Analysis of CO₂ spatiotemporal variations in China using tower data and a weather-biosphere-online-coupled model, WRF-VPRM

Xinyi Dong¹,², Man Yue¹,², Yujun Jiang³,⁴, Xiaoming Hu⁵, Qianli Ma⁴, Jingjiao Pu³, and Guangqiang Zhou⁶

¹School of Atmospheric Science, Nanjing University, Nanjing, 210023, China
²Joint International Research Laboratory of Atmospheric and Earth System Sciences & Institute for Climate and Global Change Research, Nanjing University, Nanjing, 210023, China
³Zhejiang Meteorological Science Institute, Hangzhou 310008, China
⁴Zhejiang Lin’an Atmospheric Background National Observation and Research Station, Hangzhou 311307, China
⁵Center for Analysis and Prediction of Storms, University of Oklahoma, Norman, Oklahoma, 73072, USA
⁶Shanghai Key Laboratory of Health and Meteorology, Shanghai Meteorological Service, Shanghai, 200135, China

Correspondence to: Yujun Jiang (yjjiang@pku.org.cn) and Xiao-Ming Hu (xhu@ou.edu)

Abstract. Dynamics of atmospheric CO₂ has received considerable attention in the literature, yet significant uncertainties remain within the estimates of contribution from terrestrial flux and the influence of atmospheric mixing. In this study, we apply the Weather Research and Forecasting model coupled with Vegetation Photosynthesis and Respiration Model (WRF-VPRM) in China to characterize CO₂ the dynamics with tower data collected at a background site Lin’an (30.30°N, 119.75°E) of CO₂ in the atmosphere. The online coupled weather-biosphere WRF-VPRM simulations are able to simulate biosphere processes (photosynthetic uptake and ecosystem respiration) and meteorology in one coordinate system. Simulations are conducted for three years (2016-2018) with fine grid resolution (20 km) to detail the spatiotemporal variations of CO₂ fluxes and concentrations. This is the first attempt to apply the weather-biosphere model for a multi-year simulation. We apply WRF-VPRM for a multi-year simulation (2016-2018) with integrated data from a satellite product, flask samplings, and tower measurements to diagnose the dynamic spatiotemporal variations of CO₂ in fluxes and concentrations in China. We find that the spatial distribution of CO₂ is dominated by anthropogenic emissions, while its seasonality (with maximum concentrations in April 15 ppmv higher than minimum minima in August) is dominated by terrestrial flux and background CO₂. Observations and simulations reveal a consistent increasing trend in column-averaged CO₂ (XCO₂) of 2.46 ppmv (0.6%/yr) resulting from anthropogenic emission growth and biosphere uptake. WRF-VPRM successfully reproduces ground-based measurements of surface CO₂ concentration with mean bias of -0.79 ppmv (-0.20%) and satellite derived XCO₂ with mean bias of 0.76 ppmv (0.19%). The model-simulated seasonality is also consistent with observations, with correlation coefficients of 0.90 and 0.89 for ground-based measurements and Orbiting Carbon Observatory-2 (OCO-2) satellite data, respectively. However, evaluation against Lin’an tower data reveals uncertainty within the model for simulating the intensity and diurnal variation of terrestrial flux, which contributes to overestimation by ~5.35 ppmv (1.26%).
Lin'an tower observations also reveal from a background site Lin’an (30.30˚N, 119.75˚E) revealed a strong correlation (-0.8598) between vertical CO$_2$ and temperature gradients, suggesting a significant influence of boundary layer thermal structure on the accumulation and depletion of atmospheric CO$_2$.

1 Introduction

Climate research requires accurate characterization of atmospheric CO$_2$, which is closely affected by the both atmospheric transport and terrestrial sources and sinks (Bauska et al., 2015; Keenan et al., 2016). Our current knowledge largely comes from interpreting ground- or space-based measurements and model simulations. While observation is limited by spatial and temporal coverages, modelling approaches also suffer from various uncertainties (Shi et al., 2018). Modelling assessment of CO$_2$ is usually conducted through two methods: first, process- or data-driven biosphere models in which terrestrial fluxes are diagnostically calculated with theoretical functions (Tian et al., 2015) or determined through semi-empirical relationships derived from ground measurements and/or satellite products with machine learning techniques (Papale and Valentini, 2003); second, inverse modelling in which prior flux estimates applied in atmospheric transport models are calibrated by observational data and/or satellite products to determine posterior flux (Peylin et al., 2002; Kountouris et al., 2018). Process-driven biosphere models have difficulties capturing spatial and temporal variabilities at fine resolution because parameters calibrated from a limited number of site observations are applied across a variety of land covers (Todd-Brown et al., 2013). Atmospheric inverse modelling is predominantly affected by the presumed prior flux, and different assimilation techniques can give different and even conflicting results (Peylin et al., 2013). These fundamental features highlight the limits of these approaches for accurately modelling carbon dynamics.

