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Carbon Weather: Toward the next generation of regional greenhouse gas inversion systems

Quantifying CO_2 Uptakes Over Oceans Using LIDAR: A Tentative Experiment in Bohai Bay

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Abstract Oceans are widely regarded as major offsets for anthropogenic carbon emissions, leading to an evident lower measured atmospheric CO_2 concentration than expected. It is thus of great significance to develop effective means to monitor CO_2 fluxes over oceans globally. In this work, we utilized observations obtained by an airborne CO_2-IPDA LIDAR to evaluate the potential of such means in estimating sea-air CO_2 flux. During a flight experiment in 2019, we have estimated the CO_2 exchange rate, -1.5 ± 0.18 mmol/m^2/h, between the Bohai Bay and the atmosphere using equilibrium atmospheric boundary layer theory. These findings indicated that the forthcoming space-borne CO_2-IPDA LIDAR is capable of identifying CO_2 uptakes over oceans qualitatively, which would be a novel means and a basis for monitoring CO_2 fluxes over global oceans.

Plain Language Summary Oceans are regarded as important offsets to anthropogenic CO_2 emissions, delaying an increase in atmospheric CO_2 concentration. However, there is a lack of understanding of the intensity, distribution, and variability of marine carbon sinks. China is due to launch a space-borne LIDAR for measuring atmospheric CO_2 concentration globally in 2021. In this work, we utilized data collected during a flight test for the forthcoming satellite mission to explore the ability of such a novel means in revealing and estimating CO_2 fluxes of oceans quantitatively. Results confirmed an evident CO_2 uptake in the Bohai Bay and exhibited a promising prospect for the novel CO_2 measuring means.

Key Points:

- We detected XCO_2 gradient during a flight campaign in China
- Retrieve XCO_2 in the atmosphere boundary layer by cloud slicing method
- Provide a model to retrieve CO_2 flux over ocean by CO_2-IPDA system

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

Current observations show that the growth of the atmospheric CO_2 concentration is evidently lower than expected, suggesting that anthropogenic carbon emissions have been offset by some unclear carbon sinks. Oceans and terrestrial ecosystems are supposed to be responsible for the missing carbon sinks (Sabine et al., 2004; Takahashi et al., 2009). Scientists have utilized varieties of means to explore carbon fluxes of terrestrial ecosystems quantitatively in the recent decades, trying to narrow gaps between observations and estimates of models (Eldering et al., 2017; Watson et al., 2009). Comparing with progresses in estimating carbon fluxes of terrestrial ecosystems, we still have some barriers to a better understanding on carbon fluxes of oceans. We do know oceans offset anthropogenic carbon emissions but there are lacks of quantitative and accurate estimates of CO_2 fluxes over oceans. Therefore, we don't have insight knowledge on distributions and dynamics of ocean carbon uptakes. Some pieces of evidence suggest that there may be a trigger point beyond which CO_2 uptakes of oceans would rapidly decline or even shift to a net CO_2 emission (DeVries et al., 2019). Therefore, designing more measurement methods to estimate ocean CO_2 fluxes more reliably and efficiently is of great importance.

The difference of CO_2 partial pressures in seawater ($p(CO_2)_{sw}$) and overlying air ($p(CO_2)_{air}$) would cause a net transfer of CO_2 flux between ocean and atmosphere (Wanninkhof, 2014). Experiments for measuring ($p(CO_2)_{sw}$) during ship tracks have been implemented in different regions of the global sea, which help to

