Characterization of Regional Combustion Efficiency using ΔXCO: ΔXCO₂ Observed by a Portable Fourier-Transform Spectrometer at an Urban Site in Beijing※

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ABSTRACT

Measurements of column-averaged dry-air mole fractions of carbon dioxide and carbon monoxide, CO₂ (XCO₂) and CO (XCO), were performed throughout 2019 at an urban site in Beijing using a compact Fourier Transform Spectrometer (FTS) EM27/SUN. This data set is used to assess the characteristics of combustion-related CO₂ emissions of urban Beijing by analyzing the correlated daily anomalies of XCO and XCO₂ (e.g., ΔXCO and ΔXCO₂). The EM27/SUN measurements were calibrated to a 125HR-FTS at the Xianghe station by an extra EM27/SUN instrument transferred between two sites. The ratio of ΔXCO over ΔXCO₂ (ΔXCO:ΔXCO₂) is used to estimate the combustion efficiency in the Beijing region. A high correlation coefficient (0.86) between ΔXCO and ΔXCO₂ is observed. The CO:CO₂ emission ratio estimated from inventories is higher than the observed ΔXCO:ΔXCO₂ (10.46 ± 0.11 ppb ppm⁻¹) by 42.54%–101.15%, indicating an underestimation in combustion efficiency in the inventories. Daily ΔXCO:ΔXCO₂ are influenced by transportation governed by weather conditions, except for days in summer when the correlation is low due to the terrestrial biotic activity. By convolving the column footprint [ppm (μmol m⁻² s⁻¹)] generated by the Weather Research and Forecasting-X-Stochastic Time-Inverted Lagrangian Transport models (WRF-X-STILT) with two fossil-fuel emission inventories (the Multi-resolution Emission Inventory for China (MEIC) and the Peking University (PKU) inventory), the observed enhancements of CO₂ and CO were used to evaluate the regional emissions. The CO₂ emissions appear to be underestimated by 11% and 49% for the MEIC and PKU inventories, respectively, while CO emissions were overestimated by MEIC (30%) and PKU (35%) in the Beijing area.

Key words: FTS, remote sensing, ΔXCO:ΔXCO₂, combustion efficiency, megacity

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Article Highlights:
• Daytime column-averaged dry-air mole fractions of atmospheric CO₂ and CO are provided in urban Beijing based on a portable FTS since 2019.
• The CO₂:CO emission ratio estimated by MEIC and PKU is 42.54% and 101.15% higher than the observed ΔXCO:ΔXCO₂ (10.46 ppm⁻¹), indicating an underestimation of the combustion efficiency in inventories.
• The MEIC underestimates CO₂ emissions by about 11% and overestimates 30% CO emissions by 30%; PKU underestimates CO₂ emissions by 49% and overestimates CO emissions by 35%.

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1. Introduction

The increasing anthropogenic emissions of CO\(_2\) are the main driving force behind global warming induced by greenhouse gases (GHG) (Stocker et al., 2013). Due to increased energy consumption, global fossil CO\(_2\) emissions reached 9.4 ± 0.5 GtC yr\(^{-1}\) over the last decade (2007–17) (Le Quéré et al., 2020). The mitigation of anthropogenic GHG emissions is often accompanied by strengthened air pollution controls (West et al., 2013). The toxic air pollutant CO is an indirect greenhouse gas because it produces positive radiative forcing (0.2 W m\(^{-2}\)) through its oxidation reaction with hydroxyl radicals (OH) in the troposphere (Myhre et al., 2013). There was a declining trend of 2% yr\(^{-1}\) in CO emissions over East Asia during 2005–16, which mainly resulted from the active clean air policies implemented in China (Zheng et al., 2018a), the largest developing and energy-consuming nation in the world with a very high contribution from megacities.

Urban areas account for 2% of the world's land but are responsible for 40%–70% of the global anthropogenic CO\(_2\) emissions (Satterthwaite, 2008, 2010; Ye et al., 2020). Carbon monoxide (CO) in the urban atmosphere is a product of incomplete combustion alongside the formation of CO\(_2\) during household combustion, industrial activities, and motor transportation, which results in a general positive correlation between CO and CO\(_2\) in urban areas. Due to the relatively short lifetime for CO of only a few weeks, it can serve as an unique tracer for atmospheric fossil-fuel CO\(_2\) emissions and combustion efficiency by analyzing the ratio of CO:CO\(_2\). The estimates of combustion efficiency at the urban scale could be calculated from the CO\(_2\) and CO statistical emission inventories; such an approach is called the “bottom-up” method. Different inventories display significantly different CO\(_2\) and CO emissions in China, especially at regional and city scales (Zhao et al., 2012; Dai et al., 2020). Accurate measurements of GHG and pollutants in cities are essential to yield significant information about the regional carbon budget and to propose strategies to control these emissions (“top-down” method). The correlation between CO and CO\(_2\) could be assessed by directly calculating the slopes of observed CO and CO\(_2\) volume mixing ratios (Wang et al., 2010; Worden et al., 2012). However, the slopes of CO and CO\(_2\) are easily affected by annual background growth and biogenic sources. Thus, regression fits of excess CO (ΔCO) and excess CO\(_2\) (ΔCO\(_2\)) from continuous observations have been extensively studied to quantify the anthropogenic contribution to carbon variation and source combustion efficiency (Wunch et al., 2009; Silva et al., 2013; Popa et al., 2014; Feng et al., 2019a; Shan et al., 2019; Cai et al., 2021; Park et al., 2021). Megacities in China show a lower combustion efficiency with a higher ΔCO:ΔCO\(_2\) ratio than in developed countries (Silva et al., 2013; Silva and Arellano, 2017; Park et al., 2021). Studies in Hefei, China have found the Emissions Database for Global Atmospheric Research (EDGAR) and the Peking University (PKU) emission inventories overestimated ΔCO:ΔCO\(_2\) in central China during 2015–16 (Shan et al., 2019).

