

Quantifying and Attributing CO Emissions over Central Asia using TROPOMI and Explicit Observational Uncertainty

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Abstract: Carbon monoxide (CO) is [an important](#) atmospheric constituent [affecting](#) both air quality and climate. Using TROPOMI CO and HCHO column retrievals within the Model-Free Inversion Estimation Framework (MFIEF), [we derived](#) daily gridded CO [emission estimates over](#) Central Asia, [including](#) Xinjiang, Kazakhstan, Kyrgyzstan, and Uzbekistan, [for 2019–2024. Regional mean CO emissions decreased by 37.9% between 2019 and 2024, with a significant seasonally adjusted trend of \$-0.33 \mu\text{g m}^{-2} \text{s}^{-1} \text{yr}^{-1}\$. The identified emission hotspots also weakened over the study period. Higher emissions during winter and early spring are consistent with enhanced combustion associated with residential heating and industrial activity. A perturbation-based uncertainty analysis propagates assumed observational uncertainties through the inversion and indicates that approximately 69% of the daily grid-cell emission estimates fall outside the adopted uncertainty bounds, highlighting the potential influence of atmospheric variability, dust loading, small sources, and observational uncertainty on the reliability of grid-scale emission estimates. The spatial distribution of satellite-derived grid-scale CO emission estimates reveals differences in urban emissions relative to bottom-up inventories, indicates a moderate positive association between coal consumption and inferred CO emissions at selected industrial locations, and highlights emissions associated with persistent underground coal fires, which are not explicitly represented as a separate source category in conventional anthropogenic emission inventories. This study demonstrates the potential of MFIEF as an observation-driven approach for characterizing regional CO emission patterns and temporal variability in data-sparse regions and provides complementary information for emission-inventory development and source characterization when production and location data are not available or are out of date.](#)

Keywords: CO emissions; TROPOMI; Spatiotemporal distribution; Uncertainty analysis; Central Asia

1. Introduction

Air pollution and climate change are significant environmental problems resulting from economic development, urbanization, energy consumption, and transportation (Adams et al., 2020; Bel and Holst, 2018; Liang et al., 2019). [Air pollution from combustion-related emissions can adversely affect human health, atmospheric chemistry, and ecosystem stability, whereas greenhouse gas accumulation contributes to long-term climate change and associated environmental risks.](#)

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- 删除了: The pixel-by-pixel satellite-inferred emissions and bottom-up activity estimates, while also identifying missing sources such as underground coal fires. identifies grids containing emissions in urban centers both lower and higher than those in bottom-up estimates databases, a somewhat linear relationship between coal consumption and emissions in industrial areas, and new sources from new small industry and coal fires. .
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- 删除了: improvement and mitigation strategies, and highlights the framework’s potential extension to CH₄ and CO₂ retrieval-based emission estimation

Fossil fuel combustion, particularly coal utilization, represents a major common driver of both atmospheric pollutant emissions and greenhouse gas emissions, emphasizing the importance of accurately characterizing emission sources and their temporal variations. CO is one of the lesser considered short-lived climate pollutants (Hanaoka and Masui, 2020; Khalil and Rasmussen, 1988, 1984; Szopa et al., 2021). It is a major sink of the hydroxyl radical (OH), which in turn affects the concentrations of methane (CH₄) and ozone (O₃) (Dai et al., 2024; Patra et al., 2014; Shepherd et al., 2024; Yin et al., 2015; Zhang et al., 2006).

CO is formed due to the incomplete combustion of fossil fuels, chemical reactions when reducing iron and other minerals during extraction and processing, brick and cement production, and biomass burning (Holloway et al., 2000; Khalil and Rasmussen, 1990; Koppmann et al., 2005). In the atmosphere, CO is oxidized to carbon dioxide (CO₂) and is also produced through the oxidation of volatile organic compounds (VOCs) (Joshi et al., 2023; Koppmann et al., 2005; World Health Organization, 2021). Under unpolluted atmospheric conditions, CO concentrations can exhibit substantial seasonal variability, partly due to seasonal changes in OH abundance and atmospheric mixing (Lelieveld et al., 2016; Spivakovsky et al., 2000). In industrial areas, CO has been observed to have elevated concentration when coal consumption and overall production increase and/or combustion efficiency decreases (Munsif et al., 2021; Tian et al., 2022; Zhao et al., 2012). This includes during the winter when additional combustion is often found for residential heating and extra production occurs to meet end-of-year industrial deadlines (Batey, 1997; Li et al., 2025). Similarly, CO is observed to be elevated locally when biomass burning occurs, including wildfires, agricultural burning, and residential solid-fuel use (Lin et al., 2020; Wang et al., 2021, 2025).

Contemporary approaches to estimating CO emissions encompass several primary technical pathways, each with distinct advantages and region-specific limitations. Bottom-up inventories combine activity data (e.g., fuel consumption, industrial production, vehicle kilometers traveled) with sector-specific emission factors, as implemented in databases such as EDGAR (Crippa et al., 2020; Olivier et al., 1999) and China's Multi-resolution Emission Inventory for China (MEIC) system (Zheng et al., 2018). These inventories provide detailed sectoral and temporal information but often suffer from outdated activity data, uncertain emission factors, and incomplete representation of unregulated or diffuse sources, particularly in developing regions. Observation-constrained analyses of global NO_x and CO emission variability can provide complementary information on combustion sources that also contribute to CO₂ emissions (Wang et al., 2025). Direct atmospheric observations, obtained from ground-based networks (e.g., World Meteorological Organization Global Atmosphere Watch (WMO GAW) stations) and satellite instruments (e.g., MOPITT, TROPOMI), offer spatially consistent and temporally continuous coverage, enabling detection of large-scale pollution events such as biomass burning (Buchholz et al., 2022; Worden et al., 2013). However, retrieval uncertainties, cloud contamination, and limited sensitivity near the surface restrict their capacity for precise emission quantification. Chemical transport model (CTM) inversions (e.g., GEOS-Chem, WRF-Chem) integrate atmospheric chemistry and dynamics to optimize emissions (Jiang et al., 2015; Kopacz et al., 2010; Tang et al., 2022), providing physically consistent estimates but requiring substantial computational resources and reliable prior inventories that are often unavailable in data-sparse regions. Isotopic analyses (Brenninkmeijer et al., 1999; Röckmann et al., 2002) can effectively distinguish anthropogenic and biogenic CO sources, yet their application is constrained by the need for high-precision laboratory measurements and limited spatial representativeness. Despite these methodological advances, CO emissions remain more

uncertain than other well-monitored species such as CH₄, NO₂, and SO₂ (Lorente et al., 2021; Schneising et al., 2019; Wang et al., 2022; Wei et al., 2020). These challenges are particularly acute in Central Asia, where sparse monitoring, limited inventories, and heterogeneous sources—ranging from industrial activities to persistent underground coal fires—have hindered comprehensive assessments and model validation. Persistent underground coal fires in Xinjiang may begin through spontaneous oxidation of exposed coal and may also be initiated or intensified when mining exposes coal seams and fractures. Because these processes are partially related to human activity, we use the neutral term “persistent underground coal fires” and interpret them separately from regulated industrial sources.

Central Asia, as a core area of the Belt and Road Initiative, the emission control of its coal-intensive industries is crucial for regional air quality and transboundary pollution transport. Due to the rapid increase in coal use, industrial development, and economic growth, CO emissions are not well characterized because observational networks are sparse and emission patterns are changing rapidly (Akhmedzhanov and Karadanov, 2020; Rupakheti et al., 2021; Zeng and Shen, 2022; Zhang et al., 2011; Zhao et al., 2022). This region of the world has diverse and unique geographical and climatic conditions, environmental conditions, and development patterns, based heavily on coal and minerals (Pirajno et al., 2011; Zeng and Shen, 2022). Understanding CO emissions here is of growing importance for assessing the efficiency of coal use, especially for both non-power-generating industries and power production, in part due to the continued future growth expected in this area, and potential for addressing and formulating environmental and climate mitigation strategies (National Energy Administration, 2015; State Council, 2023).