Researchers have attempted to reconcile differences between “bottom-up” biosphere models and “top-down” atmospheric inverse models, and recent studies have demonstrated increasing levels of agreement owing to improved understanding of both approaches, such as better parameterization of biosphere processes (Dayalu et al., 2018), more accurately constrained estimates of prior flux (Crowell et al., 2018; Feng et al., 2019), and advanced measurement/satellite instruments that provide high quality data for assimilation (Gaubert et al., 2019); however, critical model disagreements still persist (Kondo et al., 2020). To bridge the gap between terrestrial flux and atmospheric mixing, a type of weather-biosphere coupled model (Ahmadov et al., 2007; Mahadevan et al., 2008) has been developed to simulate biosphere processes and meteorology conditions in one coordinate system, allowing their interactions to be properly addressed. A few case studies (Ahmadov et al., 2009; Kretschmer et al., 2012; Park et al., 2018) have demonstrated the potential advantages of coupled weather-biosphere models over pure biosphere/inverse models for short term (a few weeks) simulations, but whether the coupled model is able to reproduce the spatial distributions and temporal variations and subsequently estimate carbon fluxes at regional scales with high confidence remains a crucial issue to be addressed.
Understanding the spatiotemporal characteristics of atmospheric CO₂ is a key priority in China because of the central role it plays in regulating the climate and environment. In recent years, tremendous efforts have been made in China to control anthropogenic emissions from fossil fuel combustion for both air quality and climate mitigation purposes (Zheng et al., 2018). While the sources and sinks of air pollutants have been thoroughly examined and well documented (Huang et al., 2020), the dynamics of CO₂ at regional to national scales remain poorly understood due to lack of long-term observations and limited modelling studies (Han et al., 2020). Li et al. (2020) applied a weather-biosphere model with tower observations to analyse CO₂ fluxes and concentrations over mixed forest and rice paddy in northeast China, but the one-year simulation limits the attempt to investigate interannual CO₂ variation which is subject to substantial change (Fu et al., 2019b). Wang et al. (2019) applied satellite products and in-situ observations with inverse modelling to derive posterior carbon fluxes and reported 100% uncertainty for constraining global terrestrial flux. Fu et al. (2020) applied GEOS-Chem simulation with offline Carbon Tracker (Peters et al., 2007) as input to estimate impacts of terrestrial flux and anthropogenic emissions on the annual variation of CO₂ concentrations, but regional-scale assessment was limited by coarse grid resolution (2°×2.5°). Machine-learning technique has also been employed to upscale site observations to regional-scale (Yao et al., 2018; Zhu et al., 2014), but the estimations of carbon budget and dynamics retain large uncertainty due to the diversity of biomass among sites and suffer from coarse grid resolution. These pilot studies have shed light on improving the understanding of spatiotemporal characteristics of CO₂ in China with modelling or observational methods, but an integrated investigation with both modelling and observations at fine-scale is urgently needed to expand diagnostic understanding of localized and regional transport, flux, and concentration of CO₂ to inform emission management and climate adaption policies (Fu et al., 2019a; Niu et al., 2017; Wang et al., 2019).

In this study we apply the Weather Research and Forecasting model coupled with the Vegetation Photosynthesis and Respiration Model (WRF-VPRM) (Hu et al., 2020; Mahadevan et al., 2008) to simulate and characterize the spatiotemporal variation of atmospheric CO₂ in China from 2016-2018, and also to validate this weather-biosphere model with recent advanced satellite and tower observations. WRF-VPRM has been applied in a few case studies over the United States (Hu et al., 2020), Europe (Kretschmer et al., 2012), northeast China (Li et al., 2020), and South Korea (Park et al., 2020); this study is the first attempt to apply and evaluate it for a multi-year simulation at fine scale (20 km) over China. We first describe the modelling methods and data employed followed by model validation against observations from multiple datasets, and then present the spatiotemporal variations and estimates of contributions from anthropogenic emissions, terrestrial flux, and background concentrations. Finally, we probe into investigate tower data and reveal the boundary layer thermal structure impacts on atmospheric CO₂ accumulation and depletion.

2 Method

The WRF-VPRM simulation in this study is configured with 48 vertical layers and 20 km grid resolution. Initial and boundary conditions are We conduct nested WRF(Version 3.9.1.1)-VPRM simulations over China (domain shown in Fig.1(a)) and Yangtze River Delta (YRD) region (domain shown in Fig.1(d)) at 20 km and 4 km grid resolution, respectively. Both
simulations were configured with 47 vertical layers with model tops at 10hPa. Model configuration in this study followed the work by Hu et al. (2020) and Li et al. (2020). We applied the YSU planetary boundary layer (PBL) scheme (Hong et al., 2006), Morrison microphysics (Morrison et al., 2009), Duhia short-wave radiation (Dudhia, 1989), RRTM long-wave radiation (Mlawer et al., 1997), Grell-3 cumulus scheme (Grell and Devenyi, 2002), and Noah land-surface scheme (Chen and Dudhia, 2001), with more details summarized in Table S1. In general, the 4km-grid simulation showed no significant difference as compared to the 20km-grid simulation (demonstrated in Figure S1 and Figure S2), thus the 20km-grid simulation was used to characterize the spatiotemporal distributions of CO$_2$ over China, and the 4km-grid simulation was only used to compare with tower data collected at a background site in YRD. Discussions in the next section will mostly refer to the 20km-grid simulation unless otherwise specified. Initial and lateral boundary conditions for the 20km-grid simulations were derived from the mole fraction product of CarbonTracker (Peters et al., 2007) with 3°×2° resolution. The latest update of column average CO$_2$ (XCO$_2$) concentration assimilation product from CarbonTracker (CT2019) with 1°×1° resolution (Jacobson, 2020) was also employed to compare with the WRF-VPRM simulation. The anthropogenic emission inventory is from the Open-source Data Inventory for Anthropogenic CO$_2$ (ODIAC) with 0.1°×0.1° resolution (Oda et al., 2018) shown in Fig.1(a); ocean flux is from climatology estimation (Takahashi et al., 2009); and vegetation fractions and enhanced vegetation index (EVI, shown in Fig.1(b)) are from MODIS (Huete et al., 2002). CO$_2$ from initial and boundary conditions, anthropogenic emission, and terrestrial biogenic flux are tagged as BCG, ANT, and BIO, respectively, to allow the contributions from each process to be identified and quantified through one simulation.

WRF-VPRM calculates ecosystem respiration (ER) and gross ecosystem exchange (GEE) with the following functions as:

\[
ER = \alpha \times T + \beta \tag{1}
\]

\[
GEE = -\lambda \times T_{\text{scale}} \times W_{\text{scale}} \times P_{\text{scale}} \times (1 + PAR/PAR_0)^{-1} \times EVI \times PAR \tag{2}
\]