promote our understanding on ocean CO_2 flux (Bushinsky et al., 2019; Lanso et al., 2015). Those works proposed reliable models to calculate $p(CO_2)_{sw}$ using easily accessible in-situ measurements, such as the sea temperature, the chlorophyll and the salinity (Bai et al., 2015). However, there is a lack of means to measure $p(CO_2)_{sw}$ with a fine spatial resolution globally, resulting in ineffectively monitoring of ocean uptakes. Moreover, these models would not be appropriate to study monthly or seasonal characteristics of $p(CO_2)_{sw}$, as these field data are normally collected over a certain period by sensors equipped on ships. Therefore, there is an urgent need to explore the feasibility of measuring sea-air CO_2 gradients using remotely sensed data from spaceborne sensors. IPDA, an abbreviation for integrated path differential absorption, LiDAR can measure column-weighted dry-air mixing ratio of CO_2 (abbreviated as XCO_2 hereafter) with high accuracy and sensitivity. Both the United States and China have scheduled to launch satellites with IPDA LIDAR onboard shortly. Flight tests carried out by researchers of the United States and China demonstrated results with a precision of ~1 ppm can be achieved by airborne CO_2-IPDA LIDAR under various circumstances (Abshire et al., 2014; Zhu et al., 2020). Those results exhibited reliable performances of the forthcoming spaceborne CO_2-IPDA LIDAR, which would serve as novel means to monitor the carbon cycle and provide us valuable opportunities to quantitatively estimate CO_2 fluxes over oceans. Hence, it is now of great significance to explore the feasibility of measuring CO_2 fluxes using observations from a CO_2-IPDA LIDAR.

In this study, we evaluated the CO_2 exchange rate of the ocean using observations acquired during a flight test carried out in China on March 14, 2019. Firstly, an evident horizontal gradient of XCO_2 has been witnessed, exhibiting a lower XCO_2 over the ocean. Then, XCO_2 in the atmospheric boundary layer (ABL) and the free troposphere (FT) have been retrieved using the cloud slicing method (Mao et al., 2018; Ramanathan et al., 2015) to reflect the vertical gradients of atmospheric concentrations of CO_2. On that basis, we estimated the CO_2 exchange rate of the ocean using the concept of equilibrium of ABL (Betts & Ridgway, 1989; Gibert et al., 2007).

2. Instrument Description and Methods

2.1. Airborne ACDL LIDAR System

An airborne Atmospheric Carbon Dioxide LIDAR (ACDL), which consists of a CO_2-IPDA LIDAR, 1064 nm Mie LIDAR and 532 nm Mie LIDAR (details are given as Figure S1) (Wang et al., 2020; Zhu et al., 2020). In this study, we mainly used the data collected by the first two LIDARs.

2.2. Retrieving Method of XCO_2

$$XCO_2 = \frac{\ln\left(\frac{P_{off}\cdot P^0_{on}}{P_{on}\cdot P^0_{off}}\right)}{2\times 10^{-6}\cdot IWF} \tag{1}$$

$$IWF = \int_{R_G}^{R_A} \frac{\left(\sigma_{on}\left(p(r),T(r)\right)-\sigma_{off}\left(p(r),T(r)\right)\right)\cdot N_A\cdot P_{(r)}}{R\cdot T(r)(1+X_{H2O}(r))}dr \tag{2}$$

XCO_2 abbreviates for the column-weighted dry-air mixing ratio of CO_2. P_{on} and P_{off} represent the received powers of λ_{on} and λ_{off} pulses. P^0_{on} and P^0_{off} represent the transmitting powers of λ_{on} and λ_{off} pulses. IWF is the integrated weighting function of CO_2, σ_{on} and σ_{off} represent absorption cross-section of online and offline wavelengths (Shi et al., 2020), R_A is the altitude of the airborne platform, and R_G is the altitude of the hard target above sea level. $P(r)$ and $T(r)$ are atmospheric profiles of the pressure and the temperature, NA is the Avogadro constant, R is gas constant, XH_2O represents the column-averaged dry-air mixing ratio of water vapor (Shi et al., 2020; Zhu et al., 2020). All spectroscopic parameter regarding calculations of the IWF are according to the HITRAN2012 database (https://hitran.org/) (Rothman et al., 2013). The meteorological parameters used in this research were acquired by ERA5 reanalysis database, including pressure, temperature

