

1 *Supplement information for*

2 **Atmospheric CO₂ dynamics in a coastal megacity: spatiotemporal**
3 **patterns, sea-land breeze impacts, and anthropogenic-biogenic**
4 **emission partitioning**

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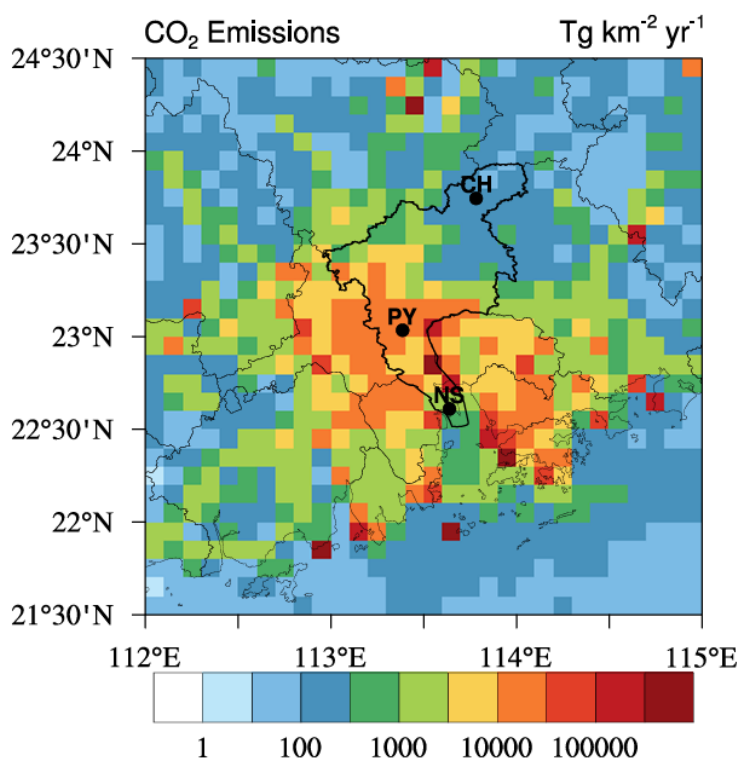
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15 In this document, we describe: (1) site selection strategy; (2) monitoring system and principles of the instruments; (3) wind
16 fields; (4) background concentrations.

17 1) Site selection strategy

18 The site selection process for Guangzhou's greenhouse gas monitoring network comprises preliminary screening, precise
19 screening, and field surveys. The preliminary screening phase involves analyzing the city's dominant wind directions,
20 topographic features, and emission distribution to establish a baseline understanding (Fig. S1). Drawing on experiences from
21 other cities and considering local conditions and construction costs, the initial plan proposed three monitoring stations:

- 22 (1) Urban station: located in the densely populated central urban area to monitor atmospheric CO₂ signals driven by
23 anthropogenic emissions.
- 24 (2) Coastal station: positioned in the southern coastal region to study the impacts of regional transport and coastal-specific
25 weather systems (notably sea-land breezes) on CO₂ concentrations.
- 26 (3) Suburban station: situated in the northern suburbs to capture CO₂ dynamics under urban ecosystem conditions.



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28 **Figure S1.** CO₂ emissions in Guangzhou and surrounding cities based on the 2023 EDGAR emission inventory (Crippa et al., 2024).

29 After determining the number of stations, satellite remote sensing was used to identify high-, medium-, and low-CO₂
30 concentration zones. A cluster analysis model screened 5–10 times the proposed number of candidate sites. These candidates
31 were further evaluated using the WRF-STILT model to analyze their sensitivity to CO₂ footprints, ensuring representative and
32 sensitive site selection. Field surveys for final site selection included the following criteria:

- 33 (1) Emission sources: greenhouse gas emission sources within a 1-km radius should be maintained at minimal levels.

- (2) Sampling accessibility: a 360° horizontal capture space around the sampling inlet.
- (3) Geological stability: long-term geological stability to avoid impacts from floods, wildfires, or landslides.
- (4) Infrastructure: minimal electromagnetic interference, stable power supply, lightning protection, and accessible communication lines.
- (5) Mobile monitoring: at least one week of mobile greenhouse gas monitoring at each candidate site.

Priority was given to newly constructed towers or existing open high towers for sampling, as these structures minimize airflow disturbances from supporting frameworks (Verhulst et al., 2017). Towers exceeding 50 meters in height were preferred; however, such platforms account for less than 1 % in Guangzhou and are often located in inaccessible areas. Thus, sites near 50 meters were prioritized to ensure well-mixed air sampling and avoid local anthropogenic or natural source-sink influences.