Beijing is located in the northern part of China (Fig. 1). It is in a representative fast-growing economic cycle and is a heavily populated and developed region. The air pollution control policies in Beijing are directly related to the country’s overall goals. The total fossil-fuel CO\(_2\) emissions of Beijing show high uncertainty with ranges from 1 to 8 Mt in 2012 according to different emission inventories and urban regions (~17% relative to the overall land area of Beijing) contributes about 64% CO\(_2\) emissions to the total emissions for Beijing (Han et al., 2020). Satellite observations found the XCO:XCO\(_2\) ratio in Beijing/Tianjin region to be one of the highest in the northern hemisphere due to its rapid economic development (Park et al., 2021). Anthropogenic emissions have been estimated to make a 75.2% contribution to the annual CO\(_2\) enhancement in Beijing in 2014 based on isotopic analysis (Niu et al., 2016). Long-term, highly accurate in situ measurements of CO and CO\(_2\) near the surface have been developed in rural and urban regions of Beijing. The Shangdianzi (SDZ) and Miyun sites are used for rural areas, and the Peking University site is used for urban areas (Han et al., 2009; Wang et al., 2010; Feng et al., 2019a; Dayalu et al., 2020). The ratio, ΔCO:ΔCO\(_2\), at the Miyun site shows a significant decrease during 2005 to 2008 (Wang et al., 2010). The observation of CO at the SDZ site shows a fast downward trend in 2006–08, is stable in 2009–13, then shows another continuous downward trend after 2013, suggestive of improved combustion efficiencies in response to the Air Pollution Prevention and Control Action Plan implemented in 2013 (Li et al., 2020). The short-term pollution reduction associated with the 2008 summer Olympics and 2020 COVID-19 lockdowns result in a decrease of ΔCO:ΔCO\(_2\), suggesting increases in combustion efficiency (Wang et al., 2010; Cai et al., 2021).

The “top-down” analysis of combustion efficiency in Beijing comes mostly from satellite and in situ observations. However, in situ surface monitoring measurements are greatly influenced by vertical mixing. The column-averaged dry-air mole fraction (\(X_{gas}\)) represents the vertically integrated concentration per dry air and is less affected by the vertical motion than in situ measurements. Therefore, the horizontal gradients of \(X_{gas}\) have a more direct relationship with the regional-scale flux (Yang et al., 2007; Wunch et al., 2011; Chen et al., 2016). Satellite observations of XCO\(_2\) combined with XCO show obvious superiority in global coverage and have been used to quantify the correlation between CO\(_2\) and CO for Beijing. However, satellite observation is limited by sampling size and temporal resolution to these specific megacities. Satellites make infrequent visits over cities and are easily affected by cloud, aerosol, and topography issues (Lei et al., 2021), leading to insufficient data for urban emissions studies. The Total Carbon Column Observing Network (TCCON) is an international Fourier-transform spectrometer (FTS) network that was originally used for satellite validation and mainly uses ground-based high-resolution Fourier-transform infrared (FTIR) measurements to record information.
about the GHG column. High-resolution FTIR stations in China have been built at Hefei, Xianghe, and Beijing (Wang et al., 2017; Bi et al., 2018; Yang et al., 2020b). Given the poor transportability of TCCON FTS stations, portable, low-cost FTS is useful as a component in long-term urban column measurements. The EM27/SUN instrument (hereafter, EM27) is a compact FTS with its own solar tracker, which is easy to set up and transport anywhere. FTS measurements in or near urban Beijing have mainly focused on satellite validation (Bi et al., 2018; Yang et al., 2020b). To date, few pub-
lished studies have rigorously quantified the combustion efficiency in the urban region of Beijing.

In this study, one year of XCO and XCO₂ measurements based on EM27 were collected at an urban site in Beijing. The objectives of this study were to (1) analyze the correlational relationship between observed enhancements of XCO and XCO₂ in urban Beijing, (2) reconcile the differences between top-down estimates of combustion efficiency with bottom-up estimates of combustion efficiency, and (3) evaluate the regional CO₂ and CO emissions estimated from bottom-up inventories using remote sensing data set. The remainder of this paper is organized as follows. A description of the instrument and method is given in section 2. Section 3 presents the results and discusses their implications and section 4 concludes.

2. Data and methods

2.1. Instrumentation

An EM27 has been set up on the roof of the Institute of Atmospheric Physics (IAP) building in Beijing since 2019. The IAP observation site (highlighted in Fig. 1b) is located between the north 3rd and 4th ring road, with heavy traffic and high CO₂ and CO emissions. The EM27 spectral resolution is less than 0.5 cm⁻¹, corresponding to a maximum optical path difference of 1.8 cm. The compound InGaAs (indium gallium arsenide) and an extended InGaAs detector are used to stably detect 3800–14 000 cm⁻¹ spectral information for accurate column concentrations of CO₂ and CO (Gisi et al., 2012; Hase et al., 2016). It takes about 60 s to record one individual spectrum with 10 interferograms averaged. The EM27 is equipped with an automated protective case with a rain sensor and a solar irradiation sensor. The cover only opens under cloudless daytime conditions (0000–0900 UTC), protecting the instrument from rain or snow and achieving automated EM27 observations. A WS500 weather station located immediately next to EM27 was used to measure surface temperature and surface pressure with accuracies of 0.2°C and 0.5 hPa, respectively, which were used to correct the a priori temperature profile needed for the XCO₂ and XCO retrievals. The Bruker Optical™ Infrared Fourier Transform Spectrometer (IFS) 125 HR used by TCCON is a high-resolution spectrometer (< 0.02 cm⁻¹) that is tied to the World Meteorological Organization (WMO) scale through numerous aircraft campaigns (Wunch et al., 2010). To ensure data quality, the EM27 has been calibrated in side-by-side experiments with 125 HR (Gisi et al., 2012; Klappenbach et al., 2015; Hedelius et al., 2017; Frey et al., 2019). High-resolution FTIR stations have been built near Beijing and are located in Xianghe, 50 km east-southeast of Beijing (highlighted in Fig. 1a) (Yang et al., 2020b). The 125 HR at the Xianghe site complies with the TCCON specifications and is used as a calibration reference for the EM27 at Beijing in this study.

A portable EM27 could act as a transfer standard between two FTIR stations (Jacobs et al., 2020). In this study, another EM27 (EM27#2) was used as a mobile transfer standard instrument for 125 HR in Xianghe and the EM27 in Beijing. The EM27#2 took measurements side-by-side with IFS 125 HR during the entire month of November 2019. To ensure stability during the shipment of EM27#2 across Beijing to Xianghe, we examined the ratio between EM27s before and after the shipment. Figures 2a and 2b show the biases between 125 HR and EM27#2. The correction equations are also shown in the figure. Figures 2c and 2d show that the ratio was consistent before and after the shipments (detailed in section 2.2), indicating that the calibration of EM27#2 and 125 HR in Xianghe could be applied to the EM27 retrieval in Beijing. The IFS 125 HR retrievals apply the TCCON standard retrieval code (GGG2014), and the EM27 uses PROFFAST, with the same meteorological data and preliminary profiles input. The results for EM27#2 show a systematic bias with 125 HR, which could be corrected by applying a linear fitting correction function (Figs. 2a and 2b). The biases between EM27 and 125HR are 0.28% and 5.3% for XCO₂ and XCO, respectively.