To address these gaps, this study applies the Model-Free Inversion Estimation Framework (MFIEF), a lightweight, mass-conserving, and observation-driven modeling system designed to infer surface CO emissions from satellite retrievals of CO and HCHO without requiring complex prior inventories or computationally intensive adjoint simulations (Li et al., 2025; Lin et al., 2020). By explicitly accounting for satellite observational uncertainties, MFIEF provides a rapid and flexible means of estimating emissions at high spatial and temporal resolution, well suited for data-sparse and dynamically evolving regions such as Central Asia (Inness et al., 2022; Savenets et al., 2022). The framework enables observation-driven characterization of the spatial and temporal variability of CO emissions and facilitates the investigation of sources that may be underrepresented in conventional inventories. Ultimately, the results contribute to improving emission inventories, informing mitigation policies, and advancing understanding of CO’s role in regional air quality and climate interactions, with broader implications for carbon neutrality and sustainable development goals (Abduh et al., 2021; World Health Organization, 2021).

2. Data and Methods

2.1 Study Area

The study area spans from 35° N to 50° N latitude and 70° E to 100° E longitude, including Xinjiang and parts of Kazakhstan, Kyrgyzstan, Uzbekistan, Tajikistan, Afghanistan, Pakistan, and adjacent regions. The climate in this region is predominantly temperate continental, including cold and dry winters and hot and arid summers, with large diurnal and annual

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150 temperature variations. The Xinjiang area has extensive coal mining and many coal-dependent industries. The region also
151 contains rural areas where residential solid fuel combustion and occasional biomass burning may contribute to seasonal CO
152 variability. In addition, coal is widely used to generate power which is then exported to areas in eastern China. This leads to
153 significant emissions of CO and other combustion byproducts. Similarly, Kazakhstan, Kyrgyzstan, and Uzbekistan also have
154 many industries related to energy and minerals, and also play a crucial role in the regional CO emissions landscape. Fig. 1
155 illustrates the study area together with the coal bases, steel plants, and power plants included in the source-category analysis.
156 These facilities are located in Xinjiang and adjacent parts of Gansu and Qinghai, whereas the satellite-based inversion was
157 conducted over the entire study domain. For the category-level temporal analysis, only steel plants and coal-fired power plants
158 that operated continuously throughout 2019–2024 were retained, resulting in a fixed sample of 4 steel plants and 53 coal-fired
159 power plants.

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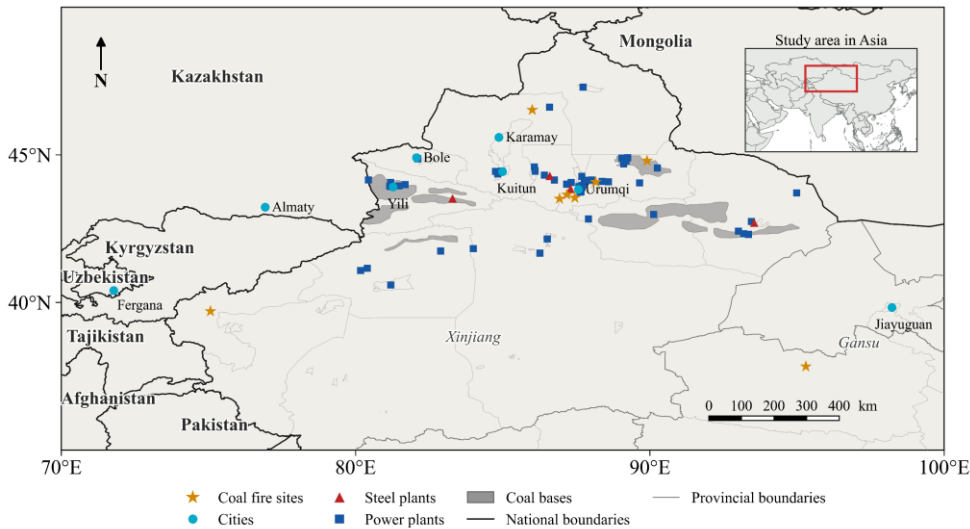
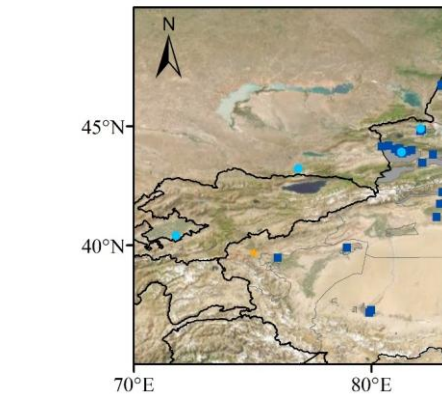
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Figure 1: study area (latitude: 35° N–50° N and longitude: 70° E–100° E) and Xinjiang's major coal bases, plants.

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161 **Figure 1.** Study area (35°–50° N, 70°–100° E). The mapped coal bases and industrial facilities provide geographic context for
162 the source-category analysis and are located in Xinjiang and adjacent parts of Gansu and Qinghai. For the 2019–2024
163 category-level statistics, only continuously operating facilities were retained, comprising 4 steel plants and 53 coal-fired power
164 plants.

165 2.2 Data

166 **TROPOMI:** The Sentinel-5 Precursor (SSP) satellite was launched in October 2017, carrying the TROPospheric
167 Monitoring Instrument (TROPOMI), a push-broom imaging spectrometer with a swath width of 2600 km (5.5 km×7 km

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186 resolution at nadir), and a local equator crossing time around 13:30, providing global coverage once daily. CO total columns
187 are retrieved under both clear-sky and partially cloudy-sky conditions using the CO absorption band near 2.3 μm in the
188 shortwave infrared (SWIR). Total-column averaging kernels for clear-sky and cloudy conditions provide insights into vertical
189 sensitivity (Borsdorff et al., 2023; Landgraf et al., 2016). In this work, the Level 3 OFFL data product from Google Earth
190 Engine (GEE) is filtered to ensure the quality assurance flag is above 0.5, and resampled using the Weighted Polygonal Remote
191 Sensing Toolkit HARP (<http://stcorp.github.io/harp/doc/>). The final analysis grid cell has a resolution of 0.05°x0.05°.

192 HCHO is a volatile organic compound (VOC), and is important due to its being both directly emitted into the atmosphere
193 by combustion, industrial activities, traffic, and fires; as well as being produced due to the chemical decay of heavier VOCs.
194 Since the atmospheric lifetime of HCHO is much shorter than that of CO₂, the loading of HCHO scales linearly with the rate
195 of in-situ production of CO (Cohen and Prinn, 2011), with the decay mostly due to OH and photolysis, and as such is sensitive
196 to the weather, climate, water availability, aerosols, and other complex atmospheric factors (Anderson et al., 2021; Dienhart et
197 al., 2021; Lui et al., 2017). The column loading of HCHO is also successfully retrieved by TROPOMI, although TROPOMI is
198 less sensitive to HCHO than to CO (Romahn et al., 2021). In this work, concurrent HCHO and CO total-column observations
199 at the same time and location are used, allowing for consistency between the retrieved species. In this study, Level-3 HCHO
200 data from GEE are used and resampled by HARP to a resolution of 0.05°x0.05°. These data can be downloaded from
201 <https://disc.gsfc.nasa.gov/datasets>.

202 Wind: European Centre for Medium-Range Weather Forecasts Reanalysis v5 (ERA5) data is a widely used reanalysis
203 dataset providing high-resolution climate data, covering multiple meteorological parameters and variables (Hersbach et al.,
204 2020). This study employs wind field products averaged at 05:00 and 06:00 Coordinated Universal Time (UTC) under the
205 monthly mean surface pressure field. The 05:00 and 06:00 UTC times correspond to the TROPOMI observation window over
206 the study region. More details can be found at <https://www.ecmwf.int/en/forecasts/dataset/ecmwfreanalysis-v5>.