2) Monitoring system

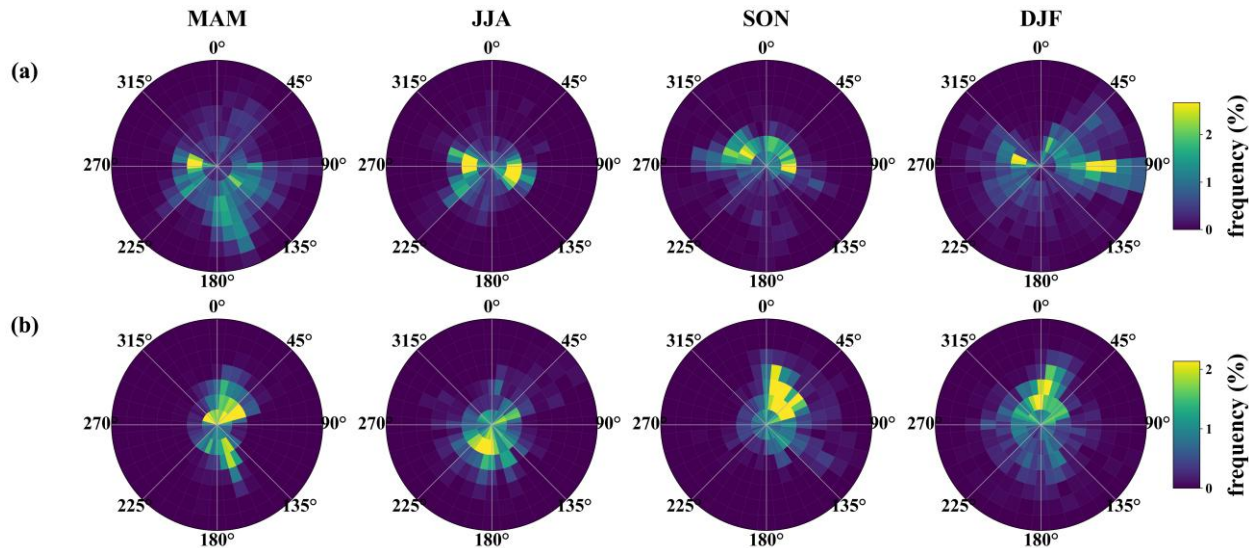
At the inlet front end, hydrophobic PTFE capsule filters and stainless steel tube clamps are installed. The capsule filters remove solid particles larger than 10 μm . Ambient air samples are drawn through 10 mm diameter PTFE-coated black aluminum-plastic tubes via a vacuum pump, with a maximum flow rate of 2 L min^{-1} . Additional filters in the tubing system remove solid particles larger than 2 μm and liquid droplets larger than 0.03 mm. The gas passes through a primary dehumidifier to reduce the dew point temperature to 2–10 °C, preventing condensation in the intake pipes. An ultra-low-temperature automatic cryogenic trap and a multi-channel gas intake system are also installed. Using built-in compressor cooling, the aluminum alloy cryogenic trap can lower the dew point temperature to –50 °C. Optimization of the control program minimizes maintenance requirements for the condensation dehumidifier, ensuring optimal system performance and reducing data artifacts during gas channel switching. Before entering the analyzers, samples pass through the calibration module, where periodic calibration curve establishment and target gas verification ensure measurement precision, accuracy, and long-term stability (detailed in Section 2.3)

The Picarro G2401 greenhouse gas analyzer employs Cavity Ring-Down Spectroscopy (CRDS). Its working principle is based on the near-infrared absorption spectrum unique to greenhouse gases like CO_2 . When an infrared laser is reflected by high-reflectivity mirrors within the measurement cavity, the light intensity decays exponentially. The gas concentration is determined by calculating the difference in ring-down time between an empty cavity and one filled with the target gas. The ABB GLA331-GGA greenhouse gas analyzer utilizes Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS). This technology creates an optical cavity using two highly reflective mirrors, enabling the laser to undergo multiple reflections between them. This amplifies the absorption signal, enhancing measurement sensitivity.

64 **3) Wind fields**

65 Wind speed and direction are critical indicators of temporal variations in atmospheric CO₂ mole fractions. Figure S2 presents
66 seasonal wind frequency monitoring results for the NS and PY stations, with wind speed resolution of 1.5 m s⁻¹ and directional
67 resolution of 10° :

- 68 (1) NS Station: average wind speed = 2.86 m s⁻¹. Dominant southerly winds in spring/summer (64 % and 57 % frequency)
69 and northerly winds in autumn/winter (66 % and 56 %).
70 (2) PY Station: average wind speed = 2.12 m s⁻¹. Prevailing northeastern and southeastern winds across seasons.



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72 **Figure S2.** Observed wind frequency as a function of wind speed and wind direction in spring (MAM), summer (JJA), autumn
73 (SON), and winter (DJF) at NS (a) and PY (b) from January 2023 to September 2024.

74 **4) Background concentrations**

75 Real-time monitoring of atmospheric background values entirely unaffected by local sources and sinks in urban areas is
76 challenging. Due to technical and logistical constraints such as site accessibility and power supply, very few monitoring stations
77 are sufficiently remote to remain permanently exposed to pristine air masses (Fang et al., 2015). With urbanization, many
78 regional background stations in suburban areas are increasingly influenced by urban carbon sources, leading to overestimated
79 urban background values (Chen et al., 2024). Here, urban background concentrations are defined as representative values of
80 air masses entering the city from upwind directions, unaffected by local sources or sinks (Verhulst et al., 2017; Mitchell et al.,
81 2018).