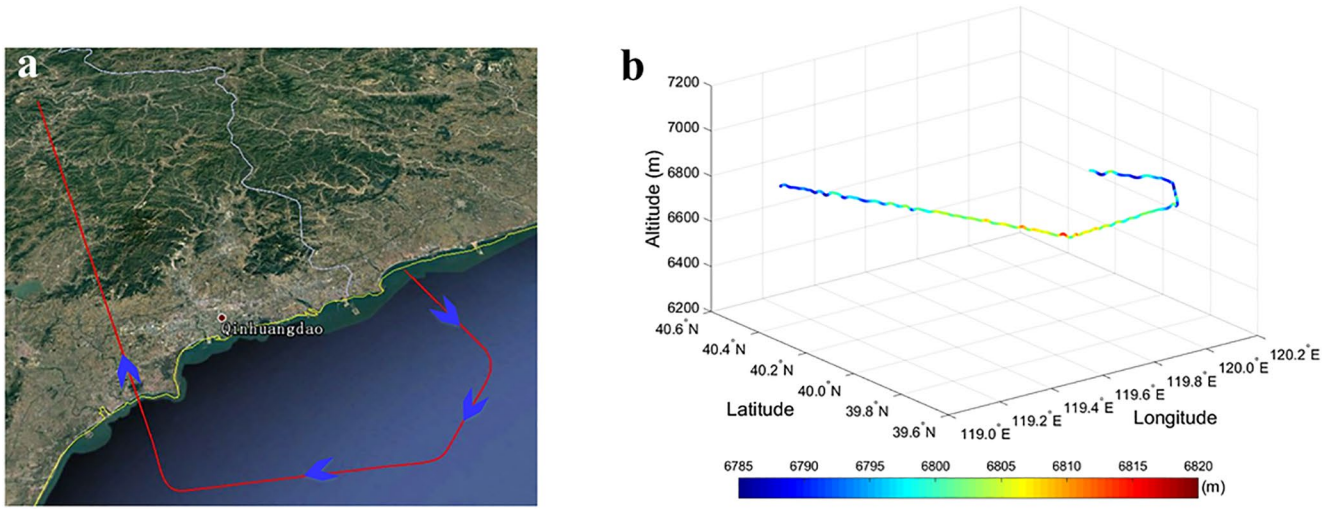

Figure 1. (a) The flight track on March 14, 2019. (b) The corresponding flight altitude on March 14, 2019.

and water vapor profile at different altitudes (Hersbach et al., 2020). In this study, the averaging time for XCO_2 retrievals is 1 s.

3. Result

3.1. Flight Campaign

We utilized data collected via a flight test on March 14, 2019 to retrieve XCO_2 over different land covers, including the ocean, the urban area and the mountainous area. Because this is the first-time developers carried out a flight test for the ACDL, there were some problems with the commissioning of the LIDAR system during the flight, resulting in a large proportion of unavailable data. A small fraction of data was used in this work and the corresponding flight track was shown in Figure 1a. The flight altitude was about 6.8 km (Figure 1b). In this study, we mainly discussed the ability of the IPDA LIDAR to estimate the CO_2 flux in Bohai Bay.

3.2. Validation of the Characteristic of CO_2 Diffusion over the Ocean

The measured atmospheric CO_2 concentration is a combination of regional backgrounds and local enhancements due to carbon sources and sinks. Signatures of surface CO_2 fluxes are diffusing through both vertical mixing and horizontal transportation. Hence, a carbon source or sink would result in a gradient of atmospheric CO_2 concentration in both the vertical and horizontal directions. In turn, the CO_2 flux could gradually be reversed by using CO_2 concentration gradients and a rational transport model. Therefore, the key purpose of this experiment is to figure out whether XCO_2 observations obtained by the IPDA-LIDAR can exhibit vertical and horizontal gradients of CO_2 concentrations.