2.2. EM27 data processing

A non-linear least-squares fitting retrieval algorithm (PROFFAST) (Hase et al., 2004) is used to analyze the spectra recorded by EM27. The algorithm has been officially used in the EM27 network, COCCON (the COllaborative Carbon Column Observing Network) (Frey et al., 2019). To obtain a high-quality spectrum, the pre-processing steps involve a direct current (DC) correction (Keppel-Aleks et al., 2007), a Norton–Beer medium apodization (Naylor and Tahic, 2007), a fast Fourier transform (Bergland, 1969), and a phase correction (Mertz, 1967). The a priori profiles obtained by TCCON are the official a priori profiles (Toon and Wunch, 2015). The inverse algorithm obtains the scaled target gas column by iteratively updating the state variables to best fit the simulated and measured spectra. Finally, the gas column (C_gas) is converted to a dry-air column-averaged mole fraction (denoted X_gas) by dividing the gas column by the simultaneously retrieved column-averaged dry-air mole fractions (C_air) Columnair, where C_air could be calculated from the O₂ column combined with the volume fraction of O₂ in the air (20.95%):

$$X_{gas} = \frac{C_{gas}}{C_{air}} . \tag{1}$$

In the post-processing process, column averaging kernels (AK) describe the altitude-dependent sensitivity of the retrieved state to the true state. As shown in Fig. 3, AK differs among instruments with different spectral resolutions. EM27 is more sensitive to changes at lower levels than 125 HR, especially for high solar zenith angles (SZAs). A pressure weighting function (PW) is used to weight the pressure thicknesses of each level (ΔP) relative to the surface pressure (P_surf):

$$PW = \frac{\Delta P}{P_{surf}} . \tag{2}$$
A Lagrangian particle dispersion model (LPDM) models a plume of atmospheric tracers from a cluster of particles. It can determine the footprint (also called an influence function or adjoint sensitivity) of multiple air parcels released from an observation site (receptor). The WRF-XSTILT, an LPDM model, is adopted to analyze the meteorologically-induced gas concentration variation by a regional column footprint simulation (Lin et al., 2003; Fasoli et al., 2018; Wu et al., 2018). The footprint is used to establish the source–receptor relationship and determine the distribution spread of entire plumes in adjacent cells over the receptor by calculating the sensitivity of the surrounding region (source) to the receptor. The column footprint \( \chi_{\text{foot}}(x_r, t_r; x_i, y_i, t_m) \) is essential for tracking the air masses for column measurements. The parameter \( \chi_{\text{foot}} \) is the sensitivity of the column measurements to the upstream and downstream surface-atmosphere fluxes. The formula to calculate \( \chi_{\text{foot}} \), with units of ppm \((\mu\text{mol} \ (\text{m}^2 \ \text{s})^{-1})^{-1}\), for each receptor is as follows:

\[
\chi_{\text{foot}}(x_r, t_r; x_i, y_i, t_m) = \frac{m_{\text{air}}}{h \rho(x_i, y_i, t_m)} \times \\
\frac{1}{N} \sum_{i=1}^{N} \Delta t_i \chi_{\text{foot}}(x_i, y_i, t_m) \Delta K(r) \Delta P(w),
\]

where \((x_r, t_r)\) is the receptor \((r)\) location, \((x_i, y_i, t_m)\) is the model’s initial time set, denoted by the model grid coordinates of location and time, \(m_{\text{air}}\) is the mean molar mass of dry air \((29 \ \text{g mol}^{-1})\), \(h\) is the atmospheric column height (in XSTILT \(h\) is set at half the planetary boundary layer (PBL) depth), \(\rho\) is the mean density of the air below \(h\), \(N\) is the total number of released particles, \(\Delta t_i\) is the residence time of particle \(i\) spent in the grid cell \((x_i, y_i, t_m)\). The total column footprint \(\chi_{\text{foot}}\) is the integrated footprint from different vertical altitudes convolved with a pressure weighting function \(\Delta P(w)\) and averaging kernel \(AK(r)\) at the receptor, which links the emission sensitivity to the observations. In the model setup, we applied the WRF model configurations verified by Dayalu et al. (2020) for the same study domains (Fig. 1a). The receptor site of X-STILT was set up at IAP.
and was run backward for seven days at a \(0.25^\circ \times 0.25^\circ\) spatial resolution to generate hourly column footprints. In each simulation, 100 air parcels are released every 100 m within 3 km and every 500 m from 3 to 6 km relative to the observation level, which tends to be denser near the surface. Higher altitudes are not used since only the lower atmosphere interacts with local emissions in the region (Hedelius et al., 2018). The AK and PW (detailed in section 2.2) of EM27 are used to weight and integrate the footprints of all levels to the column footprint and could be directly obtained from the output of the EM27 retrieval.

### 2.4. The \(\Delta XCO: \Delta XCO_2\) calculation method

#### 2.4.1. Calculation of \(\Delta XCO\) and \(\Delta XCO_2\) from measurements

The observed regression slope of \(\Delta XCO\) against \(\Delta XCO_2\) denotes the amount of CO per CO\(_2\) emissions in the atmosphere that has been captured by the FTS, signifying the emission ratio or combustion efficiency. A larger slope of the regression line reveals greater combustion efficiency in the observed XCO\(_2\). Wunch et al. (2009) proposed that the \(X_{gas}\) anomaly (\(\Delta X_{gas}\)) could be calculated by subtracting \(X_{gas}\) in the morning from the afternoon value at the same SZA. The anomalies are divided by the AK at the surface to account for the sensitivity of the column measurement to surface variations because diurnal changes are assumed to be confined to the boundary layer. This method could minimize the atmospheric effects from background or natural sources and the SZA-dependent error. The \(\Delta XCO:\Delta XCO_2\) ratio, calculated from the diurnal enhancement, is denoted as \((\Delta XCO: \Delta XCO_2)_d\).

Apart from the method based on diurnal variation,
$\Delta XCO$ and $\Delta XCO_2$ could be estimated from the regional change relative to the background. The observation site at urban Beijing records the pollution-related signal of XCO and XCO$_2$. The quantification of background XCO$_2$ concentration is the sum of the boundary and biogenic XCO$_2$ values.