82
83 This study combines dominant urban wind patterns, atmospheric transport models, NOAA Earth System Research
84 Laboratory/Global Monitoring Laboratory (NOAA GML) background stations, and Carbon Tracker assimilation data to
85 determine atmospheric CO₂ and CO background values for Guangzhou. Seasonal dominant wind directions at urban stations

86 exhibited similar characteristics (Fig. S2), prompting a focus on summer and winter for air mass origin analysis. Figure S3
 87 illustrates the spatial distribution of average afternoon (12:00–16:00) atmospheric footprints during summer (July) and winter
 88 (December) over Guangzhou. Summer footprints primarily spanned southern urban and marine regions, while winter footprints
 89 extended to northeastern and southeastern urban and marine areas. By tracking air particles entering the city and using samples
 90 from the CO₂ GLOBALVIEWplus v10.1 ObsPack (Schuldt et al., 2024), two NOAA GML background stations—Hok Tsui
 91 (HKG) and Dongsha Island (DSI)—were identified as potential marine background references for Guangzhou. Air particles
 92 from upwind directions passed through both stations in all seasons.

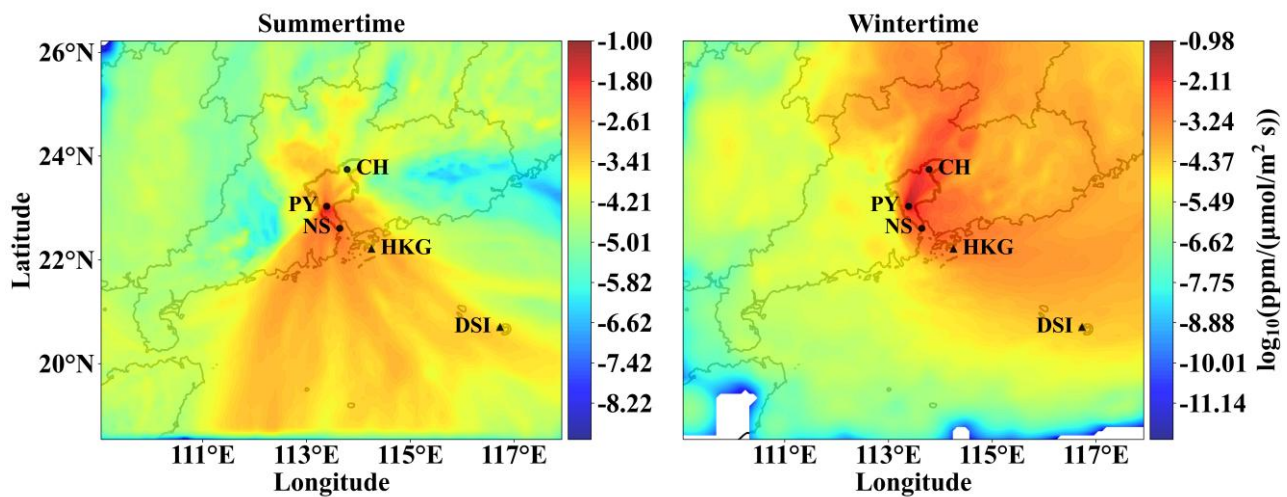


Figure S3. Spatial distribution of average atmospheric footprints during afternoon hours (12:00–16:00) in summer (July) and winter (December) based on the PY station.

96 We compared decadal CO₂ time series from these stations (Fig. S4) and applied CCGCRV curve-fitting software for smoothing
 97 (Thoning et al., 1989). HKG data showed significant deviations from smoothed trends, suggesting contamination by local CO₂
 98 surface fluxes. In contrast, DSI data consistently fell within HKG's range, indicating its suitability as Guangzhou's marine
 99 background. For further validation, we compared concentration values from NOAA's CarbonTracker (version CT-NRT.v2024-
 100 5) at grid cells where air particles last resided with DSI observations (Jacobson et al., 2024). During 2023, residuals between
 101 smoothed values were < 2 ppm, with a mean residual of 0.81 ± 0.85 ppm (Fig. S5), confirming DSI's reliability. Due to DSI's
 102 sparse weekly flask sampling, Carbon Tracker data were adopted for marine CO₂ background concentrations. For CO, lacking
 103 Carbon Tracker data, smoothed DSI CO observations served as the urban background reference.