207 EDGAR: The Emissions Database for Global Atmospheric Research (EDGAR) emissions database (Olivier et al., 1999)
208 provides monthly bottom-up emissions for CO₂ and other species. This study used monthly sectoral CO emissions from
209 EDGAR version 8.1 at a spatial resolution of 0.1°x0.1° and resampled the data to the 0.05°x0.05° MFIEF grid for spatial
210 comparison. The included sectors comprise the power industry, industrial combustion, buildings, transport, agriculture, fuel
211 exploitation, industrial processes, and waste management. EDGAR represents anthropogenic emissions estimated from activity
212 data, emission factors, and spatial allocation proxies, whereas MFIEF derives grid-scale emission signals from satellite
213 observations without explicitly separating anthropogenic and non-industrial sources. No explicit uncertainty bounds are
214 provided with the data, although studies show it can range from 20% up to 150% depending on the species and location, on
215 grids which have a source (i.e., the value is not zero or empty) (McDuffie et al., 2020). The data can be accessed at
216 <https://edgar.jrc.ec.europa.eu/>.

217 AAOD: The Multi-angle Imaging Spectro Radiometer (MISR), aboard NASA's Terra satellite, is a pioneering instrument
218 designed to characterize Earth's atmosphere and surface through simultaneous observation at nine distinct viewing angles. A
219 key advanced product derived from MISR data is the Absorption Aerosol Optical Depth (AAOD). This parameter

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- 删除了: EDGAR: The Emissions Database for Global Atmospheric Research (EDGAR) emissions database (Olivier et al., 1999) provides monthly bottom-up emissions for CO, NO_x, VOCs, and CH₄, among other species. This study specifically utilizes sectoral monthly grid data provided by the version 8.1 database, with a resolution of 0.1°×0.1°, with subsequent resampling at 0.05°×0.05°. The

quantitatively estimates the fractional optical thickness of [absorbing aerosols](#), primarily caused by light-absorbing species such as black carbon and mineral dust, [and is relevant to trace gas inversions including CO and HCHO which are based on atmospheric absorption](#). The L3 AAOD data product used in this paper has a resolution of [0.25°](#) and is resampled to [the common 0.05° grid cell only to align the datasets; this resampling does not add spatial information](#). The data can be searched from <https://disc.gsfc.nasa.gov/datasets> ([Liu et al., 2025](#)).

2.3 Research Methods

The MFIEF is utilized to estimate CO emissions ([Li et al., 2023; Qin et al., 2023](#)). Equation (1) takes into account factors such as CO column changes, chemical decay and production, and transport, and relies on retrieved data from TROPOMI CO and HCHO to approximate the weighting coefficients (α_i) of the underlying partial differential equations that govern the transformation between CO and its driving factors, using the same equations used by world-leading chemical transport models ([Cohen and Prinn, 2011](#)). Specifically, emissions cause CO to increase, acting as a source term, while chemical reactions and mixing between in-plume and out of plume regions may lead to the loss or gain of CO, and are represented by a first order linearization of differential equations acting upon and by the CO concentration. The transport of CO is also an important factor, affected by both pressure-induced and advective processes, with the advective term representing the horizontal movement of CO due to wind, and the pressure-induced transport term accounting for the convergence or divergence of the atmosphere itself, accounting for the gain or loss due to non-uniform winds across the concentration and emissions gradients. As such, the transport terms can either increase or decrease the CO column in a given location depending on the direction and magnitude of the flow ([Li et al., 2025; Lu et al., 2025a](#)), which account for topography, land-sea breezes, variable weather systems, and more simplified atmospheric conditions in which there is a substantial concentration gradient due to strong variation in emissions sources in space, with a non-uniform wind field. Combining these terms yields the CO emissions Eq. (1), where:

$$E_{CO} = \frac{dV_{CO}}{dt} + \alpha_2 \cdot V_{CO} + \alpha_3 \cdot \nabla(\mathbf{u} \cdot V_{CO}) - \alpha_4 \cdot V_{HCHO} \quad (1)$$

V_{CO} represents the TROPOMI CO column loading [after conversion to units of \$\(\mu\text{g} \cdot \text{m}^{-2}\)\$](#) . $\alpha_2 \cdot V_{CO}$ represents the sink of CO with $\alpha_2(\text{s}^{-1})$ as the inverse CO lifetime. $\alpha_4 \cdot V_{HCHO}$ represents CO production, with $\alpha_4(\text{s}^{-1})$ as the inverse HCHO lifetime. $\nabla(\mathbf{u} \cdot V_{CO})$ represents the daily zonal and meridional dispersion of CO with a unit of $(\mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1})$, and $\alpha_3(\text{m}^{-1})$ represents the transport distance. This mass conservation equation for CO provides a framework for understanding and quantifying the changes in CO columns in the atmosphere, considering the various processes that influence its distribution, following [Li et al. \(2025\)](#).

The [emission, production, and transport terms of Equation \(1\)](#) are initialized, TROPOMI observations and their retrieved uncertainties, meteorological data, and either an initial a priori emissions dataset, or our own previously inverted emissions dataset in the case of multiple iterations. The fitting is based on multiple least squares regression [for each grid cell](#) on a month-by-month basis, reflecting the differences in albedo, elevation, land-surface use, emissions type, and differences in confounding variables on each pixel specifically grouping all daily data from TROPOMI and the ECMWF reanalysis where

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删除了: The MFIEF is utilized to estimate CO emissions (Li et al., 2023; Qin et al., 2023). Equation (2) takes into account factors such as CO column changes, chemical decay and production, and transport, and relies on retrieved data from TROPOMI CO and HCHO in order to approximation of the weighting terms of the underlying partial differential equations that govern the transformation between CO and its driving factors, using the same equations used by world-leading chemical transport models (Cohen and Prinn, 2011). Specifically, emissions cause CO to increase, acting as a source term, while chemical reactions and mixing between in-plume and out of plume regions may lead to the loss or gain of CO, and are represented by a first order linearization around the CO concentration. The transport of CO is also an important factor, affected by both pressure-induced and advective processes, with the advective term representing the horizontal movement of CO due to wind, and the pressure-induced transport term accounting for the convergence or divergence of the atmosphere itself (i.e. vertical motion). Since vertical changes do not affect inverted CO total column loadings, so instead this term accounts for the gain or loss due to non-uniform winds. As such, the transport terms can either increase or decrease the CO column in a given location depending on the direction and magnitude of the flow (Cohen et al., 2024; Li et al., 2023; Qin et al., 2023), and are related to topography, land-sea breezes, weather systems, and more. Combining these terms yields the CO emissions Eq. (1), where:

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they all occur. This process provides sufficient data to compute the weighting terms α_i , while also accounting for the changes in land-surface, atmosphere, and other factors. In the case where a pixel in any given month either does not have enough data or produces a fit with a p value larger than 0.05, the values of the model are set to NaN on that [grid cell](#) during that month. To ensure physically realistic and mathematically consistent values for the coefficients α_2 , α_3 , and α_4 , physical cutoffs are applied. Any value that is not positive ($\alpha > 0$) is set to NaN.

[Equation \(1\)](#) is then used to calculate estimates of CO emissions across all [grid cells](#). To estimate the maximum probability density of the coefficients during the corresponding [grid cell](#)-by-[grid cell](#) and month-by-month basis, the solution distribution of α within the valid range (from the 10th percentile to the 90th percentile of the Probability Density Function (PDF)) is sampled 1000 times per [grid cell](#). The mean value of this distribution is assigned as the day's average emission in that [grid cell](#), and [its](#) standard deviation [was used to characterize parameter-induced variability in the estimated CO emissions](#). This process allows us to estimate the coefficients and their uncertainties, taking into account the available data and the physical constraints on the parameters, providing a more robust and accurate representation of the CO mass conservation equation in the study region.