The boundary XCO$_2$ value was obtained from trajectory endpoint of the Copernicus Atmosphere Monitoring Service (CAMS) satellite-derived global optimization mean column concentrations data (http://atmosphere.copernicus.eu, version: v20r3, last access: 7 June 2021) (Chevallier et al., 2005; Chevallier et al., 2010; Chevallier et al., 2019). These trajectories were derived from the Weather Research and Forecasting-Column Stochastic Time Inverted Lagrangian Transport (WRF-XSTILT). According to the validation report for the CAMS v1r1 data by Chevallier et al. (2020), the biases of modeled XCO$_2$ data show less bias (less than 1 ppm) in a non-urban area when compared to the TCCON retrieval results. The background XCO$_2$ values obtained from CAMS were mostly from the region less affected by human activities, so the CAMS background values were considered to be plausible. Biogenic XCO$_2$ values were from the convolution of the column footprint generated by WRF-XSTILT and the CO$_2$ vegetation flux. The CO$_2$ vegetation flux comes from a global gridded terrestrial carbon flux product developed by Zeng et al. (2020). Due to unavailable XCO background data, XCO$_2$ background values were simply obtained from the lowest observed XCO. Similar to the diurnal method described above, the AK at the surface was also taken into account in calculating the anomalies. The ratio $\Delta XCO: \Delta XCO_2$ calculated from regional enhancement is denoted as $(\Delta XCO: \Delta XCO_2)_r$.

2.4.2. Calculation of $\Delta XCO$ and $\Delta XCO_2$ from bottom-up inventories

The simulated regression slope of $\Delta XCO$ against $\Delta XCO_2$ represents the regional XCO enhancement per XCO$_2$ enhancement contributed from different sources transported to the observation site. A simple way to simulate $\Delta XCO: \Delta XCO_2$ directly from the emission inventories in a specific geometrical shape is as follows (Wunch et al., 2009; Hu et al., 2019; Shan et al., 2019)

$$\left( \frac{\Delta XCO}{\Delta XCO_2} \right)_{\text{sim}} = \frac{E_{CO}}{E_{CO_2}} \times \frac{M_{CO}}{M_{CO_2}}.$$  \hspace{1cm} (4)

Here, $E_{CO}$ and $E_{CO_2}$ are the CO and CO$_2$ emissions in the specific area, $M_{CO}$ and $M_{CO_2}$ are the molecular masses of CO and CO$_2$, and $(\Delta XCO/\Delta XCO_2)_{\text{sim}}$ is the simulated correlation slope of XCO to XCO$_2$.

The parameters $\Delta XCO$ and $\Delta XCO_2$ can be simulated by the convolution of the source footprint function (modeled by the Lagrangian atmospheric transport model) and the anthropogenic flux inventories defined as follows:

$$\left( \frac{\Delta XCO}{\Delta XCO_2} \right)_{\text{sim}} = \frac{X\text{foot,co} \times E_{CO}}{X\text{foot,co}_2 \times E_{CO_2}},$$  \hspace{1cm} (5)

where the simulated $\Delta XCO$ and $\Delta XCO_2$ are the modeled hourly anthropogenic enhancement from different sources. The $E_{CO_2}(E_{CO})$ is and represented by the CO$_2$ (CO) surface anthropogenic emissions. The footprint quantifies the sensitivity of the measurements to emissions in terms of unit concentration per unit flux. Simulated $\Delta XCO$ and $\Delta XCO_2$ values based on this method are divided by AK at the surface to account for the sensitivities of column measurements near the surface. We focused on simulations from 1100–1600 local standard time (LST, UTC + 8 h) consistent with the time that the atmosphere is well-mixed and the depth of planetary boundary layer (PBL) grow to approach its maximum in the late morning due to the solar heating of the surface. The PBL, in turn, collapses at sunset due to surface cooling, which increases the difficulty of simulation. We selected this period so that the footprint generated by X-STILT would be less susceptible to modeled PBL uncertainties (Sargent et al., 2018).

$E_{CO_2}(E_{CO})$ is obtained from monthly gridded fossil-fuel emission databases. Fossil-fuel emission products come from Peking University (PKU) and the Multi-resolution Emission Inventory for China (MEIC). The PKU product provides global CO and CO$_2$ grid emission maps up to 2014 with 0.1° spatial resolution according to a sub-national disaggregation of fuel data (Wang et al., 2013). The MEIC product provides China’s CO and CO$_2$ grid emission maps up to 2017 with 0.25° spatial resolution, including the local emission information for each power plant and industrial operation (Zheng et al., 2020). The Temporal Improvements for Modeling Emissions by Scaling (TIMES) developed by Nassar et al. (2013) is applied to the MEIC and PKU products to derive diurnal CO$_2$ emissions. Due to unavailable temporal variations for CO, we simply averaged the monthly CO emissions and assigned them to each hour. The MEIC and PKU products provide gridded regional emissions up to 2017 and 2014, respectively. Owing to the lack of an emission trend in Beijing, we apply the inter-annual variation of CO$_2$ emissions in China until 2019, as concluded from Crippa et al. (2020). Due to the unavailability of CO emissions up to 2019, a mean decreasing rate in East Asia of 2% was applied, estimated from the top-down inversion method based on MOPITT data from 2005 to 2016 (Zheng et al., 2018a).

3. Results and discussion

3.1. Time series of XCO and XCO$_2$

The monthly means and standard deviation of XCO$_2$ and XCO at mid-latitude stations in the northern hemisphere are displayed in Fig. 4. These include FTIR stations in Beijing, Pasadena (34.14°N) (Wennberg et al., 2017), Xianghe (39.75°N) (Yang et al., 2020b), Karlsruhe (49.10°N) (Hase et al., 2017), Tsukuba (36.05°N) (Morrino et al., 2018), and Paris (48.97°N) (Té et al., 2017). Table 1 summarizes the sea-
sonal mean and standard deviation of XCO\textsubscript{2} and XCO. There is no data recorded in February, which may cause a bias because anthropogenic XCO\textsubscript{2} enhancements during the Spring Festival should be significantly lower than normal. The XCO\textsubscript{2} values in Beijing ranged from 402 ppm to 423 ppm in 2019. The seasonal variation in XCO\textsubscript{2} achieves its peak in winter (414.33 ± 2.65 ppm), followed by spring (413.58 ± 1.25 ppm) and autumn (412.09 ± 2.88 ppm), and is lowest in summer (407.87 ± 3.34 ppm). The features of the XCO\textsubscript{2} seasonal variation in Beijing are similar to observations from other FTIR observation sites in the mid-latitude northern hemisphere. Intensity of photosynthesis, which is related to the latitude of the observation site, is the main reason for the seasonality in XCO\textsubscript{2} variation. The peak and trough of monthly XCO\textsubscript{2} means were found in December (415.72 ± 3.18 ppm) and August (404.87 ± 1.47 ppm), respectively, which differ slightly from other FTIR stations. The monthly XCO\textsubscript{2} in Paris, Park Falls, Karlsruhe, and Xianghe stations reach a peak in March to April, whereas Pasadena stations reach a peak in December. The monthly XCO\textsubscript{2} of Beijing, Xianghe, Park Falls, Karlsruhe reach a trough in August, whereas Pasadena reaches a trough in September.