3.2.1. The Horizontal Gradient of XCO_2

The XCO_2 retrievals obtained by the IPDA LIDAR and point mixing ratio of CO_2 obtained by the in-situ measuring equipment were demonstrated in Figure 2a. The blank regions are caused by the loss of the echo signal due to the large roll angle of the airplane as it turns, or the received signals are saturated. Figure 2a exhibits explicitly horizontal gradients of column and point CO_2 mixing ratios. Given that the downtown of Qinhuangdao locates in the plain area shown in Figure 2a, there must be anthropogenic carbon emissions in that region, especially in March when there is public heating service consuming large amounts of coals. Figure 2a shows that XCO_2 retrievals over the mountains area are lower than those over the urban area but are larger than those over the ocean, exhibiting evident XCO_2 gradients among different surfaces. The XCO_2 retrievals varied from 406.2 to 441.3 ppm during the flight track.

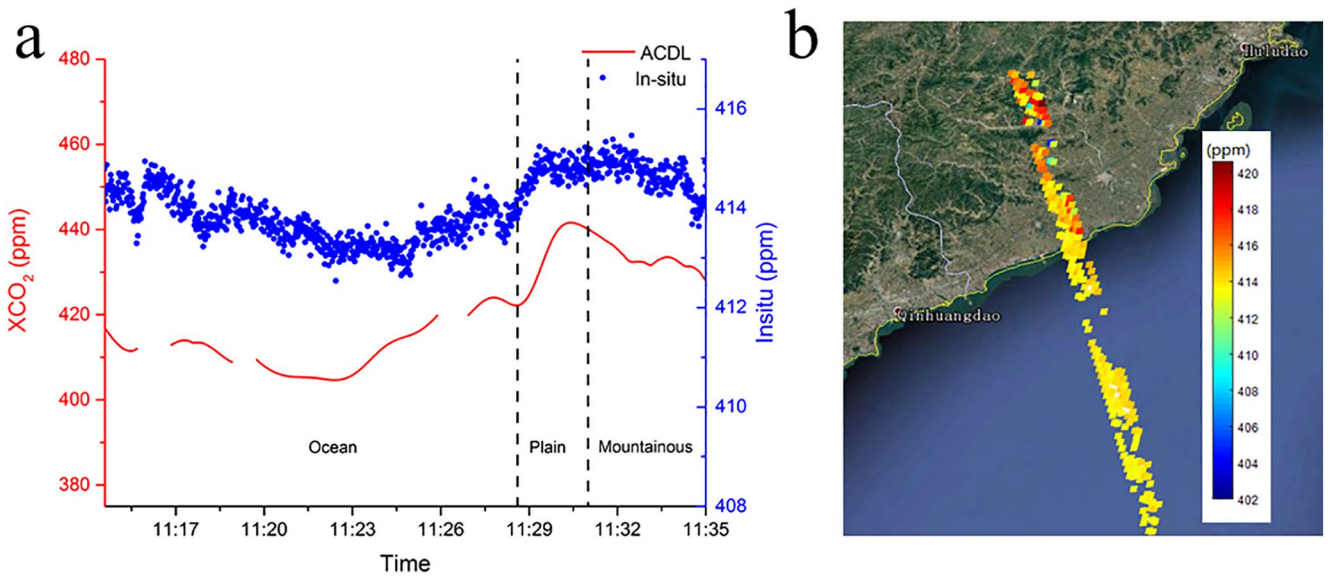

Figure 2. (a) The CO_2 concentration trend measured by the airborne IPDA system during the flight on March 14, 2019. (b) XCO_2 products of OCO-2 on March 16, 2019.

We also collected XCO_2 products of OCO-2 with a spatial resolution of 1.29 × 2.25 km to explore the land-sea gradients of CO_2 (Lu et al., 2015), OCO-2 data could be acquired from the MIRADOR platform at http://mirador.gsfc.nasa.gov. However, as XCO_2 products of OCO-2 suffers is too sparse in the selected study area on March 14, 2019. We have to utilize XCO_2 products from OCO-2 on March 16, 2019 in this section. Figure 2b demonstrates that the mean XCO_2 over ocean are about 2.0 ppm lower than those over the coastal land, also indicating a plausible CO_2 uptake in the ocean (see S2).