[For the calculation of CO emissions, grid-cell filtering based on the distribution of retrieved values is required following the method established by Zheng et al \(2026\). Random errors are expected to cluster around zero and to produce approximately symmetric positive and negative values. For each grid cell, we identify the most negative retrieved emission value, \$-0 \mu\text{g m}^{-2} \text{s}^{-1}\$, and define a symmetric interval from \$-0\$ to \$0 \mu\text{g m}^{-2} \text{s}^{-1}\$. Values within this interval are treated as being dominated by retrieval noise rather than a resolvable positive emission signal and are excluded. This screening reduces the influence of grid cells for which the inversion cannot distinguish a true positive emission signal from the effects of observational noise.](#)

[2.4 Propagation of observational uncertainty into CO emission estimates](#)

[The accuracy and precision targets for TROPOMI total CO column prior to launch were 15% and 10% respectively \(Landgraf et al., 2016; Martínez-Alonso et al., 2020; Veeffkind et al., 2012\). There hasn't been enough research on specific issues which impact the column retrievals of CO and HCHO including aerosol loadings and microphysical properties, cloud cover under low cloud conditions, changes in surface reflectivity from pixel to pixel, and the initial CO and CH₄ data, which could result in greater uncertainty than what has been mentioned \(Hu et al., 2024; Lu et al., 2024\). The uncertainty in TROPOMI CO total column has implications for CO emission calculations. First, emission calculation relies on accurate and precise CO column concentration data. Second, since emissions estimation requires the use of a non-linear gradient operator \(Lu et al., 2025b; Zheng et al., 2026\), uncertainty in the spatial domain plays an additional role. Consequently, if small perturbations within the observational uncertainty range lead to relatively large changes in inverted emissions, this indicates that the computed emission estimates may be strongly affected by noise \(Beekmann and Derognat, 2003; Rao, 2005; Reilly et al., 1987\). In such instances, the emissions cannot be regarded as stable; thus, their reliability is questionable. This makes it difficult to assess the robustness of the inverted emissions and may reduce confidence in the retrieved emission estimates.](#)

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2.4 Uncertainty

The accuracy and precision targets for TROPOMI total CO column prior to launch were 15% and 10% respectively (Landgraf et al., 2016; Martínez-Alonso et al., 2020; Veeffkind et al., 2012). There hasn't been enough research on specific issues like aerosol levels, cloud cover in low cloud conditions, changes in surface reflectivity from pixel to pixel, and the initial CO and CH₄ data, which could result in greater uncertainty than what has been mentioned (Hu et al., 2024; Lu et al., 2024). The uncertainty in TROPOMI CO total column has implications for CO emission calculations. First, emission calculation relies on accurate and precise CO column concentration data. Second, since emissions estimation requires the use of a non-linear gradient operator (Cohen and Prinn, 2011), uncertainty in the spatial domain plays an additional role. Such uncertainty may propagate through the emissions equations, leading to cases where small perturbations in the observations (i.e., due to observational or retrieval uncertainty) lead to larger perturbations in inverted emissions (Beekmann and Derognat, 2003; Rao, 2005; Reilly et al., 1987) In such instances, the emissions cannot be regarded as stable; thus, their reliability is questionable. This lack of precise quantification hinders a more robust understanding of CO (...)

485 Accordingly, this study propagates the assumed observational uncertainties through MFIEF and quantifies the corresponding
 486 range of variation in the inverted emissions and allows for a more robust product.
 487 Random perturbations were applied to both the retrieved CO and HCHO column data within conservative retrieval
 488 uncertainty bounds. The perturbations ~~were~~ randomly added to ~~(or subtracted from)~~ the observed data and a bootstrap
 489 ~~resampling was~~ performed to sufficiently sample the ~~perturbation~~ space. Specifically, each data point of CO column and HCHO
 490 column retrieved from TROPOMI, on a daily and gridded basis, ~~was randomly perturbed~~ within ~~±10% of its original value~~. In
 491 addition to ~~point-wise perturbations~~, the ~~non-linear influence on~~ spatial ~~gradients was also evaluated~~. For example, given two
 492 neighboring ~~grid cells~~, if there is an overlapping portion of the data range after the random ±10% perturbations are applied,
 493 the direction of the computed gradient, and therefore the emissions may shift. In these cases, the ~~resulting~~ gradient ~~becomes~~
 494 ~~unstable~~, and therefore the emissions estimate is also not likely trustworthy. ~~The calculated changes define the emission~~
 495 ~~uncertainty range associated with propagation of the perturbed observational inputs~~. Specifically, the ~~relative emission~~
 496 ~~response was calculated~~ by Eq. (2):

$$F_{resp} = (E_{CO(\pm)} - E_{CO}) / E_{CO} \quad (2)$$

497 ~~$E_{CO(\pm)}$~~ denotes the newly calculated CO emission, ~~E_{CO}~~ denotes the initial CO emission, ~~E_{resp}~~ denotes the change of CO
 498 emission result, i.e., the observational contribution to emission uncertainty. The distribution of these responses quantifies the
 499 observational contribution to emission uncertainty, and forms a minimum of the total top-down emissions uncertainty. Grid
 500 cells exhibiting responses larger than the adopted threshold are classified as exceeding this uncertainty range and are excluded
 501 from the final emission product, following Zheng et al., (2026). The CO emission calculation and uncertainty-screening
 502 workflow is summarized in Fig. 2.

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- 删除了: $E_{CO(\pm)}$ denotes the newly calculated CO emission, E_{CO} denotes the initial CO emission, E_{un} denotes the change of CO emission result, i.e., uncertainty. Other retrieved terms will behave similarly, meaning that if any of the variables behave in such a way locally across any grids on any given day, those subsets of grids may lead to a change in the net total inverted emissions which is too large, and the data ultimately not being reliable. This work adapts a conservative approach, setting any such data to NaN and discarding it from the ultimate inverted emissions products, following Zheng (2024). The CO emission calculation and data screening process flow chart is shown in Fig. 2.

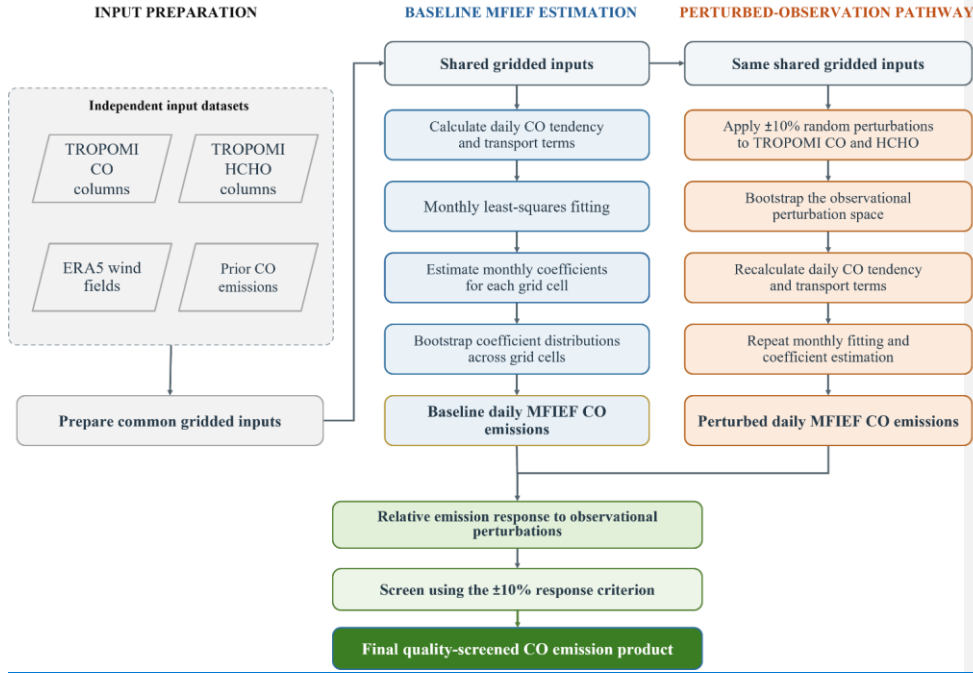


Figure 2. Workflow of MFIEF-based CO emission estimation and uncertainty-propagation analysis based on $\pm 10\%$ perturbations of TROPOMI CO and HCHO observations.

2.5 Statistical trend analysis

Regional means were calculated from daily spatial means. To quantify the temporal trend in regional CO emissions, we fitted an ordinary least-squares regression to the monthly mean emission series from January 2019 to December 2024. The model included continuous time, expressed in years since January 2019, and calendar-month fixed effects to account for seasonality:

$$E_t = \beta_0 + \beta_1 T_t + \sum_{m=2}^{12} \gamma_m M_{m,t} + \varepsilon_t \quad (3)$$

where E_t is the regional monthly mean CO emission flux, T_t is the time elapsed since January 2019, and $M_{m,t}$ represents calendar-month indicator variables. Heteroskedasticity- and autocorrelation-consistent standard errors with a maximum lag of

12 months were used to account for serial correlation in the monthly series. As a non-parametric robustness test, we also applied the seasonal Mann-Kendall test with a period of 12 months and calculated the corresponding seasonal Sen's slope. Statistical significance was evaluated at the 0.05 level. The regression used all 72 monthly observations. The percentage change between the 2019 and 2024 annual means was reported as a descriptive endpoint comparison and was not interpreted as the regression slope. Trend coefficients are reported with 95% confidence intervals.