where $T$ is the air temperature at 2m above the surface ($T_2$); $\alpha, \beta, \lambda$ are vegetation type-dependent parameters; $PAR_0$ is the vegetation type-dependent half-saturation value of photosynthetically active radiation (PAR); and $T_{\text{scale}}, W_{\text{scale}}, P_{\text{scale}}$ are scaling factors for temperature, water stress, and phenology, respectively. In this study we take the atmosphere as a reference, thus GEE has a negative sign and ER has a positive sign. The current version of WRF-VPRM is parameterized ($\alpha, \beta, \lambda$) for 7 vegetation types (Fig.1(c)): crops, mixed forest, evergreen forest, deciduous forest, shrub, savanna, and grass. For each modelling grid, ER and GEE are calculated as the weighted averages of each vegetation type based on their fractional abundance. Recent studies (Hu et al., 2020; Li et al., 2020) have investigated the uncertainty associated with this parameterization through sensitivity simulations and suggested the crops can be further divided into subcategories based on eddy-covariance (EC) tower measurement to improve the model. In this study we applied the default parameterization (values presented in Table S2) which has been demonstrated to successfully reproduce the terrestrial flux over northeast China (Li et al., 2020). In contrast, CT2019 applies a process based biosphere model, the Carnegie-Ames Stanford Approach (CASA(Zhou et al., 2020)), driven by year-specific weather and satellite data to simulate terrestrial fluxes (Peters et al., 2007). CASA also estimates photosynthetic uptake based on solar radiation and plant phenology, and estimates respiration as a
function of T2. CASA directly simulates monthly means of Net Primary Production (NPP) and heterotrophic respiration (R_h). NPP is the difference between photosynthetic uptake (equivalent to GEE) and autotrophic respiration (R_a). The summary of R_h and R_a is equivalent to ER. Thus, WRF-VPRM and CASA are essentially very similar in terms of considering methodology impact; however, it should be noted that to resolve CASA simulated NPP into GEE and R_a, CT2019 applies the assumption that GEE is twice that of NPP, which implies that for the same plants the photosynthetic carbon uptake is double the magnitude of autotrophic respiration (but of opposite sign). This assumption is applicable at monthly scale but may contribute to have difficulty reproducing the rapid changes at hourly and daily scales due to impact from weather systems, which will be demonstrated with more details in Section 3.2.

**Measurements**

Hourly measurements of CO2 concentrations were collected at the Lin’an Regional Atmospheric Background Station (30.30°N, 119.75°E, surroundings shown in Fig. 1(a)) with Picarro G1301 and G1302 trace gas analysers mounted on an observation tower at 21 and 55 meters, respectively, above ground level (AGL) and analysed online (data analysis lab shown in Fig. 1(b)). The station is located in the remote area of Hangzhou 138.6 meters above sea level in the middle of a hilly area covered by mixed forest. The hourly Lin’an station tower measurements collected between 2016-2018 provide a representative sampling of the The observation tower is 60km to the west of downtown center of Hangzhou and 195km to the southwest of Shanghai. Fig. 2(c) and (d) presents the wind rose map at Lin’an derived from hourly observations of 10m and 55m wind respectively, which clearly shows the northeast and southwest as prevailing wind directions. The station can properly represent the background atmospheric environment in YRD as demonstrated in previous studies (Deng et al., 2018; Pu et al., 2020). The tower data provides a representative sampling of CO2 gradients resulting from exchange between atmosphere mixing and terrestrial flux.

**Flask samplings of CO2**

Atmospheric samples near the surface concentrations with were collected at monthly intervals are collected and analysed for CO2 through the National Oceanic and Atmospheric Administration’s (NOAA’s) Earth System Research Laboratory (ESRL) at four sites (locations shown in Fig. 1(a)) within our modelling study domain, including Dongsha Island (DSI, 20.69°N, 116.73°E), Lulin (LLN, 23.47°N, 120.87°E), Ulaan Uul (UUM, 44.45°N, 111.09°E), and Mt. Waliguan (WLG, 36.29°N, 100.89°E). The Orbiting Carbon Observatory-2 (OCO-2) satellite product (Kiel et al., 2019) with daily intervals was employed to validate simulation of column averaged CO2 (XCO2) concentrations. A total of 204,940 OCO-2 version 9 swath data covering the simulation period was used in this study. Daily ground-based Fourier transform spectrometer (FTS) measured XCO2 at Hefei site (31.90°N, 117.17°E) also collected through the Total Carbon Column Observing Network (TCCON) for year 2016 (Wang et al., 2017). The TCCON-Hefei site was located in the northwestern rural area of Hefei city and measurements were conducted from September 2015 to December 2016 (Liu, 2018). WRF has been evaluated extensively and consistently performs well for reproducing the meteorology fields and the transport of atmospheric tracers, in China (Gao et al., 2015; Tang et al., 2016; Wang et al., 2017; Yang et al., 2019), so this study will only present the simulation performance for CO2, which hasn’t been thoroughly discussed in the literature.
3 Result and Discussion

3.1 Model evaluation

We first evaluate the capability of WRF-VPRM to reproduce concentrations of surface CO₂ and XCO₂, and we find fairly good model performance through the comparison with satellite and ground-based observations. The WRF-VPRM simulated surface layer (mid-level height AGL is 12m) CO₂ and XCO₂ averages between 2016-2018 are demonstrated in Fig.2(a) and (b) respectively. High concentrations are found over industrial areas such as the North China Plain (NCP), Pearl River Delta (PRD), and Yangtze River Delta (YRD), where the surface CO₂ and XCO₂ are above 440 ppmv and 408 ppmv, respectively; the domain averages are 411 ppmv and 406 ppmv, respectively. While most climate models assume evenly distributed CO₂ (Fung et al., 1983; Kiehl and Ramanathan, 1983), our data demonstrates a prominent gradient between industrial and remote areas (e.g., Tibet Plateau, Mongolia), especially for surface CO₂, which could be an important source of uncertainty for estimating the long-wave radiation effect (Xie et al., 2018). Spatial patterns of CO₂ and XCO₂ are in close agreement with ODIAC, indicating the dominant impact of anthropogenic emission in determining the CO₂ distribution. WRF-VPRM simulated CO₂ is generally consistent with CT2019 (Fig.2(d)), but CT2019 estimates lower CO₂ surface CO₂ (mid-level height AGL is 25m) over the coastal industrial areas YRD and PRD because the ocean module used in CT2019 provides stronger air-sea exchange than the ocean flux by Takahashi et al. (2009) used in WRF-VPRM. The two models showed better agreement for XCO₂ (Fig.2 (b) and (e)), but also differed by ~1 ppmv over Taklamakan Desert and along the eastern side of the Tibet Plateau. The OCO-2 swath data integrated into the corresponding horizontal grids of WRF-VPRM and CT2019 respectively, to validate XCO₂. Biases of WRF-VPRM and CT2019 both fall into the range of ±3 ppmv as shown in Fig.2(d) and (f), respectively, but WRF-VPRM apparently provides more details of spatial gradient. WRF-VPRM showed well-mixed underestimations and overestimations along neighbouring satellite tracks, while CT2019 tended to overestimate (underestimate) over Tibet Plateau (Taklamakan Desert) where WRF-VPRM slightly smaller biases. Fig.4(a) and (b) present the raw data pairs between models and OCO-2 with daily interval for WRF-VPRM and CT2019 respectively. In general, the WRF-VPRM model reproduced OCO-2 well, with mean bias (MB) of 0.76 ppmv and normalized mean bias (NMB) of 0.19% (Fig.3(a)); CT2019 showed MB of 0.54 ppmv and NMB of 0.17% (Fig.3(b)), suggesting an overall acceptable performance of the weather-biosphere model to reproduce the spatial distribution pattern of XCO₂ in China.