3.2.2. Vertical Gradient of XCO_2

The vertical gradient of CO_2 is harder to obtain when compared to that of horizontal gradient. Vertical profiles of CO_2 concentration not only help us gain insight into the carbon cycle process but also provide critical information for improvements on performances of passive remotes sensing of XCO_2 (Eldering et al., 2017). At present, an airborne/spaceborne IPDA LIDAR is still incapable of obtaining the range-resolved CO_2 mixing ratio with adequate precision. However, an XCO_2 gradient between the ABL and FT can be obtained under certain conditions. Moreover, such an XCO_2 gradient is a direct index to describe the CO_2 exchange between lower and upper atmospheres, thus laying the foundation for estimating underlying CO_2 fluxes. Cloud-reflected signals can be used to obtain above-cloud XCO_2 (Mao et al., 2018; Ramanathan et al., 2015). In addition, a critical foundation for obtaining XCO_2 gradient around ABL is to prove that the clouds we used as the reflector occurred at the top of ABL (Ramanathan et al., 2015). The method to determine the ABL could be found in S3.

Then, we could retrieve $XCO_{2(FT)}$ (XCO_2 above clouds) by the signals reflected by clouds. Signals obtained over the ocean are evidently weaker than those obtained over lands because of the lower reflectance of the ocean. Under such a circumstance, any cloud-attenuated beams yielded very a low SNR of received signals, it's difficult to directly retrieve $XCO_{2(ABL)}$ (XCO_2 below clouds) by these signals. Hence, we utilized another method to retrieve $XCO_{2(ABL)}$, as is shown in Figure 3. There are three bases for further determination of $XCO_{2(ABL)}$. Firstly, the integrated path was divided into FT and ABL. Secondly, XCO_2 kept constant in adjacent positions (100 measurements, or 3.33 s). Thirdly, $XCO_{2(FT)}$ can be retrieved using signals reflected by clouds. Then, we can calculate $XCO_{2(ABL)}$ using Equation 3.

$$XCO_{2(ABL)} = \frac{DAOD_t - DAOD_{FT}}{IWF_{ABL}} \quad (3)$$

$DAOD_t$ is the total differential absorption optical depth of CO_2 between the airplane and the sea surface, $DAOD_{FT}$ is the differential absorption optical depth of CO_2 between clouds and flight height (Shi et al., 2020),