3. Results

3.1 Spatiotemporal Distribution of CO Observations

Fig. 3a presents the spatial distribution of the average TROPOMI CO total column from 2019 to 2024 across the study area, revealing a domain-wide mean of 1.54×10^{18} molecules cm^{-2} . The concentration is predominantly influenced by anthropogenic activities, with elevated values ($>2.0 \times 10^{18}$ molecules cm^{-2}) located in, around, and downwind of major urban and industrial centers such as Urumqi, Fergana, Almaty, Jiayuguan, and the Zhundong Coalfield. In contrast, natural areas like the Tianshan and Altai Mountains exhibit lower concentrations, averaging around 1.0×10^{18} molecules cm^{-2} . Moderately high CO levels are also observed in the desert regions of central Xinjiang, likely due to enhanced photochemical production from VOCs under intense ultraviolet radiation. Elevated CO is also found in parts of northwestern Kazakhstan, where biomass burning and residential solid fuel combustion may contribute to seasonal variations in the regional background CO, particularly during the heating season. The statistical distribution of the CO total column, shown in Fig. 3b, deviates from a normal distribution and is distinctly bimodal, with the 25th and 75th percentiles for the entire period calculated at 1.23 and 1.83×10^{18} molecules cm^{-2} , respectively. Analysis of interannual variability shows that the highest CO levels occurred in 2021, with an annual mean of 1.59×10^{18} molecules cm^{-2} , particularly in high-value zones. Conversely, 2022 marked the lowest concentrations across both high- and low-value zones, with the mean dropping to 1.44×10^{18} molecules cm^{-2} . The values subsequently rebounded to an intermediate level in 2023 (1.52×10^{18} molecules cm^{-2}), followed by a further increase in 2024 (1.56×10^{18} molecules cm^{-2}). This is consistent with findings indicating that the general air pollution policies implemented by China are driving CO (Li et al., 2025; Liu et al., 2024) more than lockdowns related to COVID-19 (Fan et al., 2021; Zheng et al., 2021a).

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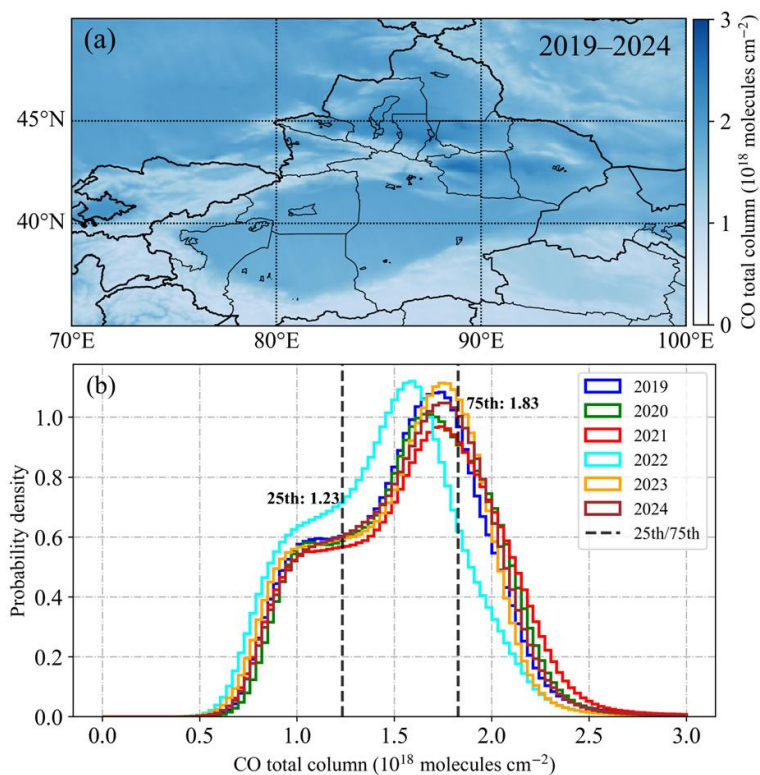
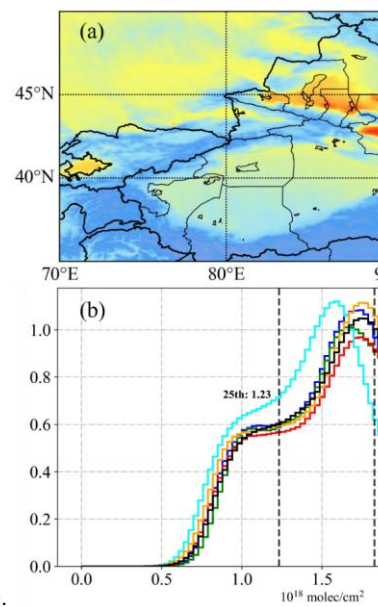


Figure 3. Spatial and statistical distributions of TROPOMI CO total columns over the study area during 2019–2024. (a) Mean spatial distribution over the six-year period. (b) Annual probability density distributions of CO total-column values over the study area.

Fig. 4 presents the available daily CO total columns from 2019 to 2024, in the $0.05^\circ \times 0.05^\circ$ resampled grid cells containing the Wugonggou underground coal-fire site (44.075°N , 88.156°E), the center of Urumqi (43.825°N , 87.575°E), Xinjiang Daan Special Steel (42.700°N , 93.542°E), and Shenhua Guoneng Hami Power Station (42.411°N , 93.009°E). Each time series represents one selected grid cell. The mean CO total columns at these four locations were 1.96 , 2.17 , 2.25 , and 2.05×10^{18} molecules cm^{-2} , respectively. All locations generally exhibited higher values in winter and lower values in summer, reflecting the combined effects of seasonal emissions, atmospheric transport, boundary-layer mixing, and faster CO oxidation associated with higher OH abundance in summer (Liu et al., 2022; Sun et al., 2023; Yang et al., 2021). The Wugonggou grid cell exhibited weaker seasonality, with a monthly amplitude of 0.32×10^{18} molecules cm^{-2} , compared with 0.71 – 0.77×10^{18} molecules

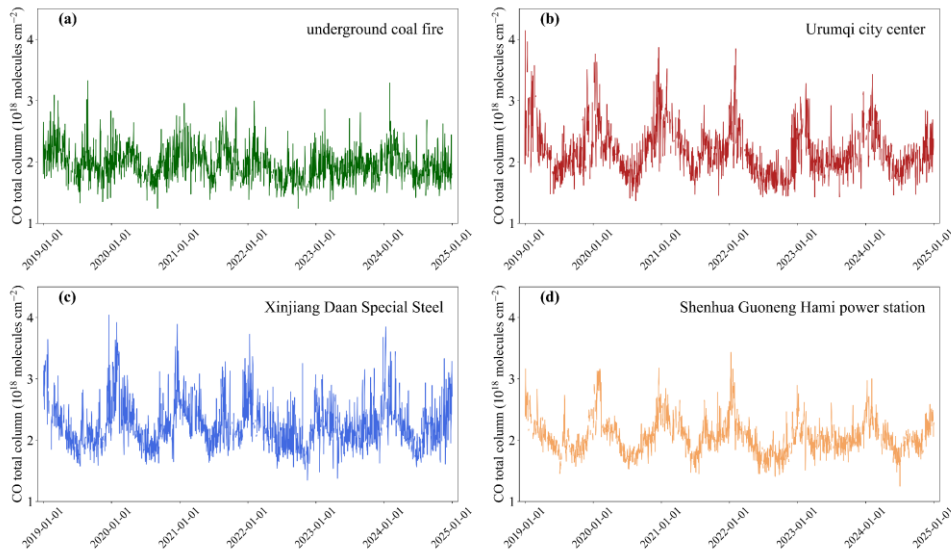


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606 cm^{-2} at the other three locations. This weaker variation is consistent with the relatively persistent nature of underground coal
607 combustion and the weaker influence of seasonally varying urban and industrial activities in the surrounding area.



