper layers.

The magnitude of $F_\alpha$ in this study was well within the range in temperate forests (Aubinet et al., 2005; Gu et al., 2012). The vertical distribution of $F_\alpha$ at the Moflux site was consistent with that at the Moflux site (Yang et al., 2007), but differed from that at the Southern Old Aspen (SOA) site where more than 85% of the increase in $F_\alpha$ in late night occurred above 9 m (Yang et al., 1999). We speculate that this difference may mainly result from the difference in the complexity of canopy structure, probably not the distance to the ridge as previously considered for advection (Aubinet et al., 2005). The Moflux flux tower is located on the top of a ridge (Yang et al., 2007), while the Moflux flux tower is on the toe of a valley sidewall. The only common feature for canopy structure and terrain the Moflux and the Moflux sites is a complex canopy (well-developed understory layer). The SOA site had an open canopy structure (total LAI was 4.1 m2 m−2, with a 2-m-tall shrub layer contributed 40%, and a trunk space height of 9–15 m) and a relative flat terrain (Yang et al., 1999). In the late night, the acceleration of the wind weakened the subcanopy inversion and moderately increased whole-canopy turbulence mixing (Mahrt et al., 2000), accordingly the increase in $F_\alpha$ mainly occurred in the upper layer (Yang et al., 1999). This situation probably does not occur in dense-canopy forest sites where the radiational cooling is often at the canopy top and the subcanopy stratification might become neutral or even unstable at night (Wang et al., 2015). Therefore, the canopy structure probably plays a more important role in the vertical distribution of $F_\alpha$ than did the topography. To optimize the vertical distribution of a profile system for the $F_\alpha$ measurement, it is needed to consider canopy structure and thermal stratification, as well as the topography (Munger et al., 2012; Yang et al., 2007).

4.2. Effects of vertical configuration of a profile on CO2 storage estimates

Profile designs varied considerably at global forest sites, most of which had 4–12 levels that distributed from ground surface to 70 m height (Table S1). We assessed the bias of subprofiles by the regression slope of subprofiles against the SMA approach. At our site, the effectiveness ($R^2$) of the best configuration increased with the number of sampling levels (N) increasing (Fig. 4a), while the systematic bias on the magnitude of $F_\alpha$ (deviation of the slope from 1) decreased (Fig. 4b). The effectiveness of a configuration was too low to get a precise estimate of $F_\alpha$ if the sample size $N < 2$ (Table S2). When the $N$ was four or more, the subprofile with optimal vertical distribution could provide a reasonable estimate of $F_\alpha$ (Fig. 4 and Table S2). Yang et al. (2007) reported that when the $N$ equaled to four or less, any combination could not measure $F_\alpha$ accurately at the complex-canopy Moflux site, but three levels was good enough for the single-canopy SOA site (Yang et al., 1999). These results confirm that more sampling lev-

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Table 1

| Season     | Period                  | $F_\alpha$ (µmol m$^{-2}$ s$^{-1}$) |
|------------|-------------------------|-----------------------------------|
|            | 0–8 m       | 8–20 m   | 20–36 m  | 0–36 m   |
| Leaf-on    | Night (21:30–5:30) | 0.214 (48.3%) | 0.121 (27.3%) | 0.109 (24.4%) | 0.444 (100%) |
|           | Morning transition (6:00–8:30) | −0.151 (32.2%) | −1.593 (34.0%) | −1.584 (33.8%) | −4.688 (100%) |
|           | Day (9:00–16:00)    | −0.128 (31.5%) | −0.115 (28.3%) | −0.164 (41.2%) | −0.407 (100%) |
|           | Evening transition (16:30–21:00) | 0.737 (27.8%) | 0.920 (34.6%) | 1.000 (37.6%) | 2.657 (100%) |
| Leaf-off   | Night (20:30–6:30) | 0.006 (108.7%) | −0.003 (61.5%) | −0.008 (147.2%) | −0.005 (100%) |
|           | Morning transition (7:00–9:30) | −0.148 (41.9%) | −0.116 (32.8%) | −0.089 (25.3%) | −0.353 (100%) |
|           | Day (10:00–16:30)  | −0.014 (21.3%) | −0.023 (34.5%) | −0.029 (44.2%) | −0.066 (100%) |
|           | Evening transition (17:00–20:00) | 0.134 (30.8%) | 0.149 (34.4%) | 0.151 (34.8%) | 0.435 (100%) |

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Fig. 3. Diurnal sections in mean CO2 storage flux per unit height ($F_\alpha$) from the three vertical sections below the measurement height of eddy covariance during the leaf-on (a) and leaf-off seasons (b), 0–8 m: below-canopy layer; 8–20 m: canopy layer; 20–36 m: above-canopy layer.
els are needed in complex-canopy forests than in simple-canopy forests.