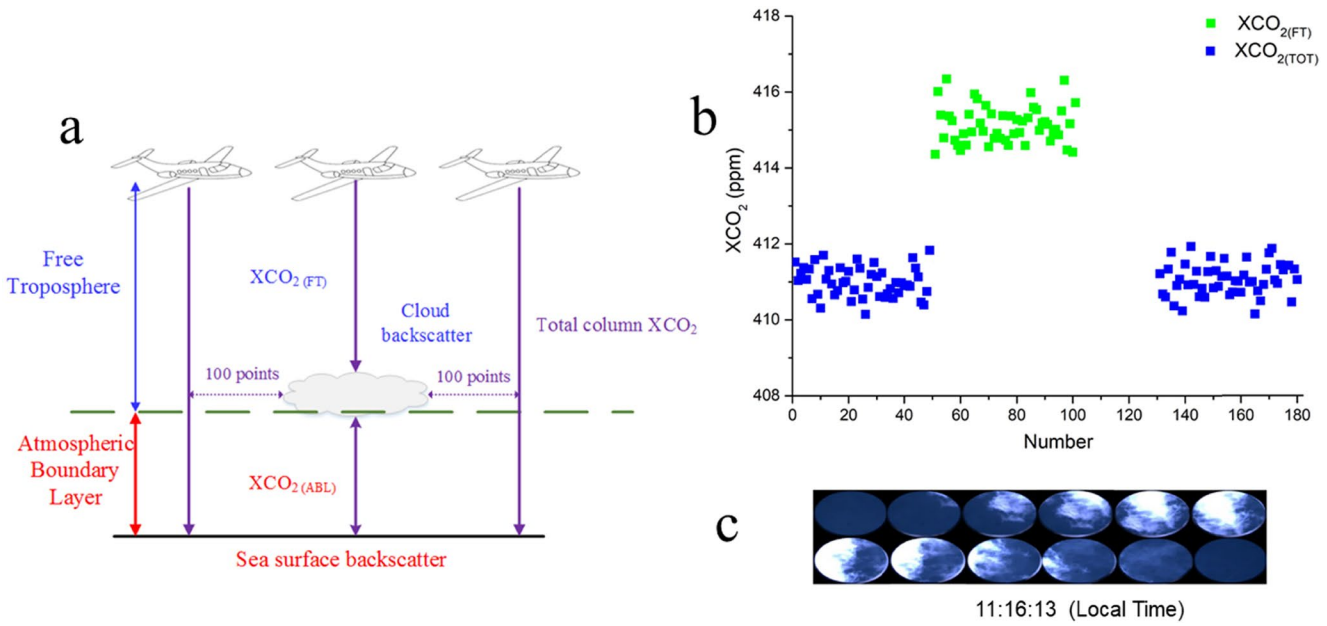

Figure 3. (a) The framework to show how to retrieve the $XCO_{2(ABL)}$ and $XCO_{2(FT)}$ through the scatter signal of clouds and sea surface on March 14, 2019. (b) A case that $XCO_{2(FT)}$ and $XCO_{2(TOT)}$ around a cloud. (c) A case that clouds exit during the detected path on March 14, 2019.

IWF_{ABL} is the integrated weighting function in ABLH. The mean value of XCO_2 during the paths that adjacent to the clouds is 411.1 ppm, the mean value of $XCO_{2(FT)}$ is 414.9 ppm, we then calculated $XCO_{2(ABL)}$ as 408.5 ppm. The gradient between $XCO_{2(ABL)}$ and $XCO_{2(FT)}$ is determined as −6.5 ppm ($XCO_{2(ABL)} - XCO_{2(FT)}$). The accuracy evaluation of XCO_2 retrieved by IPDA would be found in S4.

4. Discussion

After obtaining the vertical gradient of CO_2, we can further estimate the CO_2 flux of the ocean using the theory of equilibrium convective boundary layer. Then, we compare its outcomes with results calculated by the principle of the partial pressure of CO_2.

4.1. Sea-Air CO_2 Flux Calculation Using the Concept of Equilibrium Atmospheric Boundary Layer

The theory of equilibrium ABL was applied in forests and oceans in previous works (Dang et al., 2011; Gibert et al., 2007), the exchange rate of CO_2 would be calculated using Equation 4, the detailed process of this method as shown in S5.

$$F_c = \rho W\left(C_{FT} - C\right) \tag{4}$$

F_c is the net surface flux of CO_2, C represents the well-mixed CO_2 mixing ratio in the ABL, C_{FT} is the CO_2 mixing ratio in the FT, ρ is air density in ABL, W is the vertical velocity at the top of ABL (Betts et al., 2004; Dang et al., 2011). In this study, W was acquired by WRF with a resolution of 1 × 1 km based on the original meteorological data from ERA5 database (Kalnay et al., 1996).

Hence, the results of section 3.2 would be used to calculate the CO_2 exchange rate over oceans according to S3. We yielded air-sea CO_2 flux as −1.5 mmol/m^2/h, suggesting a net CO_2 uptake over the ocean. The magnitude of this result is consistent with the CO_2 carbon fluxes in coastal zones acquired by eddy covariance observation in previous studies (Chien et al., 2018; Gutiérrez-Loza & Ocampo-Torres, 2016; Kahl et al., 2017), see Table S1. The intensity of carbon flux exchange caused by seawater at the coastal zone is much greater than that of open sea areas (Nellemann et al., 2009; Tokoro et al., 2014).