608
609 **Figure 4.** Time series of TROPOMI CO total columns in the $0.05^\circ \times 0.05^\circ$ grid cells containing (a) the Wugonggou underground
610 coal-fire site, (b) the center of Urumqi, (c) Xinjiang Daan Special Steel, and (d) Shenhua Guoneng Hami Power Station during
611 2019–2024.

612 3.2 Spatiotemporal Distribution of CO Emissions

613 To distinguish dust-dominated regions from other aerosol types, we utilized the AAOD ratio between 443 nm and 880
614 nm. Mineral dust typically exhibits stronger absorption at shorter wavelengths due to its iron oxide content, leading to higher
615 AAOD₄₄₃/AAOD₈₈₀ ratios, whereas other aerosols like black carbon show less spectral dependence. A higher ratio thus
616 suggests a greater contribution of dust to total absorption. Based on this spectral behavior, we selected a threshold of
617 AAOD₄₄₃/AAOD₈₈₀ > 4.5 to identify and mask dust-dominated areas, thereby minimizing potential biases in CO emission
618 estimates associated with dust contamination. Grid cells exceeding this threshold were excluded, effectively removing spurious
619 emission signals caused by enhanced atmospheric absorption and scattering from absorbing aerosols which impact the retrieval
620 of CO and HCHO. As shown in Fig. 5a, the spatial distribution of AAOD ratios highlights extensive dust-affected areas,
621 primarily in the desert and arid parts of western China and southern Kazakhstan. This screening process resulted in the

删除了: The CO total column on a daily basis is provided in

上移了 [1]: Fig.

删除了: 4 for the following sites: the Wugonggou Coal Fire in Fukang, (latitude 44.075° N, longitude 88.156° E) a representative power plant, a representative steel plant, and the center of Urumqi. As observed, the CO column contains a mixture of seasonal factors (generally higher in winter and lower in summer), anthropogenic factors (generally more production near the end of the year, and additional power for heating in the coldest months including November and December), as well as some amount of random day-to-day variability. This set of findings is tied to both anthropogenic and natural forcing related to temperature, light and OH. Summer usually has more UV radiation, more OH, and less CO (Liu et al., 2022; Sun et al., 2023; Yang et al., 2021). Among the four points in Fig. 4, the underground coal fire site, with a mean CO total column of 1.96×10^{18} molec/cm² and the smallest interquartile range (25th-75th percentile: 1.78-2.11), is primarily influenced by natural conditions and exhibits lower peak values and reduced daily variability. In contrast, the central area of Urumqi (mean: 2.18; 25th-75th percentile: 1.90-2.37), the steel plant (mean: 2.09; 25th-75th percentile: 1.82-2.30), and the power plant (mean: 2.16; 25th-75th percentile: 1.88-2.32) are mainly affected by human activities and production demands, showing generally higher concentrations and stronger variability. All four locations display seasonal fluctuations with clear peaks and troughs. Statistically, emissions from the power plant closely resemble those in central Urumqi ($R^2 = 0.55$, $p < 0.001$, RMSE = 0.30×10^{18} molec/cm²), while the Wugonggou Coal Fire site aligns more closely with the steel plant ($R^2 = 0.35$, $p < 0.001$, RMSE = 0.33×10^{18} molec/cm²). These distinctions in both concentration levels and temporal behavior suggest that different types of emission sources can be identified based on their varying seasonal patterns (...)

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699 exclusion of approximately 14% of the study area, underscoring the importance of aerosol correction in satellite-based
700 emission inversion.

701 To quantify emission uncertainty associated with observational inputs, we varied the total column concentrations of CO
702 and HCHO within ranges of $\pm 10\%$ and $\pm 20\%$, representing the adopted TROPOMI retrieval uncertainty ranges. Grid-cell
703 estimates were retained when the relative change in emissions remained within the corresponding uncertainty bound. As
704 illustrated in Fig. 5b, under the $\pm 10\%$ and $\pm 20\%$ perturbation thresholds, 31.3% and 33.7% of grid cells, respectively, remained
705 within the corresponding uncertainty bound over the six-year period. Fig. 5b shows how the emission estimates change as
706 observational uncertainty propagates through the inversion, with estimates falling within the corresponding bounds retained.
707 Furthermore, Fig. 5c shows the number of days with reliable emissions after applying the $\pm 10\%$ filter, with a spatial average
708 of approximately 135 valid days per grid cell. The results indicate that stricter observational-uncertainty filtering leads to a
709 gradual reduction in retained emission estimates, quantifying how input-column uncertainty affects the inversion outcomes.
710 This is an important point, since many papers calculating emissions using satellites have assumed that reducing uncertainty
711 leads to a more precise end result (Estrada et al., 2025; Shen et al., 2023), whereas this result demonstrates that both very high
712 and very low emissions results are both impacted by observational noise.

删除了: To evaluate the impact of input data uncertainty on emission estimates, we performed a perturbation analysis by varying the total column concentrations of CO and HCHO within ranges of $\pm 10\%$ and $\pm 20\%$, reflecting uncertainties from TROPOMI retrievals and model inputs. Grid cells were classified as having “reliable emissions” only if the relative variation in estimated emissions remained within the corresponding threshold. As illustrated in Fig. 5b, under the $\pm 10\%$ and $\pm 20\%$ uncertainty thresholds, 31.3% and 33.7% of grid cells, respectively, met the reliability criterion over the six-year period. Furthermore, Fig. 5c shows the number of days with reliable emissions after applying the $\pm 10\%$ filter, with a spatial average of approximately 135 days per grid cell. The results indicate that stricter uncertainty filtering leads to a gradual reduction in credible emission estimates, highlighting the sensitivity of inversion outcomes to uncertainties in the input column concentrations.

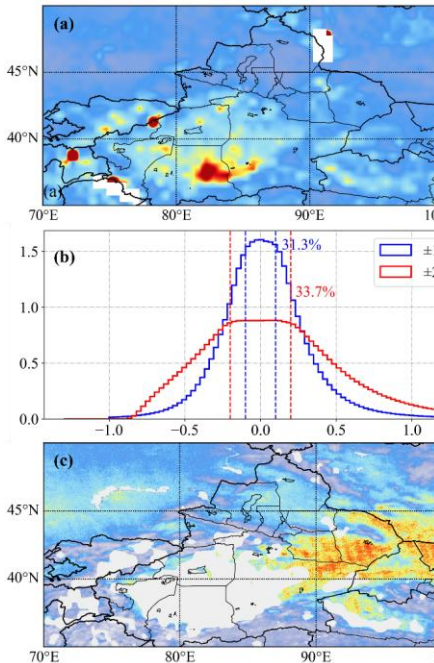


Figure 5: (a) AAOD₄₄₃/AAOD₈₈₀ Ratio; (b) Plausible CO emission share with perturbation added to both CO and HCHO columns; and (c) number of credible data in uncertainty calculations.