For a given N, the most effective combination (with the highest $R^2$) had a very low bias in $F_s$, but the combination with the lowest bias (as indicated by the slope and intercept of the fitted line between sub-combination and the baseline) was not necessary the most effective combination (Fig. 4, Table S2). So the biases are more important than the $R^2$ because our concern is to avoid any systematic error (non-unit slope and non-zero intercept). Using the OLS regression is better for prediction purpose than comparison of the difference in estimates between two methods using the SMA test in this study. Therefore we suspect that the criteria of previous studies using the OLS regression (e.g., Bjørkegren et al., 2015; Yang et al., 2007, 1999) might biased the errors of subprofiles, although the overall trends are similar. Combinations with the lowest biases in the magnitudes of $F_s$ and the highest effectiveness require sampling slightly denser in the subcanopy layer than that above, because of larger $\Delta x_c / \Delta t$ in this layer (Fig. 2). For a dense-canopy forest, it was preferred to allocate at least two levels under the canopy and two or more levels over the canopy (Table S2; Yang et al., 2007). To optimize a profile to measure the $F_s$, one needs to balance the $\Delta x_c / \Delta t$ and the layer thickness (Yang et al., 2007).

The diurnal variation in $F_{s,p36n}$ (discrete sampling by the profile) was consistent with the $F_{s,EC}$ (continuous sampling by the EC system), and similar to that of $F_{s,p}$ (Fig. 5). The $F_s$ reached its minimum in the morning, peaked in the early evening and declined rapidly until the late night, inconsistent with those by Aubinet et al. (2005). These results support the alternative method of friction velocity threshold for filtering the nocturnal NEE (Van Gorsel et al., 2009; van Gorsel et al., 2007).

We further quantified the difference in $F_s$ estimates between the tower-top method and the profile measurement. The SMA test between $F_{s,EC}$ and $F_{s,p36n}$ showed that the slope (0.929) was not significantly ($P > 0.05$) different from unit, and the intercept ($-0.059$) was neither significantly ($P > 0.05$) different from zero (Fig. 6). However, the $F_{s,EC}$ was systematically lower in magnitude than the $F_{s,p}$, with the greatest difference occurring during the morning transition phase (Fig. 5). The slope (0.654) was significantly ($P < 0.001$) lower than one, but the intercept was not significantly ($P > 0.05$) different from zero (Fig. 6). This indicated that the tower-top measurement underestimated the $F_s$ by 34.6% compared with the reference 8-level profile system, qualitatively in accordance with previous studies (Iwata et al., 2005; Gu et al., 2012; Bjørkegren et al., 2015). The number of sampling levels and the vertical distributions varied considerably among sites (Table S1). This partly results in the disagreement on whether the tower-top method can safely replace the profile method. We noticed that previous studies that considered the two methods being interchangeable all lack adequate information, such as the sampling period (for one cycle of the whole profile), the percentage of the effective measurement time (with the purging time subtracted) to the average interval (Table S1), the time scale of the dataset, etc. Such obscurities should be clarified in order to increase the reliability of the interchangeableness of two methods.

In order to assess the effect of the two estimates of $F_s$ on NEE, we compared the tower-top based NEE ($F_s + F_{s,EC}$) and the profile-based NEE ($F_s + F_{s,p}$) in Table 2. The deviations of the slopes from one were within 5%, while the intercepts for the leaf-on daytime (0.560 µmol m$^{-2}$ s$^{-1}$) and leaf-on nighttime (−0.864 µmol m$^{-2}$ s$^{-1}$) were highly significantly ($P < 0.001$) different from zero. The large intercepts demonstrated the tower-top method underestimated the daytime $CO_2$ uptake and the nocturnal $CO_2$ release because the NEE was negative in daytime and positive.
at night in the leaf-on season. According to Papale et al. (2006),
using the tower-top method to estimate \( F_s \) lead to an error of
25 g C m\(^{-2}\) yr\(^{-1}\) on the annual NEE; and the underestimations can
be up to the order of 100 g C m\(^{-2}\) yr\(^{-1}\) on the gross primary
productivity and ecosystem respiration. Therefore, it should be
cautious to use the tower-top method to measure \( F_s \) at least at forest sites.
If a profile system is not available in old dataset, one may explore
the possibility to retrieve a “storage correction factor” through a
regression between the storage measured with the profile system
and a series of variables (Papale et al. 2006), models considering
the mean diurnal variation (Hutyra et al. 2006), or just a simple
linear model similar in Fig. 6b. The associated uncertainty may
be less important than the systematic bias of underestimation of
photosynthesis and respiration.