We further analyse WRF-VPRM validation against OCO-2 for the seven vegetation types in each season and find no prominent difference (evaluation statistics summarized in Table 1). Regarding vegetation type, the model showed the largest normalized mean bias (NMB) MB over deciduous forest of -0.25% and 0.12 ppmv in summer and winter, respectively, both over deciduous forest which only cover a very small portion in northeast China (see dominant vegetation types in Fig.1(c)). The three most abundant coverage vegetation types in China are grass, crops, and mixed forest. XCO₂ simulated by WRF-VPRM over grass areas is slightly overestimated by 0.08~0.16 ppmv throughout the year, and the NMB over mixed forest is -0.11% to 0.15%. This indicates a good performance of the model over the
vast majority of areas of China. Performance over crops generally showed larger discrepancy than other vegetation types, with NMBMB ranging from 0.16% to 0.66 ppmv in summer to 0.29% to 1.19 ppmv in winter, suggesting the model tends to slightly overestimate column concentration of CO₂ over cropland. Li et al. (2020) reported that WRF-VPRM underestimated biosphere carbon over rice paddy sites (by ~3%) in northeast China and suggested the parameterization of α, β, λ as the most important cause. Cropland differs significantly across China with various types of species such as rice, wheat, and corn, for which literature reports substantially different rates of ecosystem respiration and photolysis uptake (Gao et al., 2018; Yang et al., 2016; Zhu et al., 2020). Thus, applying one set of parameters to represent all crops may be responsible for the lingering uncertainty of simulated XCO₂. In terms of seasonal difference, WRF-VPRM performs best showed slightly smaller bias in summer (NMB=0.12%) and worse larger bias in winter (NMB=0.23%), and the correlation coefficients were all ~0.8, consistent with application over the U.S. (Hu et al., 2020) which also reported slightly better performance in summer than other seasons, indicating good agreement with the OCO-2 satellite product.

Fig. 4 also presents the overall simulation bias against ground-based observations employed in this study at the their raw temporal intervals (daily monthly for OCO-2, daily for TCCON data at Hefei ESRL sites, hourly for tower data at Lin’an, and monthly daily for data TCCON at Hefei). At the ESRL sites, surface (Fig. 4(c)), surface CO₂ concentrations were simulated well with minor overestimation by 0.69 ppmv (0.17%) at the ESRL sites (Fig. 3(f)). However, evaluation Evaluation at the Lin’an station shows significant overestimations for CO₂ by 5.34 ppmv (1.25%) and 5.41 ppmv (1.27%) at 21 m (Fig. 3(d)) and 55 m (Fig. 3(e)) AGL, respectively. The was performed with the 4 km-grid simulation. The mid-level heights of WRF-VPRM’s first, second, and third layers were 12.3 m, 36.9 m, and 61.6 m, respectively, and simulations were linearly interpolated to 21 m and 55 m to compare with the tower data. The evaluation at 21 m AGL (Fig. 4(d)) shows slight overestimation by 0.02 ppmv, but the evaluation at 55 m height (Fig. 4(e)) shows relatively large overestimations by 1.06 ppmv. The discrepancy is largely attributable due to the combined effect of vertical allocation of anthropogenic emission within the model as recently recognized (Brunner et al., 2019). Biosphere and parameterization of VPRM. Tracer transport models (such as WRF-VPRM and CASA) and inverse modelling methods allocate anthropogenic CO₂ emission into the near surface layer due to lack of injection height information, which will likely subsequently lead to systematic overestimation of surface CO₂ concentration in industrial areas, though a regional scale (750×650km) modelling study around the city of Berlin (Brunner et al., 2019) reported that distributing anthropogenic emission into the surface layer overestimated near-surface CO₂ concentration by 14% in summer and 43% in winter as compared with considering the vertical profiles of local anthropogenic sources. Lin’an observation tower is located at a densely vegetated area. Validation against OCO-2 suggested that WRF-VPRM did not show significantly different performance over different vegetation types as shown in Table 1. As compared to the ESRL background sites which were located in more remote areas with little anthropogenic emission (Fig. 1(a)), Lin’an was more frequently affected by regional anthropogenic emissions which were transported from Shanghai and Hangzhou due to the prevailing northeast wind (Pu et al., 2014), indicating that the emission allocation discrepancy may induce more prominent error at Lin’an. In fact, the 20 km-grid WRF-VPRM simulation bias at Lin’an were 5.34 and 5.41 ppmv at 21 m and at 55 m
respectively (Figure S2), significantly larger than the bias at ESRL sites. In addition, both the 20km-grid and 4km-grid simulations showed relatively larger bias at 55 m than 21 m due to smaller topography roughness and higher wind speed which increases with height according to observations (Figure S3). CT2019 also substantially underestimated anthropogenic emission at Lin’an, but the first, second, and third layers’ mid-level heights are 25 m, 103 m, and 247 m, respectively, so we did not compare it directly.

present the direct comparison with the tower data, but analysed the simulated diurnal variation as will be discussed in Section 3.3. Fig. 3(c) and (d) reveal that observed average CO₂ concentrations at Lin’an (428 ppmv) are substantially higher than those at ESRL sites (407–410 ppmv). The evaluation at Lin’an station also infers the prominent high CO₂ level in YRD due to the intensive regional anthropogenic emission as compared with ESRL sites at remote locations. Pu et al. (Pu et al., 2014) analysed the back trajectories for hourly measurements collected at Lin’an station between 2009–2011 and demonstrated that it was frequently affected by prevailing northeast winds carrying polluted airmasses from upwind cities including Hangzhou, Shanghai, and northeast parts of Jiangsu where manufacturing factories were densely located. Simulated XCO₂ is also compared with those WRF-VPRM and CT2019 were well consistent with the TCCON Hefei site observations, and a very good agreement is found as shown in Fig. 4(f), with MB −0.79 ppmv and NMB of −0.2%. In general, recent 0.78 ppmv respectively, and NMB by −0.20% and −0.19% respectively. The 4km-grid simulation showed very similar result to the 20-grid simulation for CO₂ (Figure S1 and Figure S2). Recent atmospheric inverse modelling studies (Fu et al., 2019; Wang et al., 2019; Xie et al., 2018) reported the simulation bias of XCO₂ at 0.5–2 ppmv with posterior flux inputs. The WRF-VPRM model applied in this study has demonstrated good agreement with the observations as a process-based model through our evaluation.