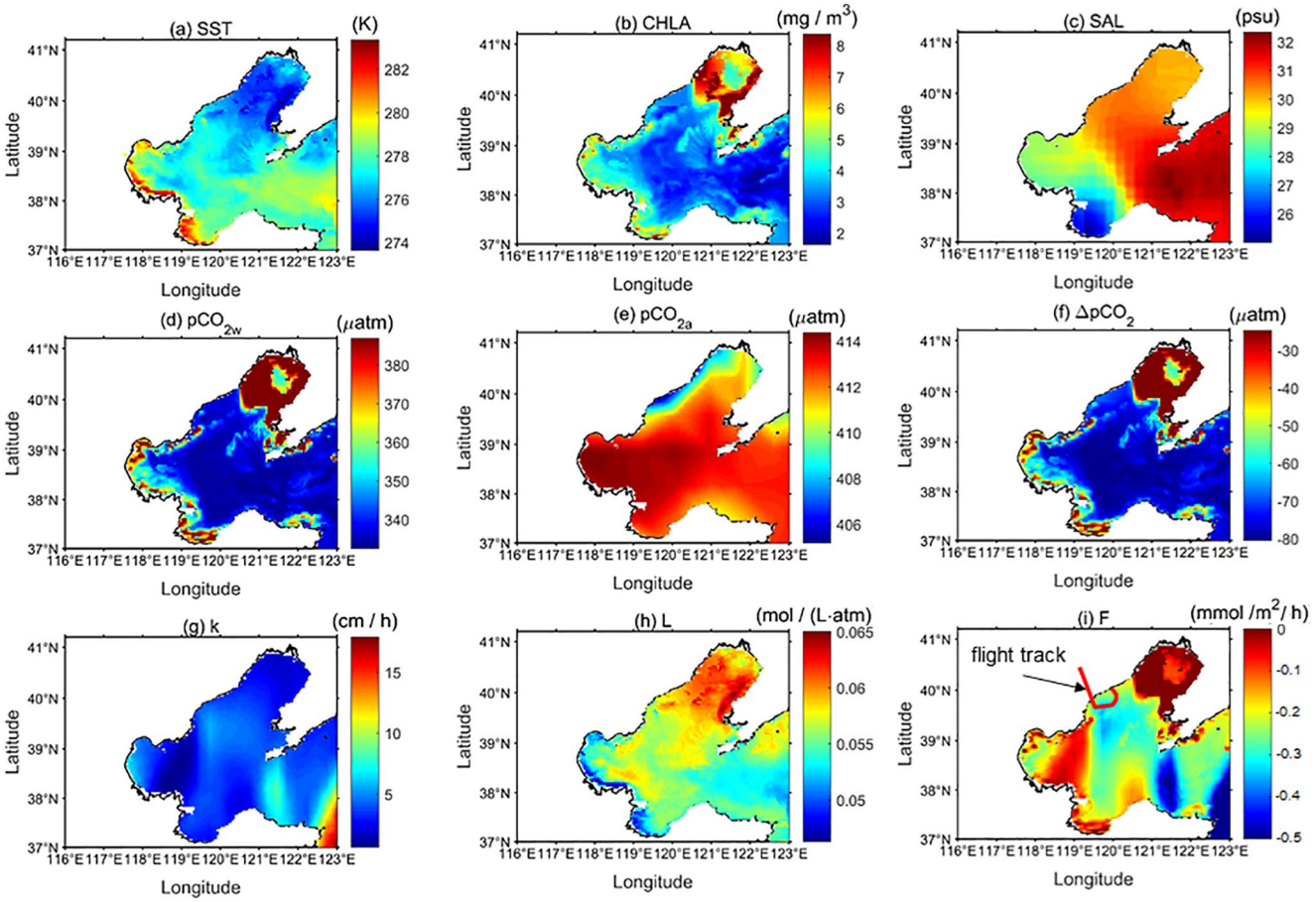

Figure 4. Information of parameters related with sea-air CO_2 flux, including (a) SST; (b) Chlorophyll a; (c) salinity; (d) pCO_2w; (e) $p(CO_2)a$; (f) ΔpCO_2; (g) K; (h) L; (i) F.