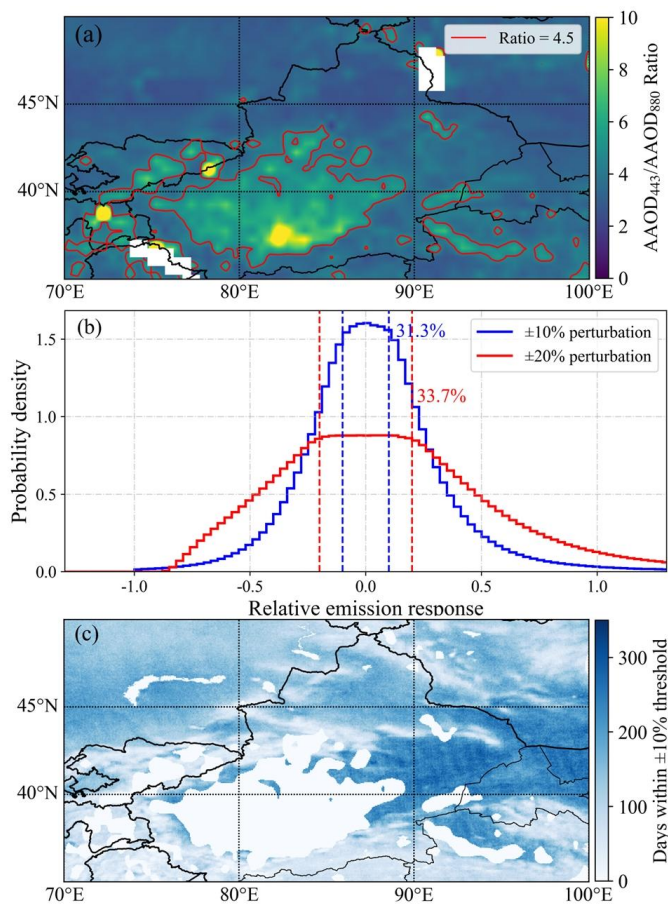


Figure 5: (a) Spatial distribution of the mean AAOD₄₄₃/AAOD₈₈₀ ratio during 2019–2024. Grid cells with ratios greater than 4.5 were identified as dust-dominated areas and excluded from the subsequent CO emission inversion. (b) Probability density distributions of the relative emission changes resulting from $\pm 10\%$ and $\pm 20\%$ perturbations in the TROPOMI CO and HCHO total-column inputs. The blue and red dashed lines indicate corresponding uncertainty bounds of ± 0.1 and ± 0.2 , respectively. The annotated percentages represent the proportions of estimates remaining within the corresponding uncertainty bounds. (c) number of valid retrievals after $\pm 10\%$ observational-uncertainty filtering.

745 Following application of the $\pm 10\%$ observational-uncertainty filter, the spatial distribution of the MFIEF-derived
746 emissions broadly corresponds to the major features of the CO total column distribution. Fig. 6 summarizes the annual spatial
747 distributions, probability density distributions, and spatial variability of the derived emissions from 2019 to 2024. Regional
748 mean emissions and the mean intensity of the identified hotspot regions decreased over the study period, while the emission
749 distributions shifted toward lower values.

750 Regional mean CO emissions decreased from $4.93\ \mu\text{g m}^{-2}\text{ s}^{-1}$ in 2019 to $3.06\ \mu\text{g m}^{-2}\text{ s}^{-1}$ in 2024, corresponding to a 37.9%
751 endpoint reduction. The seasonally adjusted trend was $-0.33\ \mu\text{g m}^{-2}\text{ s}^{-1}\text{ yr}^{-1}$ (95% confidence interval: -0.52 to $-0.15\ \mu\text{g m}^{-2}$
752 $\text{s}^{-1}\text{ yr}^{-1}$; HAC-adjusted $p < 0.001$). A seasonal Mann–Kendall test independently supported the decreasing trend: $-0.20\ \mu\text{g m}^{-2}$
753 $\text{s}^{-1}\text{ yr}^{-1}$ ($p < 0.001$). This overall trend toward lower emissions is further reflected in the evolution of emission hotspots, which
754 we defined as regions where the emission flux exceeded twice the study area's mean ($6.72\ \mu\text{g m}^{-2}\text{ s}^{-1}$). A total of 11 such
755 hotspots were identified, primarily situated between 39.83° – 46.58°N and 81.08° – 98.23°E . Their average intensity decreased
756 markedly from $9.51\ \mu\text{g m}^{-2}\text{ s}^{-1}$ in 2019 to $5.63\ \mu\text{g m}^{-2}\text{ s}^{-1}$ in 2024, representing a total reduction of 40.8% or an average annual
757 decrease of $0.68\ \mu\text{g m}^{-2}\text{ s}^{-1}\text{ yr}^{-1}$. The PDFs of the emissions (Fig. 6b) provide further evidence of this systematic reduction,
758 showing a distinct leftward shift toward lower values. The distribution peak decreased from $3.34\ \mu\text{g m}^{-2}\text{ s}^{-1}$ in 2019 to $2.85\ \mu\text{g m}^{-2}$
759 s^{-1} in 2024. Concurrently, the PDFs of the standard deviations (Fig. 6c) indicate a significant weakening of spatial
760 heterogeneity, with the peak value dropping from $1.66\ \mu\text{g m}^{-2}\text{ s}^{-1}$ to below $1.30\ \mu\text{g m}^{-2}\text{ s}^{-1}$. In summary, the results depicted in
761 Fig. 6 clearly highlight a marked downward trend in CO emissions between 2019 and 2024, characterized by reduced hotspot
762 intensity, a systematic shift of the emission distribution to lower values, and reduced spatial variability. These observed changes
763 may reflect the combined effects of emission-control policies, industrial restructuring, energy use, meteorology, sampling, and
764 non-industrial sources; their individual contributions cannot be separated here. Together, these results indicate a statistically
765 significant regional decline, reduced hotspot intensity, and weaker spatial variability over the study period.

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- 删除了: a $\pm 10\%$ uncertainty filter, the spatial pattern of CO emissions was found to be consistent with that of the CO total column, though significant interannual variability was observed. As shown in Fig. 6, a pronounced decreasing trend from 2019 to 2024 is demonstrated by the contraction of emission hotspots in
- 删除了: (a), a systematic leftward shift of the probability distribution toward lower values (b), and a marked reduction in ...
- 删除了: reflected in the standard deviation distribution (c). The annual mean CO concentration in
- 删除了: area has been substantially declined, from $4.93\ \mu\text{g/m}^2/\text{s}$ in 2019 to $3.06\ \mu\text{g/m}^2/\text{s}$ in 2024, with a reduction of approximately 38%.
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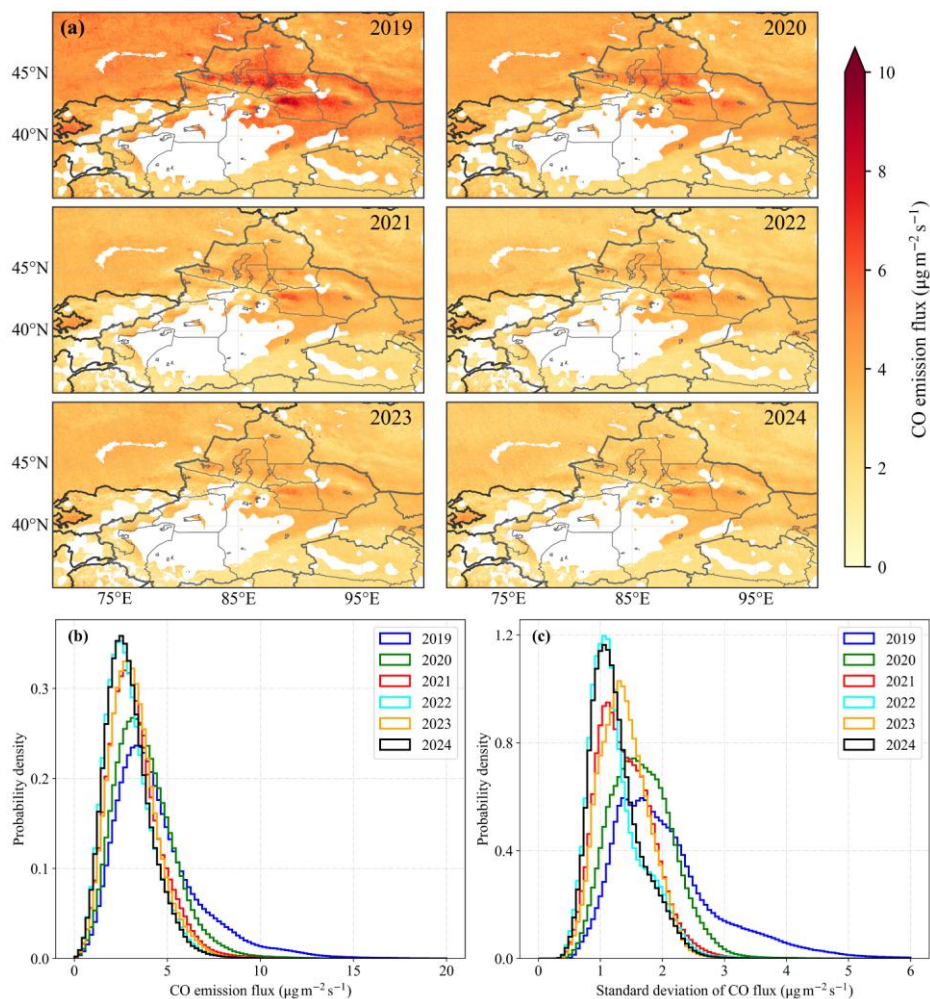
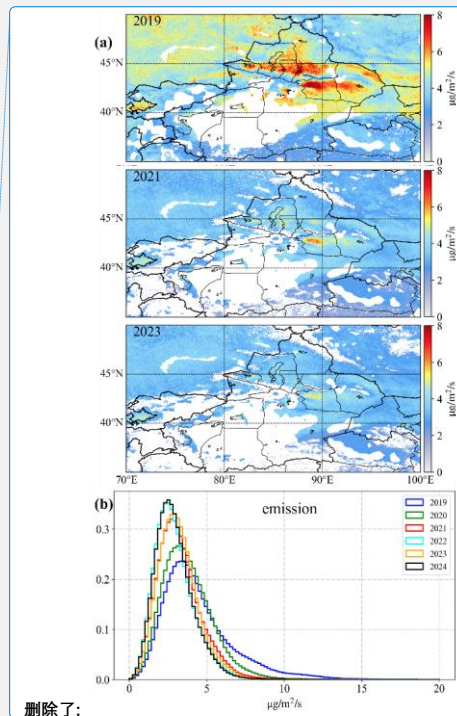