4.3. Uncertainty of CO\(_2\) storage estimates

The uncertainty of \( F_s \) was higher at night than in the day
as expected (Fig. 7). Surprisingly, the peak of uncertainty of \( F_s \)
ocurred in late night, rather than in the transition phase of the
boundary-layer when the magnitude of \( F_s \) peaked. The uncertainty
of \( F_s \) for the 28-min window in the transition periods during
the leaf-on season (around 0.4 µmol m\(^{-2}\) s\(^{-1}\)) was relatively small
(~10%) compared to the magnitude of \( F_s \), but it was large in the
nighttime. During the leaf-on season, the relative uncertainty of
\( F_s \) (the ratio of the SD of \( F_s \) to the magnitude of \( F_s \)) was 10% in the
transition periods, 50% between 8:00–16:00, and about 100% in the
nighttime. Additionally, the uncertainty of \( F_s \) systematically diminished
with the length of the time window (for averaging CO\(_2\) mixing
ratio) increasing (Fig. 8), although its diurnal pattern was similar for
different time windows (Fig. 7). For the leaf-on season, the uncertainties for the 2-min window in the daytime (2.47 µmol m\(^{-2}\) s\(^{-1}\)) and in the nighttime (4.44 µmol m\(^{-2}\) s\(^{-1}\)) were 12.4 and 12.7 times of those for the 28-min in the corresponding time. The large
uncertainty of \( F_s \) for small window might be unacceptable to NEE
calculating.

The uncertainties in \( F_s \) are rarely assessed in the literature.
Heinesch et al. (2007) estimated an upper limit of the uncertainty
due to sampling time was about 0.42 µmol m\(^{-2}\) s\(^{-1}\) (10% of \( F_s \)) for
eight measurements in the 30-min window at the Vielsalm site,
using one-point measurement at the 1-m height. Their estimate of
upper limit of absolute uncertainty was similar to or higher than
the uncertainty in this study (Fig. 8). However, the uncertainty of
\( F_s \) was estimated to be 0.9 µmol m\(^{-2}\) s\(^{-1}\) (300%) by the wavelet
analysis in an open-canopy forest (van Gorsel et al., 2011). The
disparities among these studies may be attributed to three potential
factors: (1) Differences in vertical structure of forest canopy
cause discrepancy in period of oscillation of \( X_c \). The gravity waves

**Table 2**

| Time period | \( N \) | \( R^2 \) | Slope | 95% CI of slope | Intercept | 95% CI of intercept |
|-------------|--------|--------|-------|-----------------|-----------|-------------------|
| The whole year | 12632 | 0.750 | 1.009* | 1.000, 1.018 | 0.020 | −0.049, 0.088 |
| Leaf-on daytime | 3170 | 0.789 | 0.027** | 1.010, 1.043 | 0.560** | 0.357, 0.763 |
| Leaf-on nighttime | 1598 | 0.578 | 1.030 | 0.998, 1.063 | −0.864** | −1.228, −0.502 |
| Leaf-off daytime | 3798 | 0.748 | 1.009 | 0.992, 1.025 | 0.040 | −0.021, 0.102 |
| Leaf-off nighttime | 4666 | 0.657 | 1.047** | 1.025, 1.066 | −0.013 | −0.072, 0.046 |

Fig. 7. Diurnal variations in standard deviations (SD, used as an uncertainty measure) of CO\(_2\) storage flux \( F_s \) calculated from the eight-level CO\(_2\) dry molar fraction for the averaging window size varying from 2 min to 28 min.