### 3.2 CO₂ seasonal variation and trend in China

We next analyse the seasonality of CO₂ and XCO₂ and find that the terrestrial flux played a more influential role than anthropogenic emission. WRF-VPRM successfully reproduced seasonal variations of CO₂ at ESRL sites, with a correlation coefficient of 0.90 (Fig. 4(a)). The WRF-VPRM 4km-grid simulation showed a correlation of 0.82 with the Lin’an tower is only 0.67 (Fig. 4(c)). We will probe into bias at Lin’an in the next section (observation of 2011 and demonstrated). Both the model and measurements showed prominent seasonal cycles for surface CO₂ concentrations, with maxima in April (413–419 ppmv) and minimums in August (399–404 ppmv) as shown in Fig. 4(b). The model suggested that the anthropogenic CO₂ contribution was 2.6 ppmv in both months, while the biogenic contributions were 3.1 ppmv and −1.2 ppmv in April and August, respectively (Fig. 4(d)). Anthropogenic emission (Fig. 4(b)) showed a flat curve with relatively higher values in December due to fuel combustion for heating (Zheng et al., 2018). EVI meanwhile showed maximums in July and August (Fig. 4(f)). During summer, photosynthetic uptake almost completely offset anthropogenic emission, causing the minimum CO₂ concentration observed in August, while the higher anthropogenic emission in December and respiration flux in April lead to the two corresponding peaks. The anthropogenic XCO₂ contributions from were 0.5 and 0.6 ppmv in April and August, respectively, and the...
biogenic contributions were 0.8 ppmv and -1.5 ppmv, respectively, suggesting that the seasonality of XCO2 was also primarily dominated by terrestrial flux. Furthermore, the seasonality at high-latitude ESRL sites (UUM and WLG) was stronger than at Lin’an and low-latitude sites (DSI and LLN) because of the larger temperature and photosynthetically active radiation (PAR) gradients. Annual average anthropogenic and biogenic XCO2 contributions were 7.1 ppmv and -1.9 ppmv, respectively, indicating that biosphere uptake is an important carbon sink offsetting 27% of anthropogenic emission and slowing the growth of atmospheric CO2.

XCO2 showed similar seasonality, with minimums in August and maximums in April and December (Fig.45(b)). Both WRF-VPRM and CT2019 showed good agreement with TCCON Hefei observations with correlations of 0.89 and 0.88, respectively (Fig.45(e)). However, we note that WRF-VPRM simulated drastic changes (e.g., the grey shaded period in Fig.45(e)) that were not reproduced by CT2019. Figure 56 shows the daily concentrations of XCO2 overlaid with horizontal wind speed at 10m AGL from WRF-VPRM and CT2019 and highlights large discrepancies over Hefei, (Figure S4 shows the same comparison but using WRF-VPRM 4km-grid simulation data). Between April 1st and 3rd 2016, an 850 hPa trough associated with a surface cold front moved southeastward from Mongolia to the North China Plain (NCP) (weather maps shown in Fig.56(g)-(i)). At the leading edge of the front, a convergence zone associated with a low pressure center formed, which led to significant cloud formation and subsequently reduced short-wave radiation. As a result, photosynthetic carbon uptake was reduced, leading to enhancement of atmospheric CO2. Meanwhile, the cold front transported anthropogenic CO2 from NCP to YRD, and the convergence zone along YRD ahead of the front facilitated the accumulation of air pollutants and CO2 from anthropogenic emissions. With its coarse spatiotemporal resolution, CT2019 had difficulty reproducing such regional weather systems that can lead to rapid and localized changes in CO2 concentration and terrestrial flux, indicating the importance of fine resolution modelling to better represent the small spatial scale and rapid temporal scale variations of CO2 (Agusti-Panareda et al., 2019).

We also find a notable increasing trend for the 3-year study period. Observed CO2 annual enhancement was 0.56%/yr at the ESRL sites and 2.3 ppmv/yr (0.62%/yr) at Lin’an. The observed average CO2 concentrations at Lin’an (428 ppmv) were substantially higher than those at ESRL sites (407-410 ppmv). The prominent higher levels of CO2 and slightly higher absolute growth rate at Lin’an can be attributed to the influence of the transport regional anthropogenic emission, which is growing at rate of 0.82%/yr as suggested by ODIAC. Domain-wide XCO2 was also found to increase by 0.52%/yr (2.3 ppmv/yr) as suggested by OCO-2 and 0.61%/yr (2.5 ppmv/yr) as suggested by the simulation. WRF-VPRM reproduced the trends in good agreement with ground and satellite observations. Model simulated budgets suggested that the increasing trends for anthropogenic, biogenic, and background XCO2 were 0.8%/yr, -9.17%/yr, and 0.59%/yr, respectively; the trends for anthropogenic, biogenic, and background CO2 were 4.95%/yr, -0.73%/yr, and 0.59%/yr, respectively. Our findings are consistent with recent measurements and inverse modelling studies but provide process-based estimates for anthropogenic emission and terrestrial flux. Wu et al. (Wu et al., 2012) reported measured CO2 concentration at Changbai Mountain forest site in northeast China increased by 1.76 ppmv/yr between 2003-
2010. With the atmospheric inversion modelling method, Fu et al. (2019b) estimated surface CO₂ in East Asia increased by 2-3 ppmv/yr between 2004-2012. These trends suggest that although anthropogenic emission increases at a steady rate in East Asia, photosynthetic uptake also serves as an increasing carbon sink due to enhanced EVI (0.29%/yr). However, as the interannual variability (IAV) of terrestrial flux is usually critically large and is affected by both vegetation itself and climate conditions (Fu et al., 2019b; Niu et al., 2017), simulation over longer time periods is necessary in future studies to conclusively comment on the changing trend of biosphere CO₂ in China.