Table 1. Seasonal variability of average XCO\textsubscript{2} and XCO in 2019 observed by EM27.

| Season                  | XCO\textsubscript{2} (ppm) | XCO (ppb)   |
|-------------------------|-----------------------------|-------------|
| Spring (March to May)   | 413.58 ± 1.25               | 109.58 ± 5.49|
| Summer (June to August) | 407.87 ± 3.34               | 120.50 ± 4.28|
| Autumn                  | 412.09 ± 2.88               | 112.37 ± 5.58|
| Winter (December to February) | 414.33 ± 2.65 | 115.78 ± 3.36|

Values of XCO ranging from 85 to 192 ppb are observed in Beijing. The XCO in Beijing was higher than the values from other cities in the northern hemisphere and

Fig. 4. Monthly variations in (a) XCO\textsubscript{2} and (b) XCO observed at Beijing, Xianghe, Karlsruhe, Pasadena, Tsukuba, and Paris during 2019. The geographical coordinates of each site are shown in the figure legend. The error bars are the monthly standard deviation of XCO\textsubscript{2} and XCO.
lower than that of the Xianghe station (Fig. 4b), implying the presence of high regional emissions in the Beijing–Xianghe region. As can be concluded from Table 1, XCO is highest in summer (120.50 ± 4.28 ppb), followed by winter (115.78 ± 3.36 ppb) and autumn (112.37 ± 5.58 ppb), and is lowest in spring (109.58 ± 5.49 ppb). The seasonal variation in Beijing is similar to the variation in Xianghe. The increase of CO during winter in Beijing could be from the increased heating time of vehicle catalysts at low temperatures (Han et al., 2009) and domestic heating. The seasonal variations in Karlsruhe, Pasadena, and Paris show XCO values that are highest in spring and lowest in summer and autumn. The XCO in summer for the Beijing-Xianghe region presents a seasonal variation opposite of the other three urban sites. Carbon monoxide (CO) concentration was also found to be lowest in summer from 2006 to 2018 at the Shangdianzi (SDZ) in situ site, a rural region near Beijing (Li et al., 2020). This result is consistent with the Karlsruhe, Pasadena, and Paris sites. The high XCO values in summer observed in urban Beijing and Xianghe could be associated with strong anthropogenic emissions.

3.2. ΔXCO: ΔXCO2 correlation observed by the FTS

Carbon monoxide (CO) is co-emitted and co-transported to the observation site with CO2. The slopes of ΔXCO to ΔXCO2 reflect the overall combustion efficiency of the observed airmass. Figure 5 shows the daily regression slope and the Pearson correlation coefficients (R2) of ΔXCO and ΔXCO2 based on the diurnal variation method. Daily regression slopes are mostly around 10 ppb ppm–1. Daily correlation coefficients are generally larger than 0.5 on 78% of the observation days. Significant positive correlations between ΔXCO to ΔXCO2 in winter, spring, and autumn indicate that most air parcels originate from combustion sources. Approximately 61% of the daily correlation coefficients in summer (grey shaded region in Fig. 5) are small (< 0.1) and even negative, and the regression slope shows large uncertainties. This suggests that CO2 emissions are dominated by non-CO-related sources in summer. In situ observations near Beijing also captured the low correlation in summer (Wang et al., 2010). The main reason is that the CO2 signals were significantly mixed with enhanced biospheric CO2 uptake during the growing season, which could offset the anthropogenic emissions in urban areas. It is necessary for CO and CO2 to
share a common combustion process when using CO as the anthropogenic tracer for CO$_2$ to investigate regional combustion efficiency. One straightforward approach is to remove the non-CO-related observations (R$^2 \leq 0.1$). Before filtering the data, $\Delta$XCO:$\Delta$CO$_2$$_{d}$ observed for urban Beijing is 8.94 ± 0.13 ppb ppm$^{-1}$ with an R$^2$ of 0.80. Removal of the strong biologically-affected days contributed to the elevated ($\Delta$XCO:$\Delta$CO$_2$)$_{d}$ of 10.46 ± 0.11 ppb ppm$^{-1}$ and an R$^2$ of 0.86.

We extended our analysis to comparisons of ($\Delta$XCO:$\Delta$CO$_2$)$_{d}$ based on other FTS station datasets of the mid-latitude northern hemisphere in and around 2019 (Table 2). The FTS stations are set up in either an urban or suburban environment with varied emissions sources: the Pasadena site is located on the southern coastal air basin of California with a population of nearly 17 million, the Tsukuba site is located in a highly urbanized city near Tokyo with a population of 2.28 million, the Paris site is in the most populous city in France (2.24 million), the Karlsruhe site is a smaller urban region surrounded by forest, which has a population of nearly 0.3 million. Stronger correlations between $\Delta$XCO and $\Delta$CO$_2$ exist with high regression slope values in the densely populated urban regions (Beijing, Pasadena, Tsukuba, Paris, and Karlsruhe). The values of ($\Delta$XCO:$\Delta$CO$_2$)$_{d}$ observed in Chinese cities (10.46 ppb ppm$^{-1}$ in Beijing, 6.76 ppb ppm$^{-1}$ in Xianghe) are significantly higher than stations in other nations. In addition, ($\Delta$XCO:$\Delta$CO$_2$)$_{d}$ in Beijing is the highest among all the cities we included, implying a high pollution per amount of CO$_2$ emissions and relatively lower combustion efficiency over Beijing.