Fig. 8. Changes in standard deviations (SD, used as an uncertainty measure) of CO\(_2\) storage flux \( F_s \) calculated from the eight-level CO\(_2\) dry molar fraction with averaging window size for different periods.
in the open canopy lead to large fluctuation of CO$_2$ concentration; and the period is about 3 min (van Gorsel et al., 2011). In contrast, the canopy at the Vielsalm site and the Maaershan site are dense and complex. A wavelet analysis indicated that the CO$_2$ concentration commonly oscillated in 1–2 min at the Maaershan site (Wang, unpublished data). (2) Different sampling protocols are applied. van Gorsel et al. (2011) and the present study used the profile data and with different designs (Table 5), but Heinesch et al. (2007) used the high-frequency data at 1 m level and assumed a similar uncertainty at all levels. (3) Different proxies of uncertainty are adopted. Heinesch et al. (2007) and this study adopted the SD as a measure of uncertainty, while van Gorsel et al. (2011) used the wavelet analysis to quantify the theoretical uncertainty. Nevertheless, the uncertainties caused by the temporal fluctuation should be appreciated compared with the spatial variability in CO$_2$ concentrations (de Araújo et al., 2010; Montagnani et al., 2009).

4.4. Effects of time averaging of CO$_2$ mixing ratio on CO$_2$ storage estimates

As the length of the averaging time window increasing from 2 min to 28 min, the $R^2$ and the slope of the SMA fitted line were both convergent to one (Fig. 9a and b), and the intercept was approaching to zero (Fig. 9c). Because the range of intercept ($-0.454$ µmol m$^{-2}$ s$^{-1}$–$0.162$ µmol m$^{-2}$ s$^{-1}$) was much lower than that of the $F_{30}$ ($-3.022$ µmol m$^{-2}$ s$^{-1}$–$2.034$ µmol m$^{-2}$ s$^{-1}$, Fig. 5), the slope was the dominant contributor of the error. The trend of the slope indicated that the underestimate in $F_3$ increased as time window of $X_c$ became longer. The $F_3$ based on the 2-min time window was 1.4 times of that by the 30-min window for all the time, 1.5 times of that during the leaf-on nighttime, and 1.2 times during the leaf-off nighttime. For the same length of window from different periods, the magnitude of the bias ranked in the order of the leaf-on nighttime > the leaf-off nighttime > the leaf-on daytime > the leaf-off daytime (Fig. 9b). These findings indicated that using the 30-min mean $X_c$ to calculate the $F_3$ could result in a significant negative bias, which were consistent with Finnigan (2006), Yang et al. (2007) and Bjorkegren et al. (2015). In contrast, Ohkubo et al. (2008) reported that the length of time averaging had no effect on the $F_3$ in a tropical rainforest site. We mathematically assessed the two approaches [(1) calculating $F_3$ based on time-derivative of the 30-min mean $X_c$, and (2) averaging all $F_3$ for all short-time windows in 30-min], and obtained identical results (Supplementary material S1). Therefore, using time-averaging of a single profile of CO$_2$ concentration, instead of spatial averaging of instantaneous CO$_2$ concentration, leads to a significant underestimate of $F_3$.

Theoretically, the $F_3$ is the change rate of the instantaneous CO$_2$ concentration ($\Delta X_c/\Delta t$) integrated in the control volume (Finnigan, 2006). The $\Delta X_c/\Delta t$ is nonlinear because of the “U” or “V” shape of the mean diurnal variation in CO$_2$ concentration for a given height (e.g. Jiao et al., 2011). Performing any averaging of CO$_2$ concentration before calculating the $F_3$ removes some high-frequency components of change (including both noise and real change), which will lead to underestimate the $\Delta X_c/\Delta t$ and consequently underestimate the $F_3$. The extent of underestimation will increase with window size of averaging time increasing (Fig. 9; Bjorkegren et al., 2015; Yang et al., 2007). This corresponds to the “transient” (short time scale) nature of $F_3$ in non-steady state conditions (Gu et al., 2012).