3.3 Diurnal variation of near-surface CO₂ and influence factors

Finally, we examine the diurnal variation of meteorology and CO₂ data at Lin’an station as shown in Fig. 6 to reveal the temporal dynamics and atmospheric mixing of CO₂ at local scale. While both 21m (Fig. 7(a)) and 55m (Fig. 7(b)) CO₂ show prominent diurnal changes, the variations were larger in summer (JJA) than winter (DJF) and were larger at 21m than 55m, indicating the dominant influence of terrestrial flux over anthropogenic emission in determining the near surface CO₂ concentration. Fig. 6(c) presents the diurnal change of wind speed collected at 50m of the Lin’an tower. The higher wind speed between 10:00-22:00 local time suggests strong regional transport and mixing of CO₂ mainly occurs during this period. Fig. 6(d) and (g) present the WRF-VPRM and CT2019 simulation bias, at 21m and 55m respectively, against Lin’an tower data at 21m (note the Y scales are different), and Fig. 7(e) and (f) present the bias of CT2019 at 21m and 55m respectively. We find that both models prominently overestimate during night time underestimated during nighttime, which shall be attributed to the bias in simulating NEE. Li et al. (2020) reported the model overestimated nighttime NEE at a mixed forest site Wuying (47.15°N, 131.94°E) by 34% during the growing season (May-Sep.) according to EC eddy-covariance tower measurement. Fig. 6(f) and (h) present the simulated NEE by WRF-VPRM and CT2019, respectively, which show close correlations with the CO₂ simulation biases. While Lin’an is also covered by mixed forest, our evaluation suggests that WRF-VPRM may have also estimated lower nighttime ecosystem respiration during the non-growing season at Lin’an as it has a warmer climate condition than Wuying (Figure S5), and CT2019 has even greater bias for presenting the diurnal cycles of terrestrial flux.

We also find that planetary boundary layer height (PBLH) significantly affects diurnal accumulation and depletion of atmospheric CO₂ as shown in Fig. 28(a). During daytime in the growing season, photosynthetic uptake results in lower CO₂ concentration; meanwhile, PBLH is also high and allows rapid vertical mixing between near surface and upper air. During nighttime when photosynthesis stops, CO₂ from ecosystem respiration starts to accumulate in the shallow stable boundary layer, while the residual layer remains largely decoupled. Thus, atmospheric constituents with surface sources normally exhibit a vertical profile in which concentrations decrease with height in the stable boundary layer (Hu et al., 2020; Hu et al., 2012). Such boundary layer characteristics are confirmed by CO₂ vertical gradients at Lin’an. In this study, CO₂ at 55m height was consistently lower than the near surface air at 21m during nighttime due to accumulation of respired CO₂ in the stable boundary layer. As photosynthetic uptake depleted the near surface CO₂ and daytime boundary layer convection developed, the CO₂ gradient was gradually weakened from 06:00 to 11:00 LT and remained minimal through
The relationship between the near-surface CO₂ profile and boundary layer stability is further statistically examined. Fig. 28(b) presents the correlation between air temperature gradient (ΔT/ΔH) and CO₂ concentration gradient (ΔCO₂/ΔH) calculated with annual averaged diurnal profiles of tower observations, which averaged for 2016-2018, where ΔT, ΔH, and ΔCO₂ is the differences of temperature, height, and CO₂ concentration between the two tower levels, respectively. Fig. 8(b) clearly demonstrates the influence of boundary layer stability on the CO₂ vertical profile, as the correlation between ΔT/ΔH and ΔCO₂/ΔH reaches -0.98. On one hand, a more stable PBL with a strongly positive temperature gradient would promote surface CO₂ accumulation and lead to a strongly negative CO₂ gradient, especially under inversion conditions when upper air has higher temperature (orange area in Fig. 28(b)). Conversely, a strongly negative temperature gradient indicates stronger radiation, and subsequently greater photosynthesis and CO₂ depletion in the near surface layer, which would result in a positive CO₂ gradient (green area in Fig. 28(b)) implying a lower CO₂ concentration at the surface. While the diurnal variations of ΔCO₂ are primarily dictated by local biogenic CO₂ fluxes and boundary layer dynamics, the two minor daytime peaks of ΔCO₂ at Lin’an, at 10:00 and 18:00 LT (Fig. 28(a)) likely suggest influence of transport of CO₂ from urban plumes in the region; for example, from Hangzhou which is 60 km away from the tower. Due to rush-hours anthropogenic emissions, CO₂ enhancement at Hangzhou relative to a background site exhibits a prominent bimodal curve with two peaks during early morning and early evening (Pu et al., 2018). Depending on meteorological conditions, particularly wind fields, urban CO₂ plumes from cities such as Hangzhou may be transported to the Lin’an site. Due to higher altitude and stronger winds, wind profile increases with height at Lin’an according to observations (Figure S1). 55m at the Lin’an tower has a larger footprint than 21m, thus 55m on the tower is more likely affected by the urban plumes in the region than 21m. The 10:00 and 18:00 LT ΔCO₂ peaks at Lin’an likely suggest stronger CO₂ enhancement at 55m than at 21m from influence of regional anthropogenic emissions; the slight delay of these ΔCO₂ peaks relative to rush hours (at about 08:00 and 17:00 LT) further corroborate the hypothesis of delayed influence of transport of urban CO₂ from Hangzhou. Even though 55m has a larger footprint than at 21m and thus may be more likely affected by regional urban emissions, turbulent vertical mixing may reduce the different influence from regional urban emissions, which explains the fact that ΔCO₂ peaks are only minor. The influence of boundary layer conditions on CO₂ variability has been discussed in several studies through analysis of mountain site ground-based observations (Arrillaga et al., 2019; Esteki et al., 2017; Li et al., 2014), but our study applied tower data as direct evidence to demonstrate the significant impact of PBL thermal structure, which has rarely been documented. More importantly, although WRF-VPRM failed to capture the bimodal ΔCO₂ peaks at rush hours, because monthly ODIAC data lacked an hourly profile, our analysis reveals the critical importance of careful configuration of the PBL scheme and spatiotemporal distribution of anthropogenic emission for weather-biosphere modelling of atmospheric CO₂.