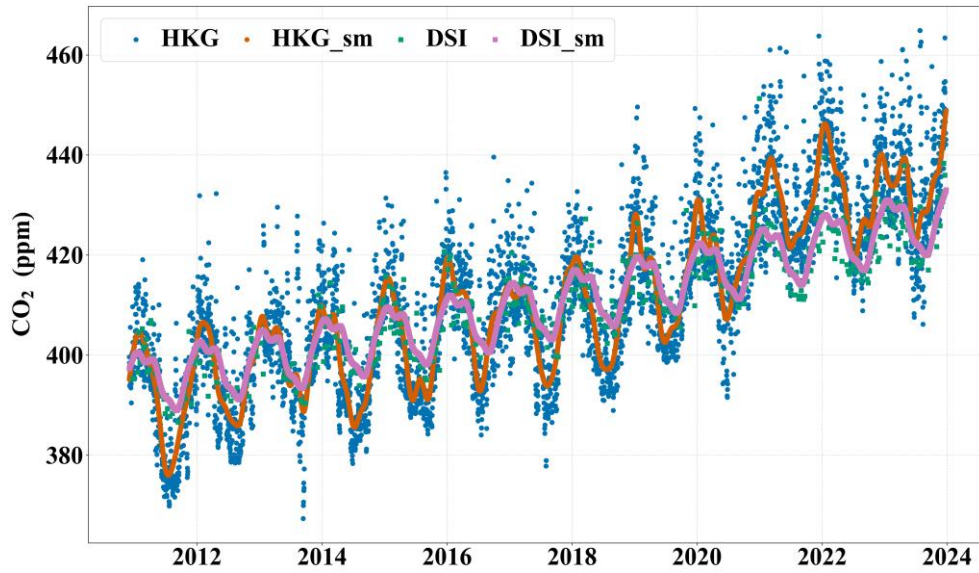


Figure S4. Time series of atmospheric CO₂ concentrations showing both raw monitored data and smoothed values at HKG and DSI stations from November 2, 2010, to December 31, 2023.

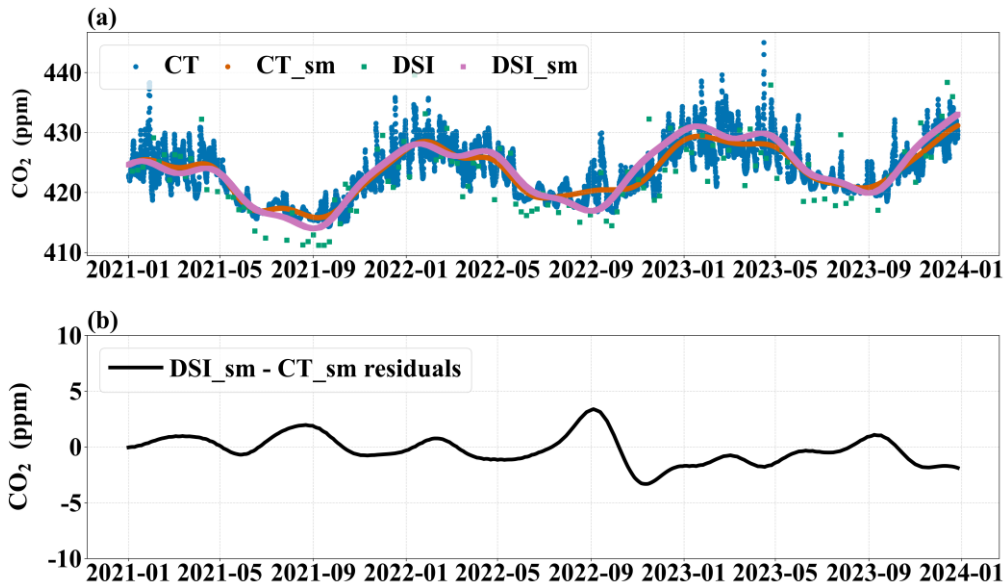


Figure S5. (a) Time series of atmospheric CO₂ concentrations showing both raw monitored/simulated data and smoothed values for DSI and Carbon Tracker from January 1, 2021, to December 26, 2023. (b) Residuals between DSI and Carbon Tracker smoothed values.

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Table S1 Summary of sea-land breeze day (SLBD) identification methods

Study area	Data	SLBD identification criteria	References
Pearl River Estuary	Observational data	Nighttime offshore winds or calm; ≥ 4 consecutive daytime onshore winds; $\Delta P \leq 5$ hPa; land-sea $\Delta T > 3$ °C	(Qiu and Fan, 2013)
Tianjin	Observational data	≤ 1 isobar line on surface weather maps; land-sea $\Delta T > 0$ °C; ≥ 3 consecutive onshore/offshore winds from sunrise to 2 h post-sunset	(Hao et al., 2017)
West African Coast	Reanalysis & observational data	Offshore/calm winds 6 h before sunrise and 2 h after sunrise; ≥ 2 consecutive onshore winds post-sunrise; $\Delta T > 0$ °C; ≥ 4 h daily sunlight	(Coulibaly et al., 2021)
Zhuhai	Observational data	24 h average wind speed < 10 m s ⁻¹ ; ≥ 3 occurrences of land/sea breeze; absolute wind direction difference between 02:00 and 14:00 = 90–270°	(Zhang et al., 2024)
Pearl River Estuary	Observational data	≤ 1 isobar line at 08:00 and 20:00; ≥ 4 consecutive land/sea breeze hours or ≥ 4 occurrences within 5 consecutive hours; no precipitation 3 h before/after SLB transition	(Mai et al., 2024)
Coastal China	Reanalysis & observational data	24 h average wind speed < 10 m s ⁻¹ ; ≥ 4 consecutive land/sea breeze hours; alternating land-sea breeze cycles within 24 h	(Huang et al., 2025)

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Table S2 Seasonal temperature averages (°C) at each station

Station	Season	20:00-05:00	09:00-18:00	0:00-23:00
CH	Autumn	23.95	26.12	23.95
	Spring	21.84	23.36	21.84
	Summer	27.80	29.77	27.80
	Winter	14.26	16.07	14.26
PY	Autumn	27.19	28.70	27.19
	Spring	24.18	25.41	24.18
	Summer	30.21	31.67	30.21
	Winter	17.38	18.61	17.38
CH	Autumn	27.57	29.32	27.57
	Spring	24.10	25.10	24.10
	Summer	30.10	31.32	30.10
	Winter	17.91	18.85	17.91