The underestimation of time averaging of CO$_2$ mixing ratio for calculating $F_3$ also has implications on the energy storage and energy balance related to the EC method. Leuning et al. (2012) found that for the La Thuile dataset, the slope of the linear regression (forced through the origin) between turbulent heat flux (the sum of sensible and latent heat fluxes) and available energy (the difference between the net radiation and the heat flux through the soil surface) increased by 0.15 from 0.75 for the half-hourly time-step (when half-hourly averages were used for regression) to 0.90 for the daily time-step (when daily averages were used for the regression); this is an estimate of the energy storage in canopy (biomass and air). However, direct estimate of energy storage in canopy for the half-hourly time-step is on average only 7% even for 15 tall-canopy (height > 8m) ecosystems (Wilson et al., 2002). We speculated that the half-hour energy storage in the canopy calculated from the half-hour mean temperature and moisture might be substantially underestimated compared with the real values in forest ecosystems.

We further examined the biases in calculated NEEs by taking time averaging for $X_c$ for calculating $F_3$ (Table 3). We found that the magnitude of NEE was overall underestimated by more than 5% with the 30-min-window-based $F_3$. The rate of CO$_2$ uptake was significantly underestimated during the leaf-on daytime, while the rate of CO$_2$ release was underestimated during the nighttime. We speculate that the widely observed underestimation of the
nocturnal ecosystem respiration (Lavigne et al., 1997; Speckman et al., 2015) may be reduced if a short time-window for averaging $\chi_c$ was used for calculating $F_s$, because of the large positive intercept (1.058 $\mu$mol m$^{-2}$ s$^{-1}$) in the leaf-on season and large slope (1.140) in the leaf-off season (Table 3). However, the long-term NEE might be less affected than the gross primary production and ecosystem respiration after flux partitioning (Papale et al., 2006) due to partial offsets of the underestimations of both CO$_2$ uptake and release.

Performing the time-averaging of $\chi_c$ measured by a single profile can reduce the random error in $F_s$, but may increase the systematic underestimation of its magnitude. So the random error and systematic error in $F_s$ were irreconcilable for a single profile system. The response time of 2 min for the single profile system in present study is one of the fastest systems in the literature (from 60 s to 1800 s, Table S1). The systematic bias may be minimized if a single cycle measurement is used to calculate the $F_s$ despite of its potential random error. Adding a buffer volume before IRGA may be a possible way to reduce the random error in $\chi_c$ due to the temporal oscillation (Marcolla et al., 2014). However, using a buffer volume is also a low-pass filtering of the CO$_2$ concentration, which is similar to taking a time averaging. Hence the buffer volume should be carefully balanced between noise-filter and high frequency loss, ideally measuring the spatial averaging instantaneous $\chi_c$. A fast response planar-averaging profile is a possible way to solve this problem. For example, simultaneous sampling from multiple points (“line sampling”) at the same height level can reduce the uncertainty due to spatial (horizontal) variation in $\chi_c$ (Marcolla et al., 2014), but this doesn’t seem practical in most cases. The novel design of the profile system, e.g., AP200 (Campbell Scientific Inc, USA), uses a buffer volume with a residence time of 2 min and completes a cycle of eight-level sampling within 2 min, which may effectively decrease the stochastic variation due to gust (Campbell Scitific, 2010). Combination of a fast-response line or plane sampling strategy and an appropriate buffer volume to mixing the air may be the direction of new generation of CO$_2$ profile for $F_s$ measurement. In theory, the $F_s$ measurement doesn’t directly require a profile of CO$_2$ mixing ratio at all. Instead, a profile is used to better represent the spatial mean of CO$_2$ mixing ratio. Therefore, if $F_s$ is the only concern, it is recommended sampling all levels simultaneously and measuring the mean concentration after mixing in a small buffer volume. Such design can measure the nearly instantaneous CO$_2$ mixing ratio of the whole profile, but has no information on the vertical gradient of CO$_2$ mixing ratio. In this case, the vertical configuration of all levels is forced to distribute with equal distance in order to better represent the whole control volume, or at least the vertical direction (c.f. Noormets et al., 2007). And the flow rate must be identical for all levels, which can be controlled with the same length of tubing as the AP100 system.