Two uncertainties contribute to the calculation of ocean fluxes, originating from the measurement uncertainties of $XCO_{2(ABL)}$ (ε_a) and $XCO_{2(FT)}$ (ε_b). The total uncertainty of CO_2 flux estimation (ε) would be calculated as:

$$\varepsilon = \pm \frac{\sqrt{\varepsilon_a^{\,2} + \varepsilon_b^{\,2}}}{\left|\left(C_{FT} - C\right)\right|} \cdot 100 \tag{5}$$

The uncertainty of ε_a is 0.62 ppm, the uncertainty of ε_b is 0.47 ppm, and the total uncertainty of CO_2 flux is about 11.7%.

4.2. Estimation of CO_2 Flux Based on the Partial Pressure of CO_2

Another widely used method to estimate sea-air CO_2 flux is based on the theory of partial pressure of CO_2 (Tao et al., 2012; Wanninkhof, 2014), the parameters and method description as shown in S6 and Table S2.

Parameters and results of net sea-air CO_2 flux calculated by this method are shown in Figure 4. Figure 4 demonstrates most of the Bohai Bay are sinks for CO_2. The exchange rate of CO_2 from the atmosphere into the ocean varies from 0.1 to 0.5 mmol/m^2/h. The estimated net air-sea exchange rate by this method is −0.32 mmol/m^2/h (see S7) (Nehrkorn et al., 2010), almost four times lower than the result calculated in section 4.1. The coastal air-sea CO_2 flux in coastal zone undergoes sharp changes during daytime (Kahl et al., 2017). Many studies have found that the carbon flux of CO_2 near the coast is much greater than that calculated by the principle of CO_2 partial pressure (Gutiérrez-Loza & Ocampo-Torres, 2016; Tokoro

et al., 2014). We expect that the future space-borne CO_2-IPDA satellite would a large number of observations to explore the causes of this phenomenon.

5. Conclusion

In this study, evidence indicating a strong CO_2 uptake in the Bohai Bay was demonstrated. It is witnessed that the horizontal gradient of XCO_2 between the ocean and the coastal land is more than 15 ppm and the vertical gradient of XCO_2 between ABL and FT is about 6.5 ppm. Both results strongly suggest an evident CO_2 sink in the ocean during the experiment period. Moreover, the exchange rate of CO_2 between ocean and atmosphere has been determined quantitatively using the theory of equilibrium convective boundary layer as -1.5 ± 0.18 mmol/m^2/h. The result of CO_2 flux calculated by CO_2 partial pressure principle also indicated studied region was carbon sink during the discussed time. This work confirms that it is feasible to quantitatively estimate CO_2 fluxes over oceans using an IPDA-LIDAR. Given that the space-borne CO_2-IPDA LIDAR is expected to be launched at the end of 2021, it would provide us another means to estimate CO_2 fluxes over oceans globally, helping solve the mystery of the missing carbon.

Data Availability Statement

We acknowledge Shanghai Academy of Spaceflight Technology provides initial LiDAR data (https://datadryad.org/stash/share/O9yWVRh8_-skyvwCFvcXM23KbI95Y6bQDsaW_nRbbXQ). We thank NASA and the OCO-2 project for making OCO-2 data available for this work (http://mirador.gsfc.nasa.gov). We thank MODIS project for making Chlorophyll and sea surface temperature data available for this work (https://oceandata.sci.gsfc.nasa.gov/MODIS-Aqua/Mapped/Daily/4km). We thank the team of ERA5 allowing us to use the data of meteorological fields (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5).

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