* CH station's temperature measurements were collected from an atmospheric monitoring station 1 km away

Table S3 Marine background concentration uncertainties

Uncertainty estimates		summer	winter
CO ₂ (ppm)	CT _{co2,r}	0.46	0.45
	CT _{co2,s}	0.84	0.54
	BG _{u,co2}	0.96	0.70
CO (ppb)	OBS _{co,r}	2.10	11.79
	OBS _{co,s}	12.50	14.08
	BG _{u,co}	12.68	18.36

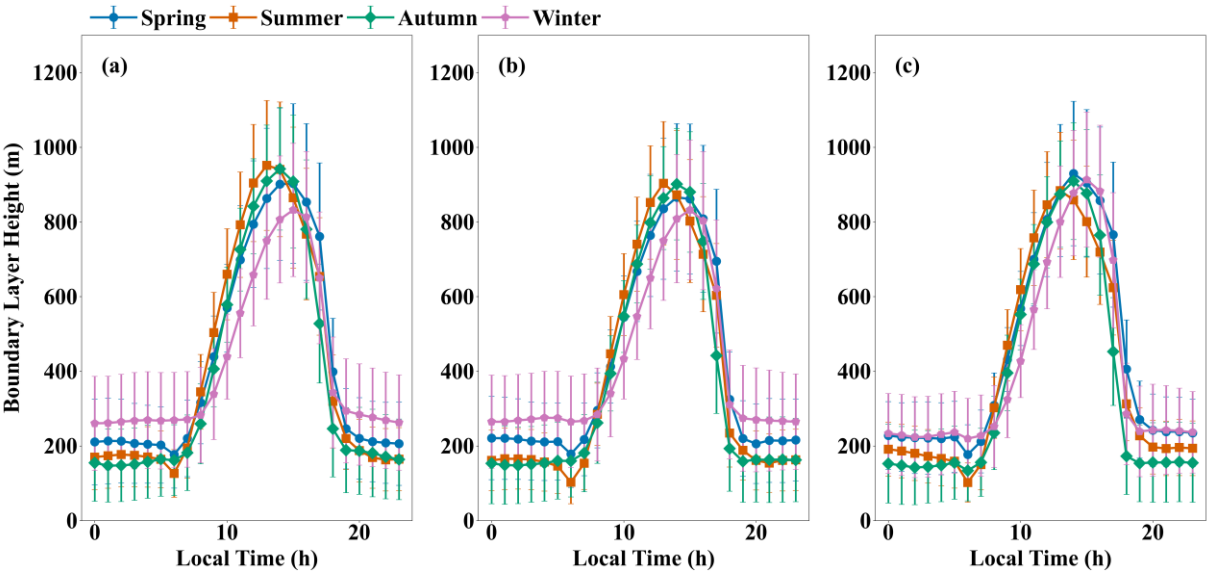


Figure S6. Diurnal variations in atmospheric boundary layer height at the (a) NS, (b) PY, and (c) CH stations across seasons. The planetary boundary layer height data were obtained from the ERA5 reanalysis product with a spatial resolution of $0.25^{\circ} \times 0.25^{\circ}$ (Hersbach, 2023). Error bars indicate ± 1 standard deviation (SD).