The rest of the daytime; at midday when photosynthesis reaches maximum intensity, CO₂ at 21m was even lower than at 55m. WRF-VPRM generally reproduced the diurnal profile but noticeably underestimated the intensity of nighttime CO₂ difference (ΔCO₂) likely due to the bias for simulating night time terrestrial flux as discussed above or underestimation of nighttime boundary layer stability by the PBL scheme (Hu et al., 2012).
4 Summary and Conclusions

In this study, the spatiotemporal variations of CO$_2$ in China are investigated with measurements from multiple datasets and a weather-biosphere coupled model simulation for 2016-2018. We find consistent higher concentrations over industrial areas with excessive anthropogenic emission and lower concentrations over densely vegetated areas. Observed CO$_2$ concentrations at Lin’an (427 ppmv) are significantly higher than remote ESRL sites (408 ppmv) although they are all established as “background” stations, indicating the dominant influence of anthropogenic emission at regional scales. The Lin’an tower data shows a large negative correlation (−0.8599) between vertical CO$_2$ concentration and air temperature gradients, suggesting the significant influence of boundary layer stability on CO$_2$ accumulation and depletion. The online coupled weather-biosphere model WRF-VPRM enables process-based estimations of contributions from anthropogenic emission (0.59 ppmv (0.15%)), terrestrial flux (0.16 ppmv (-0.04%)), and background concentration (405.70 ppmv (99.89%)) to average total XCO$_2$.

Respective simulation biases of surface CO$_2$ and XCO$_2$ are 0.69 ppmv (0.17%) and 0.76 ppmv (0.19%) against ESRL site observations and OCO-2 satellite product with correlations of 0.87 and 0.90, indicating overall good performance of the WRF-VPRM model. Maximum CO$_2$ concentrations are found in April and minimums are found in August for all three years, and the seasonality is reproduced well by the model, which also reveals that terrestrial flux and background concentration dominated the seasonality rather than anthropogenic emission.

A steadily increasing trend in XCO$_2$ by ~2.46 ppmv (−0.6%/yr) during the study period is demonstrated consistently by both model simulation and satellite product. Budget analysis suggests that anthropogenic emission increased by 0.83%/yr contributing to the 0.81%/yr growth rate of anthropogenic XCO$_2$ enhancement, 27% of which was offset by biosphere uptake. It is noted that terrestrial flux has significant inter-annual variability, thus a more robust estimation of the terrestrial flux trend should be obtained through a long-term study in the future. The background XCO$_2$, representing contributions from global circulation, increased by 2.37 ppmv (0.59%/yr), suggesting that CO$_2$ level in China was growing at the same rate as the rest of the world.

The most significant modelling bias is identified from validation against the Lin’an tower data, which WRF-VPRM 4km-grid simulation overestimated by about 4.281 ppmv (1.26%) with a correlation coefficient of 0.9282. The allocation of anthropogenic emission into the surface layer is partially responsible for this modelling bias because Lin’an is closely affected by upwind industrial mega cities in YRD, suggesting the need to include vertical profiles of fossil fuel combustion to properly redistribute the ODIAC for modelling purposes. However, in addition, diurnal variations of the bias suggest that the modelling discrepancy is likely due to also induced by large uncertainty associated with simulating nighttime ecosystem respiration. Representation and parameterization of photosynthetic carbon uptake in VPRM has been continuously improved during the past 10 years since its first release (Hu et al., 2020), but ecosystem respiration parameterization is still too simplified to fully represent the autotrophic and heterotrophic respiration of biomass. Li et al. (2020) and our study both reveal the urgent need to better calibrate VPRM parameterization over different vegetation types in China, and other methods such as inverse modelling is necessary to further validate the anthropogenic fluxes from ODIAC.
Nevertheless, WRF-VPRM is demonstrated to be a reliable tool to model the dynamics of CO₂ and exchange between the atmosphere and terrestrial flux. Most importantly, as the online coupled modelling system is able to simulate meteorology and biosphere processes simultaneously, it promotes the opportunity to investigate the interactions between atmospheric mixing and terrestrial flux (Carvalhais et al., 2014; Schimel et al., 2015) while comprehensively considering various factors from both sides that affect CO₂ in one coordinate frame, which could be a very helpful tool to support policy makers for balancing short-term carbon cycles at regional scales.

Data availability

The modelling output is accessible by contacting the corresponding author (yjjiang@pku.org.cn, xhu@ou.edu)

Author contributions

The concept and ideas to design the integrated simulation and observation analysis were devised by YJ, X-MH, and XD. Simulation was performed by X-MH. OCO-2 satellite product was collected and processed by X-MH. CT2019 assimilation data and ground-based observations was collected by XD. Tower measurement was conducted, processed, and analysed by QM, JP, and YJ. Model evaluation was performed by MY. The manuscript was prepared by XD and X-MH with input and feedback from YJ, MY, QM, JP, and GZ.

Competing interests

The authors declare that they have no conflict of interest.

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### Table 1. Evaluation statistics\(^1\) for WRF-VPRM 20km-grid simulation against OCO-2 satellite product at daily intervals