According to the in situ observations, Wang et al. (2010) found a $\Delta$CO:$\Delta$CO$_2$ value of 41.7 ppb ppm$^{-1}$ at the Miyun background site in Beijing from 2007 to 2008. Han et al. (2009) derived a $\Delta$CO:$\Delta$CO$_2$ value of 43.4 ppb ppm$^{-1}$ during the 2006 winter at an urban site in Beijing. Using XCO retrievals from NASA/Terra Measurement of Pollution in the Troposphere (MOPITT) and XCO retrievals from the Japan Aerospace Exploration Agency Greenhouse gases Observing Satellite (GOSAT), Silva et al. (2013) estimated $\Delta$XCO:$\Delta$CO$_2$ in Beijing/Tianjin to be 43.5 ppb ppm$^{-1}$ from 2009 to 2010. These values are significantly larger than our estimated value (10.46 ppb ppm$^{-1}$). The reason could be that China has implemented pollution control policies since 2013. As a result, the combustion efficiency significantly increased as CO emissions decreased (Zheng et al., 2018b; Feng et al., 2019b; Li et al., 2020). Shan et al. (2019) estimated $\Delta$CO:$\Delta$CO$_2$ to be 15.99, 8.02, and 5.67 ppb ppm$^{-1}$ based on in situ, ground-based FTS, and satellite measurements (GOSAT, MOPITT), respectively, at Hefei from 2015 to 2016. Li et al. (2020) estimated $\Delta$CO:$\Delta$CO$_2$ to be 25.5 ppb ppm$^{-1}$ at the SDZ site near Beijing in 2018. The $\Delta$XCO:$\Delta$CO$_2$ value we calculated (10.46 ppb ppm$^{-1}$) is close to the value estimated from ground-based FTS at Hefei in 2015−16 (8.02 ppb ppm$^{-1}$) and is about 60% lower than the near-surface observed value at SDZ (25.5 ppb ppm$^{-1}$). The value of $\Delta$CO:$\Delta$CO$_2$ based on FTS (8.02 ppb ppm$^{-1}$) at Hefei is about 50% lower than in situ observations (15.99 ppb ppm$^{-1}$) (Shan et al., 2019). In situ observations only capture the near-surface signal of a small sampling area in the local planetary boundary layer. In contrast, the FTS observations detect the whole layer of the atmosphere, which may weaken the near-surface signal. The FTS observations have larger footprint compared to in situ observations and could be more representative to the regional flux (Wunch et al., 2016).

### 3.3. Effect of regional transportation on $\Delta$XCO:$\Delta$CO$_2$

Transportation governed by weather conditions plays an important role in the day-to-day variations in CO and CO$_2$ in Beijing (Feng et al., 2019a; Panagi et al., 2020). Air pollution is concentrated in Beijing’s southern and eastern parts (Feng et al., 2019a). We identified source regions for each observation based on X-STILT footprints. Pathways are characterized by sources in the northwest (NW) and north China plain (NCP) according to the year-round average 24-hour backward footprints (Figs. 6c and 6e), which share 62.93% and 26.72% of the observation days, respectively. Higher XCO$_2$ and XCO occurred when air masses originated from the NCP region. Clean air masses originating from the NW are less affected by human activities, which may cause the observed decrease in XCO$_2$ and XCO.

When an air mass passes over different source regions, the correlation between CO and CO$_2$ shows different patterns. As shown in Figs. 6b and 6d, ($\Delta$XCO:$\Delta$CO$_2$)$_{d}$ originating from the clean region is 8.23 ± 0.1 ppb ppm$^{-1}$ and from the polluted region is 11.46 ± 0.2 ppb ppm$^{-1}$. Advection that brings air masses containing emissions from the NCP contributed to an elevated proportion of ($\Delta$XCO:$\Delta$CO$_2$)$_{d}$.

| FTS station (longitude, latitude) | Time period           | $\Delta$XCO:$\Delta$CO$_2$ (ppb ppm$^{-1}$) | R$^2$   |
|----------------------------------|-----------------------|---------------------------------------------|--------|
| Beijing, CHN (39.98°N, 116.39°E) | 2019.1−2019.12       | 10.46 ± 0.11                                | 0.86   |
| Xianghe, CHN (39.75°N, 116.96°E) | 2018.7−2019.7         | 6.76 ± 0.70                                 | 0.52   |
| Karlsruhe, DE (49.10°N, 8.44°E)  | 2018.1−2019.12        | 1.84 ± 0.21                                 | 0.52   |
| Pasadena, US (34.14°N, 118.13°E)| 2019.1−2019.12        | 4.06 ± 0.18                                 | 0.61   |
| Tsukuba, JP (36.05°N, 140.12°E) | 2018.1−2019.9         | 4.68 ± 0.22                                 | 0.58   |
| Paris, FR (48.97°N, 2.37°E)     | 2019.1−2019.12        | 3.06 ± 0.06                                 | 0.76   |
which exceeded the annual slope (10.46 ± 0.11 ppb ppm$^{-1}$).

### 3.4. Comparison of the observed and modeled regional XCO$_2$ and XCO enhancement

The regression slope of ΔXCO:ΔXCO$_2$, based on the regional enhancement method (ΔXCO:ΔXCO$_2$)$_r$, is estimated to be 9.06 ± 1.89 ppb ppm$^{-1}$, which is consistent with the value of (ΔXCO:ΔXCO$_2$)$_d$ (10.46 ± 0.11 ppb ppm$^{-1}$). The ratio (ΔXCO:ΔXCO$_2$)$_d$ is calculated solely based on the diurnal variation observed by EM27. In contrast, (ΔXCO:ΔXCO$_2$)$_r$ is estimated by subtracting the background value from the observations. Uncertainty in background value yields more uncertainty for (ΔXCO:ΔXCO$_2$)$_r$, compared to (ΔXCO:ΔXCO$_2$)$_d$. 

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**Fig. 6.** (a) Correlations of ΔXCO and ΔXCO$_2$ in 2019. (b, d) Correlations of ΔXCO and ΔXCO$_2$ in 2019 originating from the NW and NCP upwind sources. $R^2$ is the correlation coefficient of ΔXCO and ΔXCO$_2$. $\text{slope}_{2019}$ is the regression slope of ΔXCO and ΔXCO$_2$. (c, e) Maps of mean 24-hour backward footprint [ppm / (μmol m$^{-2}$ s$^{-1}$), lg(x)] with 0.25° resolution at IAP, Beijing, starting at 1200 LST in 2019, originating from the NW (c) and NCP (e). The closed blue line indicates the BTH area. Only footprint values larger 10$^{-2}$ ppm / (μmol m$^{-2}$ s$^{-1}$) are displayed.
The estimated values for $\Delta XCO$ and $\Delta XCO_2$ from regional enhancement versus the background were used for comparison with the model simulation at hourly timescales (Figs. 7 and 8). The modeled $\Delta XCO_2$ (hereafter $\Delta XCO_{2,\text{sim}}$) was simulated from the MEIC and PKU anthropogenic emission inventories ($\Delta XCO_{2,\text{MEIC}}$ and $\Delta XCO_{2,\text{PKU}}$). The observed enhancement $\Delta XCO_2$ (hereafter $\Delta XCO_{2,\text{obs}}$) was the difference between the observed urban $XCO_2$ ($XCO_{2,\text{obs}}$) and the $XCO_2$ background ($XCO_{2,\text{back}}$) from model. Summer data is excluded due to the unavailable background data for CO (detailed in the next paragraph). The same trend is shared by $XCO_{2,\text{back}}$ and $XCO_{2,\text{obs}}$ as shown in Fig. 7a.