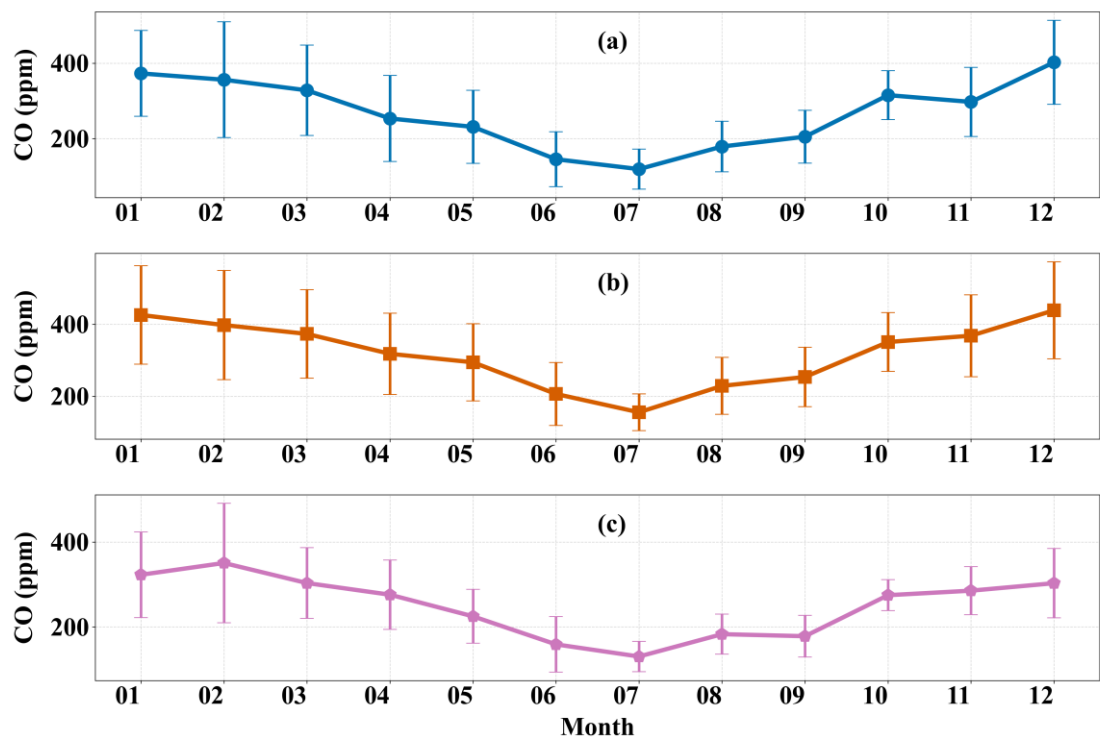


Figure S7. Monthly mean CO concentrations at the (a) NS, (b) PY, and (c) CH stations. Error bars indicate ± 1 SD.

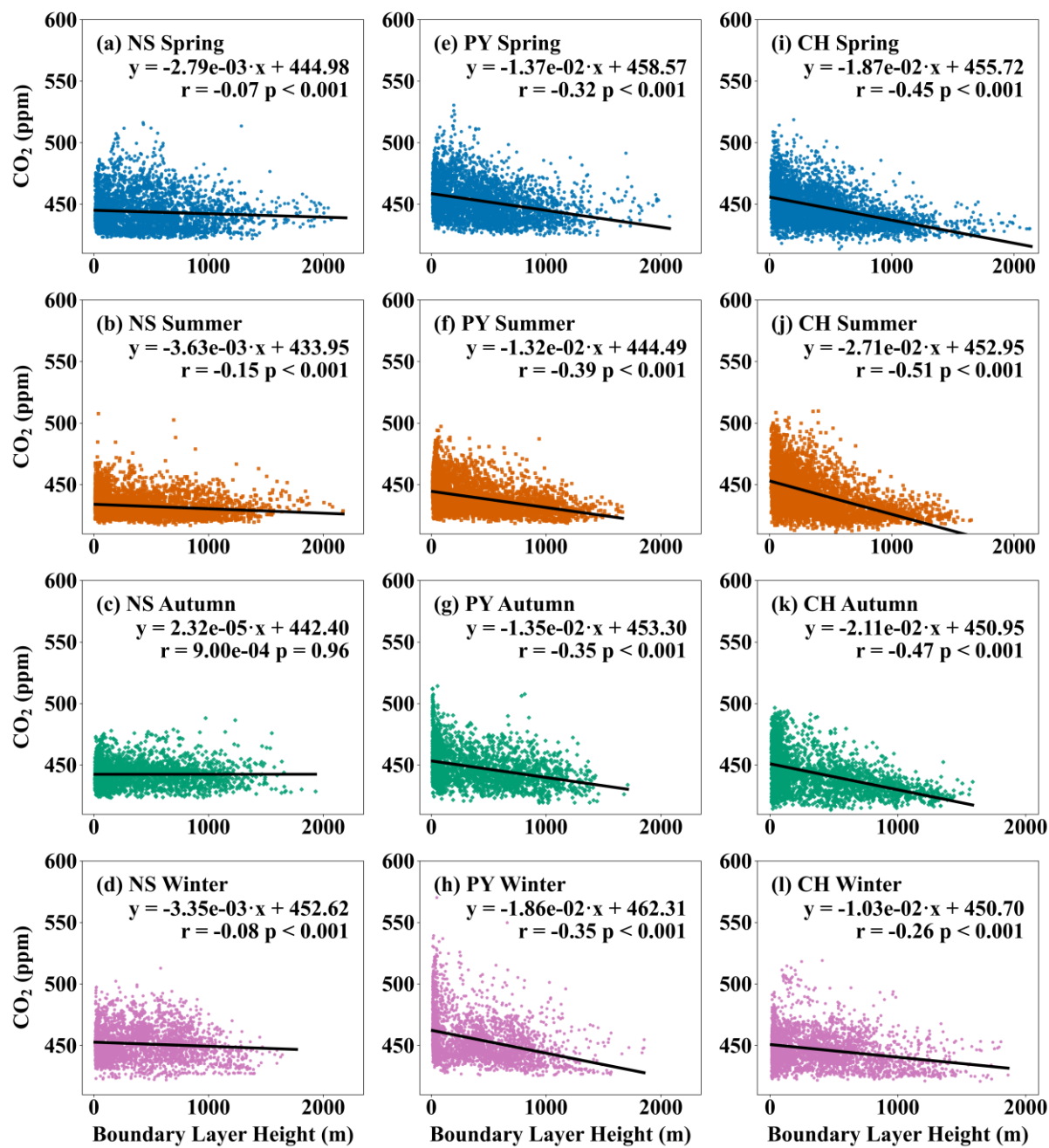


Figure S8. Correlations between atmospheric CO₂ concentrations and boundary layer height at the NS (a–d), PY (e–h), and CH (i–l) stations across seasons.