Theoretically, sampling protocols for $F_s$ measurements should consider the frequency of coherent structure or the integral time scale of scalar concentration. Coherent structure is well-organized ramp-like motion for scalar time series (Gao and Li, 1993). The typical duration of coherent structure above and within forest canopy is between 14–116 s by the wavelet analysis (Cava et al., 2004; Dias Júnior et al., 2013; Gao and Li, 1993; Lu and Fitzjarrald, 1994; Steiner et al., 2011; Thomas and Foken, 2005). However, some studies detected that the duration of coherent structure increased from unstable to stable conditions (Steiner et al., 2011) or varied within a day (Dias Júnior et al., 2013). Other motions such as canopy or gravity waves might significantly contribute to the spectral properties of the scalar time series particularly under stable conditions (Cava et al., 2004; Lee and Barr, 1998; van Gorsel et al., 2011), although they are not as frequent as does the coherent structure. This makes that the frequency of coherent structure can only be used as a rough reference for sampling interval of one cycle of all levels of a profile. According to Finnigan (2006), the integral time scale was an important parameter to determine the bias in $F_s$. The typical integral time scale of CO$_2$ density at our site was about 100 s in both leaf-on and leaf-off seasons, thus a 2-min time window might be a reasonable choice to avoid noise in CO$_2$ concentrations caused by the wave-like motions.

Another sampling problem is the decoupling of canopy/ subcanopy layer with the ambient air in clear and calm nights, when the above- and sub-canopy air layers relation can be significantly weakened. Decoupling is commonly characterized with the vertical gradient of the bulk Richardson number (a measure of stability) (Van Gorsel et al., 2009) or with vertical shear of mean wind direction (Alekeychik et al., 2013). The decoupling suppressed the vertical mixing and exchange of mass and energy (Burns et al., 2011; van Gorsel et al., 2011), thus might changed the gradient and time scale of scalar concentration measurements for the $F_s$ calculation. However, the occurrence and depth of decoupling sublayers are highly dependent on the canopy structure and atmospheric stability (Burns et al., 2011; Wang et al., 2015). For forests with sparse understory and clear trunk space, the decoupling sublayer was often constrained within the subcanopy (Heinesch et al., 2007; van Gorsel et al., 2011) and might increase its depth up to the whole canopy at very stable conditions (Alekeychik et al., 2013). For forests with dense canopy, the decoupling regime might be more complex and even extend to the daytime due to the radiative heating/cooling and thermal isolation of the canopy (Froelich and Schmid, 2006; Pyper et al., 2007; Wang et al., 2015). At our site, the above canopy decoupling (wind direction shear between height levels beyond a threshold of 20°) had a probability of 22% in the daytime and 39% in the nighttime, and the below-canopy misalignment occurred with a frequency of more than 70% in both daytime and nocturnal occasions. So it is difficult to design a sampling system according to the variations in depth and timing of decoupling. However, the topography and canopy structure might have general implications for the regime of decoupling (van Gorsel et al., 2011; Wang et al., 2015), which might be helpful for design profile systems for $F_s$ estimates.

5. Conclusions

The CO$_2$ storage in a tall-canopy forest ($F_s$) makes significant contributions to NEE estimation in the early morning and nighttime. Vertically, the $\chi_c$ and the magnitudes of its time derivative

| Time period          | $N$  | $R^2$ | Slope  | 95% CI of slope | Intercept | 95% CI of intercept |
|----------------------|------|-------|--------|-----------------|-----------|--------------------|
| The whole year       | 18970| 0.855 | 1.071  | 1.065, 1.077    | 0.121     | 0.079, 0.161       |
| Leaf-on daytime      | 4875 | 0.854 | 1.057  | 1.046, 1.068    | −0.209    | −0.339, −0.079     |
| Leaf-off daytime     | 2763 | 0.815 | 1.051  | 1.034, 1.068    | 1.058     | 0.895, 1.221       |
| Leaf-off nighttime   | 5228 | 0.798 | 1.057  | 1.045, 1.070    | −0.103    | −0.132, −0.074     |
| Leaf-on nighttime    | 6104 | 0.700 | 1.140  | 1.125, 1.156    | 0.094     | 0.065, 0.123       |

Table 3 Results of standardized major axis (SMA) line-fitting for the estimates of net ecosystem exchange of CO$_2$ (NEE) calculated from the 2 min- and 30 min-window-based CO$_2$ molar mixing ratio from the eight-level profile system, respectively. The NEE is calculated as the sum of eddy flux ($F_e$) and change in storage term ($F_s$). The model form is $F_e + F_{a0} + F_{a0} + F_{e0} = a + bF_{a0} + F_{e0}$. All line-fittings are highly significant ($P < 0.001$); and all slopes and intercepts are highly significant ($P < 0.001$) different from unit and zero, respectively. Sample size ($N$), determination coefficient ($R^2$), and 95% confidence interval (CI) are given.
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