Fig. 7. (a) Measured $XCO_2$ and CAMS background concentration. (Only 1100 to 1600 LST periods are displayed). (b–c) Hourly measured and modeled regional enhancement $\Delta XCO_2$ for each fossil-fuel emission inventory (b for MEIC, c for PKU).

Fig. 8. Hourly measured and modeled $XCO$ for (a) MEIC and (b) PKU.
Figures 7b and 7c show the correlation between \( \Delta XCO_{obs} \) and \( \Delta XCO_{sim} \). The x-intercept of the linear fitting equation of \((-1 \text{ ppm for MEIC and PKU})\) represents the \( \Delta XCO_{obs} \) value with no anthropogenic effect. The bias of \( \Delta XCO_{obs} \) and \( \Delta XCO_{sim} \) was mainly attributed to the error from emission inventories, background \( XCO \) values, and transport simulation. Both the observed and modeled \( \Delta XCO \) are in good agreement, with correlation coefficients \( (R^2) \) of 0.70 and 0.73 for MEIC and PKU, respectively. The slope of the fitting equation denotes the ratio of the observed \( \Delta XCO \) change to the modeled \( \Delta XCO \) change. Many previous studies attempted to use the slope value as a scale factor to evaluate and constrain regional \( CO \) emissions \((\text{Sargent et al., 2018; Shekhar et al., 2020; Yang et al., 2020a})\). The slope for MEIC \((0.89 \text{ ppm ppm}^{-1})\) is closer to the 1:1 line than PKU \((0.61 \text{ ppm ppm}^{-1})\). According to the regression slope value, MEIC underestimates approximately 11\% of \( CO \) emissions with emission inventories \((\text{Turnbull et al., 2011; Tohjima et al., 2014})\). Each footprint is convolved with the corresponding hourly gridded emission inventories \((PKU, MEIC)\). The modeled anthropogenic enhancement of \( CO \) at the receptor site is the sum of contributions from the sensitive emission grid flux \((\text{detailed in section 2.3})\). The linear regression slopes of the modeled \( \Delta XCO: \Delta XCO_2 \) based on PKU, MEIC, and observations are shown in Table 3. The outliers are excluded according to the three standard deviations criterion. Modeled \( \Delta XCO \) shows a good relationship with \( \Delta XCO_2 \) \((R^2 = 0.97 \text{ for MEIC and 0.96 for PKU})\). The modeled data displays a slightly greater correlation than the observed \( \Delta XCO \) and \( \Delta XCO_2 \) \((R^2 = 0.86 \text{ for the diurnal variation method, } R^2 = 0.83 \text{ for the regional enhancement method})\). The reason is that modeled values only take the anthropogenic influence of \( CO \) into account, ignoring the \( CO \)-related but not the \( CO \)-related signal, such as the resident respiration of \( CO_2 \). The observed \( \Delta XCO \) and \( \Delta XCO_2 \) values based on regional background enhancement display the weakest correlation \((R^2 = 0.83)\) due to the uncertainty of the modeled background value. The simulated regression slope of \( \Delta XCO \) and \( \Delta XCO_2 \) in 2019 is 14.91 ± 0.36 ppm ppm\(^{-1}\) for MEIC and 21.04 ± 0.70 ppm ppm\(^{-1}\) for PKU. The MEIC and PKU inventories are 42.54\% and 101.15\% higher than the observed value \((10.46 ± 0.11 \text{ ppm ppm}^{-1})\), respectively.

In recent studies, \( \Delta XCO: \Delta XCO_2 \) based on EDGAR \((PKU)\) emission inventories are about 256.59\% (219.39\%) larger than the values calculated from the FTS in Hefei 2015–16, and 207.86\% (173.31\%) larger than those in 2016–17 \((\text{Shan et al., 2019})\). Silva and Arellano \((2017)\) found that \( \Delta XCO: \Delta XCO_2 \) based on the EDGAR inventory was 50\% higher than the value estimated by satellites in the megacities of China. Wang et al. \((2010)\) found that the bottom-up estimate of \( \Delta XCO: \Delta XCO_2 \) was 19.2\% larger than the

3.5. Comparison the observed and simulated \( \Delta XCO: \Delta XCO_2 \)

Many studies have compared the observed \( \Delta CO: \Delta CO_2 \) with emission inventories \((\text{Turnbull et al., 2011; Tohjima et al., 2014; Shan et al., 2019})\). To calculate the simulated \( \Delta CO: \Delta CO_2 \) with emission inventories on a regional scale, it is essential to know that the observational site is representative of the region. A few studies roughly specified a geometric bounding box outlining the region which influences the observed value \((\text{Wunch et al., 2009; Hu et al., 2019; Shan et al., 2019})\). However, \( \Delta CO: \Delta CO_2 \) calculated based on the geometric bounding method is sensitive to the specified size of the enclosed area. A circle centered upon the EM27 observation site was specified as the influencing region. As the source area radius ranges from 50 to 500 km, \( \Delta CO: \Delta CO_2 \) varies from 17.00 to 19.77 ppm ppm\(^{-1}\) for MEIC, 32.28 to 53.74 ppm ppm\(^{-1}\) for PKU, respectively. The results show great uncertainty among different inventories and influencing areas. This method is based on the assumption that each grid in the specific geometric region of the emission map contributes equally to the observed concentration.