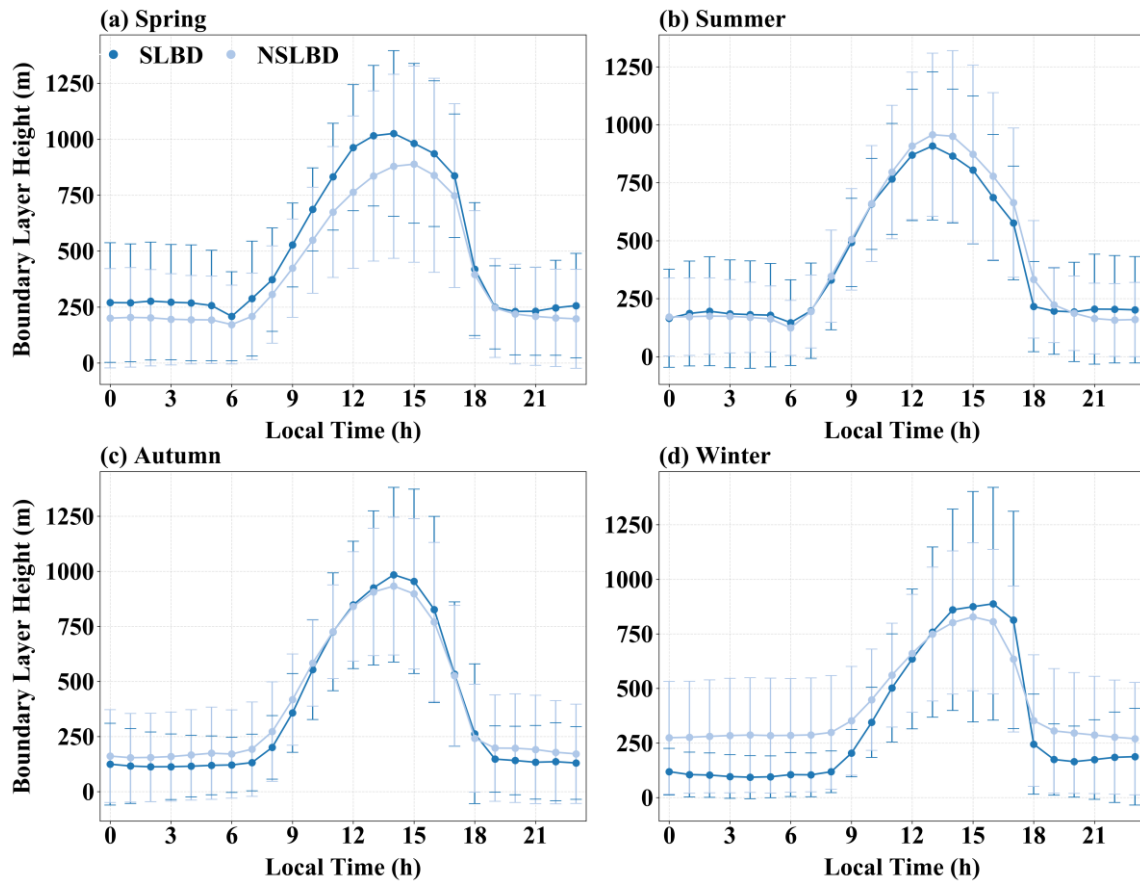


Figure S9. Diurnal variations in atmospheric boundary layer height at the NS station during sea-land breeze days (SLBD) and non-SLB days (NSLBD) across seasons. Error bars indicate ± 1 SD.