| Season | Vegetation type | Mean Obs. (ppmv) | Mean Sim. (ppmv) | MB\(^1\) \(\text{ppmv}\) | NMB\(^2\) \(\text{ppmv}\) | \(\text{se}^2\) | # of samples |
|--------|-----------------|------------------|------------------|-----------------|-----------------|----------------|----------------|
| Spring | other           | 406.85           | 407.81           | 0.0696c         | 0.23c           | 0.82           | 16123         |
|        | evergreen       | 407.52           | 407.89           | 0.36            | 0.09            | 0.73           | 1920          |
|        | deciduous       | 408.15           | 408.430          | \(0.27\)        | \(0.07\)        | 0.82           | 412           |
|        | mixed           | 407.79           | 408.21           | 0.41            | 0.10            | 0.79           | 4438          |
|        | shrubland       | 406.97           | 407.54           | 0.56            | 0.13            | 0.74           | 6550          |
|        | savanna         | 407.59           | 408.55           | 0.96            | 0.22            | 0.81           | 534           |
|        | grass           | 406.81           | 407.49           | 0.68            | 0.16            | 0.81           | 11170         |
|        | crops           | 407.50           | 408.29           | 0.79            | 0.19            | 0.82           | 13548         |
| Summer | other           | 403.90           | 404.84           | 0.93            | 0.23            | 0.88           | 13445         |
|        | evergreen       | 402.68           | 402.24           | -0.44           | -0.11           | 0.85           | 1082          |
|        | deciduous       | 400.39           | 399.39           | \(-1.01\)       | \(-0.25\)       | 0.82           | 527           |
|        | mixed           | 402.04           | 401.60           | -0.43           | -0.11           | 0.87           | 4312          |
|        | shrubland       | 403.92           | 404.41           | 0.48            | 0.12            | 0.85           | 5193          |
|        | savanna         | 404.62           | 404.60           | \(-0.02\)       | \(0.01\)        | 0.79           | 170           |
|        | grass           | 402.35           | 402.66           | 0.31            | 0.08            | 0.88           | 12588         |
|        | crops           | 402.86           | 403.52           | 0.66            | 0.16            | 0.87           | 7947          |
| Fall   | other           | 403.32           | 404.35           | 1.03            | 0.26            | 0.82           | 17054         |
|        | evergreen       | 403.93           | 403.19           | -0.74           | -0.18           | 0.71           | 1716          |
|        | deciduous       | 403.35           | 403.64           | \(0.28\)        | \(0.07\)        | 0.84           | 281           |
|        | mixed           | 403.64           | 403.95           | 0.31            | 0.08            | 0.83           | 3611          |
|        | shrubland       | 403.12           | 404.22           | \(1.10\)        | \(0.27\)        | 0.77           | 8532          |
|        | savanna         | 403.45           | 404.15           | 0.70            | 0.17            | 0.70           | 504           |
|        | grass           | 403.22           | 403.65           | 0.43            | 0.11            | 0.85           | 11176         |
|        | crops           | 403.76           | 404.80           | 1.04            | 0.26            | 0.80           | 13136         |
| Winter | other           | 404.76           | 405.80           | 1.03            | 0.26            | 0.80           | 13838         |
|        | evergreen       | 404.79           | 404.75           | \(-0.05\)       | \(-0.01\)       | 0.78           | 2671          |
|        | deciduous       | 405.38           | 406.65           | \(1.27\)        | \(0.31\)        | 0.79           | 135           |
|        | mixed           | 405.20           | 405.79           | 0.59            | 0.15            | 0.79           | 2108          |
|        | shrubland       | 404.76           | 405.84           | 1.09            | 0.27            | 0.79           | 7683          |
|        | savanna         | 404.63           | 405.83           | 1.20            | 0.30            | 0.75           | 1064          |
|        | grass           | 405.06           | 405.64           | 0.58            | 0.14            | 0.77           | 5967          |
|        | crops           | 405.17           | 406.36           | 1.19            | 0.29            | 0.79           | 15508         |

\(^1\)For each season, evaluation statistic with the worst performance (largest absolute value of NMB) is highlighted in red, and
the one with best performance (smallest absolute value of NMB) is highlighted with in bold font.

\(^2\)Mean bias was calculated as: \(MB = \frac{1}{N} \sum_{i=1}^{N} (\text{Sim}_i - \text{Obs}_i)\), and

\[
MB = \frac{1}{N} \sum_{i=1}^{N} (\text{Sim}_i - \text{Obs}_i), \quad \text{NMB} = \frac{1}{N} \sum_{i=1}^{N} (\text{Sim}_i - \text{Obs}_i)^2
\]

\text{correlation coefficient was calculated as: } \text{CC} = \frac{\sum_{i=1}^{N} (\text{Sim}_i - \text{Sim}) (\text{Obs}_i - \text{Obs})}{\sqrt{\sum_{i=1}^{N} (\text{Sim}_i - \text{Sim})^2 \sum_{i=1}^{N} (\text{Obs}_i - \text{Obs})^2}}

\text{where } \text{Sim} \text{ is the average of simulations, } \text{Obs} \text{ is the average of observations.}
For each season, evaluation statistic with the worst performance (largest absolute value of MB) is highlighted in red, and the one with best performance (smallest absolute value of MB) is highlighted with in bold font.
Figure 1: Annual averages of (a) ODIAC emission, (b) MODIS EVI, and (c) dominant vegetation type; and (d) photo of Lin’an observation tower; (e) photo of data analysis and recording system in the 20km simulation domain, and (d) terrain height of the 4km simulation domain. The locations of the ESRL sites, TCCON Hefei site, and Lin’an tower site are indicated with red circles, rectangles, and diamonds respectively in (a). The 4km domain is indicated with the red dash rectangle in (b), and the locations of Hangzhou and Shanghai are indicated with yellow triangles in (d).
Figure 2: Photos of the (a) Lin’an regional atmospheric background station and (b) the data analysis lab; and wind rose map at Lin’an derived from wind speed and wind direction observations for 2016-2018 at (c) 10m and (d) 50m.
Figure 2: 2016-2018 averages of WRF-VPRM simulations of (a) 1st layer (mid-layer height is 12km) CO$_2$ concentration, and (b) XCO$_2$ concentration; (c) WRF-VPRM simulated XCO$_2$ bias against OCO-2; (d)-(f) is same as (a)-(c) but for CT2019 (1st layer mid-level height is 25m). Markers in (c) represent the locations of ground-based sites.
Figure 34: Data pairs for OCO-2 against (a) WRF-VPRM and (b) CT2019; (c) TCCON-Hefei against WRF-VPRM and CT2019; Lin’an tower against WRF-VPRM 4km-grid simulation at (d) 21m and (e) 55m; and (f) ESRL TCCON-Hefei against WRF-VPRM and CT2019.
Figure 45: Monthly variations of (a) CO$_2$ at ESRL sites, (b) total (black) and background (BCG, grey) CO$_2$ (line) and XCO$_2$ (area and bar), (c) CO$_2$ at Lin'an station (averaged for daytime 21m and 55m data); (d) contributions from anthropogenic (ANT, orange) and biogenic (BIO, blue) for CO$_2$ (lines) and XCO$_2$ (bars); (f) ODIAC emission and MODIS EVI; and (e) Daily variation of XCO$_2$ at TCCON-Hefei site.
Figure S6: Daily XCO$_2$ from CT2019 (a-c) and WRF-VPRM (d-f), weather map from Korea Meteorological Administration (g-i). The blue box represents location of Hefei.
Figure 67: Seasonal mean diurnal variations of observed CO₂ at (a) 21m and (b) 55m, and (c) wind speed at 50m; WRF-VPRM simulation biases of CO₂ at (c) 21m and (d) 21m and (e) 55m, and (f); CT2019 simulated biases at (e) 21m and (f) 55m; Simulated NEE; from (g)-(i) are same as (d)-(f) but for WRF-VPRM and (h) CT2019.
Figure 78: (a) Average (2016-2018) diurnal variations of simulated (black line) and observed (red line) ΔCO₂ and simulated (blue line) PBLH at Lin’an station; and (b) correlation between CO₂ gradient between 55m and 21m (ΔCO₂/ΔH) and temperature gradient (ΔT/ΔH) at Lin’an station (diurnal data is averaged for each year respectively).