The region of influence and the sensitivity of each influencing grid to the observations vary over time. Using surface hourly backward column footprints for each measurement is a common and robust way to quantify the sensitivity of the atmospheric concentration changes at the receptor to upwind source regions using units of concentration per unit flux \((\text{Turnbull et al., 2011; Tohjima et al., 2014})\). Each footprint is convolved with the corresponding hourly gridded emission inventories \((PKU, MEIC)\). The modeled anthropogenic enhancement of \( CO \) at the receptor site is the sum of contributions from the sensitive emission grid flux \((\text{detailed in section 2.3})\). The linear regression slopes of the modeled \( \Delta XCO: \Delta XCO_2 \) based on PKU, MEIC, and observations are shown in Table 3. The outliers are excluded according to the three standard deviations criterion. Modeled \( \Delta XCO \) shows a good relationship with \( \Delta XCO_2 \) \((R^2 = 0.97 \text{ for MEIC and 0.96 for PKU})\). The modeled data displays a slightly greater correlation than the observed \( \Delta XCO \) and \( \Delta XCO_2 \) \((R^2 = 0.86 \text{ for the diurnal variation method, } R^2 = 0.83 \text{ for the regional enhancement method})\). The reason is that modeled values only take the anthropogenic influence of \( CO \) into account, ignoring the \( CO \)-related but not the \( CO \)-related signal, such as the resident respiration of \( CO_2 \). The observed \( \Delta XCO \) and \( \Delta XCO_2 \) values based on regional background enhancement display the weakest correlation \((R^2 = 0.83)\) due to the uncertainty of the modeled background value. The simulated regression slope of \( \Delta XCO \) and \( \Delta XCO_2 \) in 2019 is 14.91 ± 0.36 ppm ppm\(^{-1}\) for MEIC and 21.04 ± 0.70 ppm ppm\(^{-1}\) for PKU. The MEIC and PKU inventories are 42.54\% and 101.15\% higher than the observed value \((10.46 ± 0.11 \text{ ppm ppm}^{-1})\), respectively.

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| Dataset             | \( \Delta XCO: \Delta XCO_2 \) (ppb ppm\(^{-1}\)) | \( R^2 \) |
|---------------------|---------------------------------------------|------|
| Emission inventories| MEIC 2019 14.91 ± 0.36 0.97               |      |
|                     | PKU 2019 21.04 ± 0.70 0.96                |      |
| Observations        | FTS (diurnal variation) 2019 10.46 ± 0.11 0.86 |      |
|                     | FTS (regional enhancement versus background) 2019 9.06 ± 1.89 0.83 |      |
observations at Miyun, Beijing, during winter 2006. Despite the observation and comparative methods, the emission inventories in urban China might overestimate $\Delta$CO:$\Delta$CO$_2$. The lack of consideration of CO$_2$ emissions from the respiration of the residents in dense urban regions may lead to the overestimation of bottom-up-based $\Delta$CO:$\Delta$CO$_2$ (Wang et al., 2010). Either the overestimation of CO anthropogenic flux or under-consideration of CO sinks are possible reasons for the elevated bottom-up estimates of $\Delta$CO:$\Delta$CO$_2$ (Vardag et al., 2015; Shan et al., 2019).

The main reason why MEIC and PKU overestimated $\Delta$XCO:$\Delta$XCO$_2$ surrounding Beijing is likely due to the overestimation of the regional CO emissions and underestimation of CO$_2$ emission. The $\Delta$XCO and $\Delta$XCO$_2$ discussed in section 3.4 are directly linked to the regional emission. The difference between the modeled and observed $\Delta X_{gas}$ is directly proportional to the difference between the emission inventories and the actual emission. The deviation of the regression fitting equation with the 1:1 line shows the model-observed difference. The slope value of modeled $\Delta$XCO to observed is less than one, suggestive of typical overestimations of CO emissions of 30% and 35% for MEIC and PKU, respectively. The underestimation of CO$_2$ emissions magnifies the effects of overestimated CO emission, which contributes to the larger difference between the modeled $\Delta$XCO:$\Delta$XCO$_2$ and the observed ratio. For MEIC, a relatively smaller underestimation of CO$_2$ emissions makes the modeled $\Delta$XCO: $\Delta$XCO$_2$ closer to observations.

### 4. Conclusions

Data was collected for clear days over one year for XCO and XCO$_2$ using the portable FTS EM27 in the urban area of metropolitan Beijing. The overall variation in XCO$_2$ typical variations in anthropogenic emissions (traced by the XCO variation) which are overlaid upon the biogenic and meteorological field effects. Correlation analyses between the XCO and XCO$_2$ enhancements provided useful information to identify the characteristic of combustion efficiency in Beijing. The ($\Delta$XCO:$\Delta$XCO$_2$)$_{obs}$ observed in urban Beijing (10.46 ± 0.11 ppb ppm$^{-1}$) is higher than other FTS urban stations (Karlsruhe, Pasadena, Tsukuba, and Paris), suggesting a high anthropogenic proportion of CO$_2$ emissions and lower combustion efficiency in Beijing. Daily $\Delta$XCO$_2$: $\Delta$XCO$_2$ varies remarkably with seasonality and weather conditions. The $\Delta$XCO: $\Delta$XCO$_2$ in summer shows large uncertainty, and the correlation of $\Delta$XCO and $\Delta$XCO$_2$ is weaker than the other three seasons. According to the air mass pathways arriving in Beijing tracked by the WRF-XSTILT model, the observation site had upwind sources from the NW and NCP of 62.93% and 26.72% of overall observation days, respectively. Air masses passing over the NCP region increased the proportion of $\Delta$XCO to $\Delta$XCO$_2$ (11.46 ± 0.20 ppb ppm$^{-1}$), which exceeded the slope with a clean upwind source (8.23 ± 0.10 ppb ppm$^{-1}$). When backward column footprints were combined with emission inventories, observations could be quantitatively compared with the emission inventories. The MEIC and PKU inventories are 42.54% and 101.15% higher than the observed values, respectively. After comparing the observed regional enhancement with the modeled $\Delta$XCO and $\Delta$XCO$_2$, the main reason for the difference is the overestimation of the regional CO emissions and underestimation of CO$_2$ emission. The less drastic underestimation of CO$_2$ emissions for the MEIC results in the improved modeling of $\Delta$XCO:$\Delta$XCO$_2$ compared to PKU. This work highlights the necessity for long-term column measurements in the heavily CO-emitting Beijing region. However, one station could only capture limited information on a regional scale within a larger urban signal. The background value obtained from the model contains a degree of uncertainty. Compared to in situ surface observations, FTS stations only record the $X_{gas}$ in clear-sky days, which can potentially lead to bias from a less homogeneous sampling. An intensive experimental FTS station combined with WRF-XSTILT could open up additional potential pathways for regional emissions studies.

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