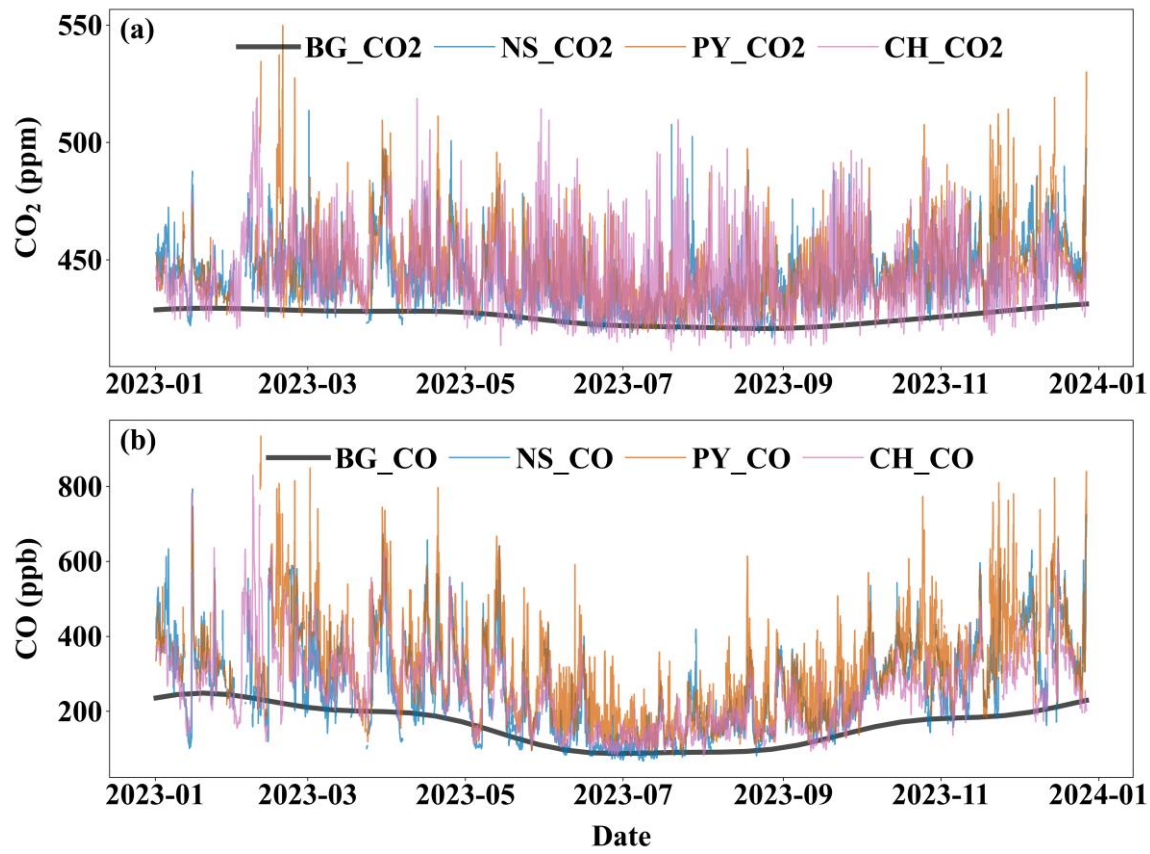


Figure S10. Time series of (a) CO₂ and (b) CO concentrations at each station compared with urban marine background levels.

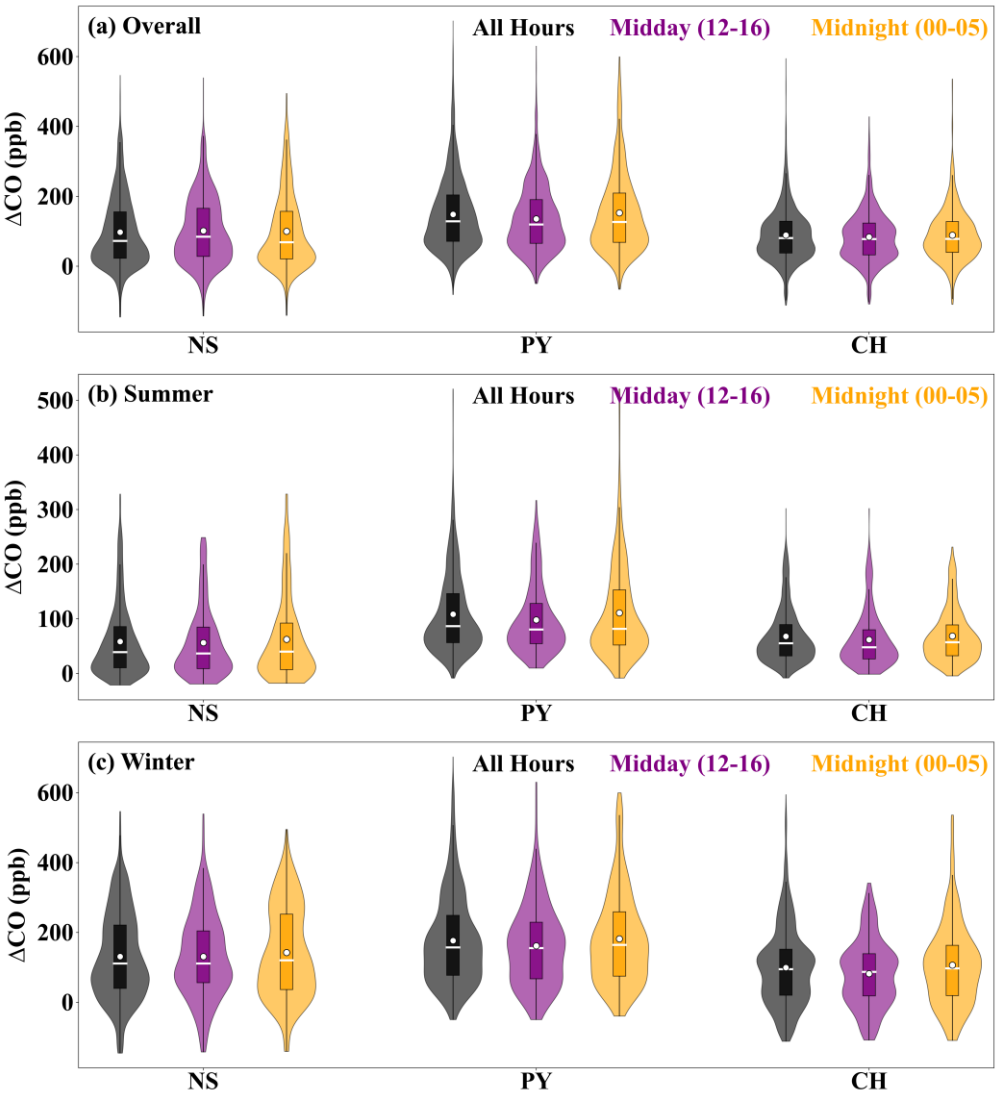


Figure S11. Hourly CO enhancement above the marine background level at each station during the (a) overall, (b) summer, and (c) winter periods. The white dots represent the mean values.

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