Figure 6. Annual characteristics of MFIEF-derived CO emissions from 2019 to 2024. (a) Annual mean CO emission flux; grid cells with fewer than 10 valid emission-estimate days in a year are shown as missing values. (b) Probability density distributions of annual mean CO emission fluxes. (c) Probability density distributions of the interannual standard deviation of CO emission fluxes. Emission-flux units are $\mu\text{g m}^{-2} \text{s}^{-1}$.



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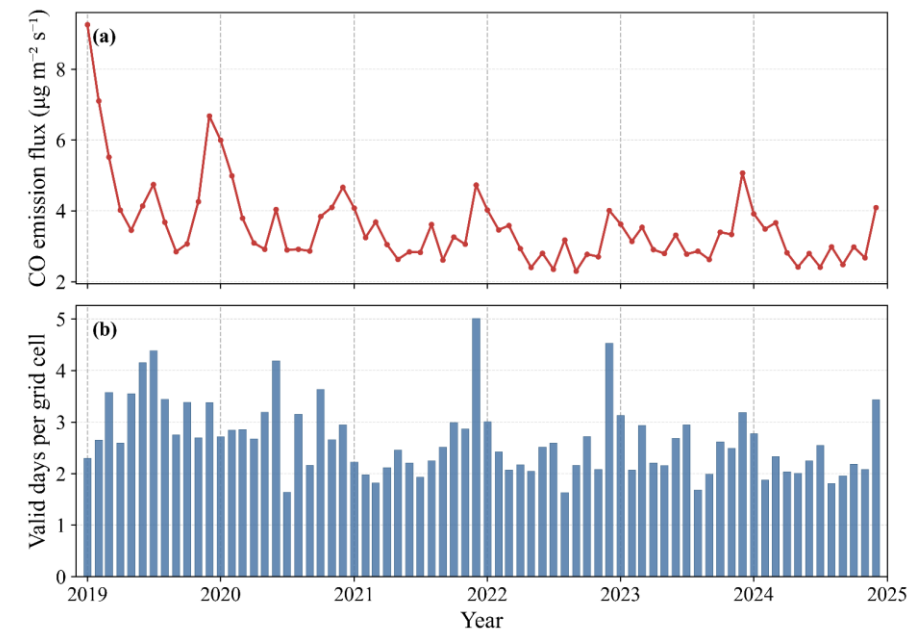
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820 Fig. 7 presents the monthly regional mean CO emissions and the mean number of valid emission-estimate days per grid
821 cell from January 2019 to December 2024. A valid day is defined as a day for which a grid cell retains an emission estimate
822 after the quality and perturbation-response screening described above; the plotted valid-day metric is the spatial mean of this
823 monthly count across the study domain. Overall, CO emissions exhibited a clear downward trend, decreasing from an
824 exceptionally high level of $9.25 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ in January 2019 to around $2.5\text{--}3.0 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ after 2022, with occasional wintertime
825 rebounds (e.g., $5.07 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ in December 2023). Seasonal fluctuations are evident, with higher emissions generally observed
826 in winter and early spring, and lower values during late spring and summer, consistent with stronger heating-related combustion
827 sources during colder times, annual production cycle which ends on Chinese New Year varying from mid-January through
828 mid-February. The number of valid observational days shows substantial variability, ranging from fewer than 2 days per month
829 (e.g., July 2020 and August 2022–2024) to more than 5 days per month (December 2021). The valid-day series is presented
830 only as a sampling diagnostic and is not interpreted as a physical driver of emission variability.

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- 删除了: Despite this variability, the temporal patterns of emissions are robust and not simply driven by observation coverage. The joint evolution of these two metrics highlights both the long-term decline in regional CO emissions and the persistence of seasonal emission peaks, which together reflect the combined influences of anthropogenic activity, energy use, and atmospheric conditions across Xinjiang and Central Asia
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831 Figure 7. Monthly mean CO emission flux and temporal sampling coverage over the study region from January 2019 to
832 December 2024. (a) Spatially averaged monthly CO emission flux derived from grid cells satisfying the observational-quality
833 criteria. (b) Mean number of valid observation days per grid cell per month, averaged over all grid cells in the study region.
834

including grid cells with no valid observations in a given month. The valid-day series is shown as a diagnostic of temporal sampling coverage rather than as an explanatory driver of emission variability.

Fig. 8 illustrates the temporal evolution of gridded CO emissions at four representative locations: the Wugonggou underground coal-fire site, Urumqi city center, Xinjiang Daan special steel plant, and Shenhua Guoneng Hami coal-fired power plant. Because the inversion is performed at a spatial resolution of $0.05^{\circ} \times 0.05^{\circ}$, these estimates represent the net emissions from all sources within the corresponding grid cell rather than emissions from an individual facility. Over 2019–2024, the mean emission fluxes were $4.50 \mu\text{g m}^{-2} \text{s}^{-1}$ at Wugonggou, $4.75 \mu\text{g m}^{-2} \text{s}^{-1}$ in central Urumqi, $5.77 \mu\text{g m}^{-2} \text{s}^{-1}$ at the selected steel-plant grid cell, and $4.43 \mu\text{g m}^{-2} \text{s}^{-1}$ at the selected coal-fired power-plant grid cell. The largest observed emission values generally occurred near the beginning of the study period, followed by a pronounced weakening of high-emission episodes. At Wugonggou, for example, the maximum daily emission flux of $14.0 \mu\text{g m}^{-2} \text{s}^{-1}$ occurred in December 2019, while the maximum observed values in subsequent years declined to 9.98, 7.65, 5.09, 5.42, and $4.97 \mu\text{g m}^{-2} \text{s}^{-1}$ from 2020 to 2024, respectively. Its annual mean similarly decreased from $6.38 \mu\text{g m}^{-2} \text{s}^{-1}$ in 2019 to $3.03 \mu\text{g m}^{-2} \text{s}^{-1}$ in 2024. The temporal evolution further indicates that the long-term reduction was particularly evident in the weakening of wintertime high-emission episodes, whereas changes during the remaining months were weaker and less monotonic. Similar attenuation of high-emission episodes was observed in the Urumqi and power-plant grid cells, while the steel-plant grid showed a smaller overall reduction. However, wintertime sampling was sparse in some years, and these seasonal differences should therefore be interpreted as descriptive temporal patterns rather than independent trends. Moreover, the causes of the observed changes cannot be uniquely separated among changes in source activity, emission-control measures, atmospheric transport and chemical loss, and temporal sampling. In particular, Wugonggou is an uncontrolled underground coal-fire source rather than an engineered industrial stack, and its temporal evolution should not be interpreted in terms of the same emission-control mechanisms as the urban and industrial grid cells.

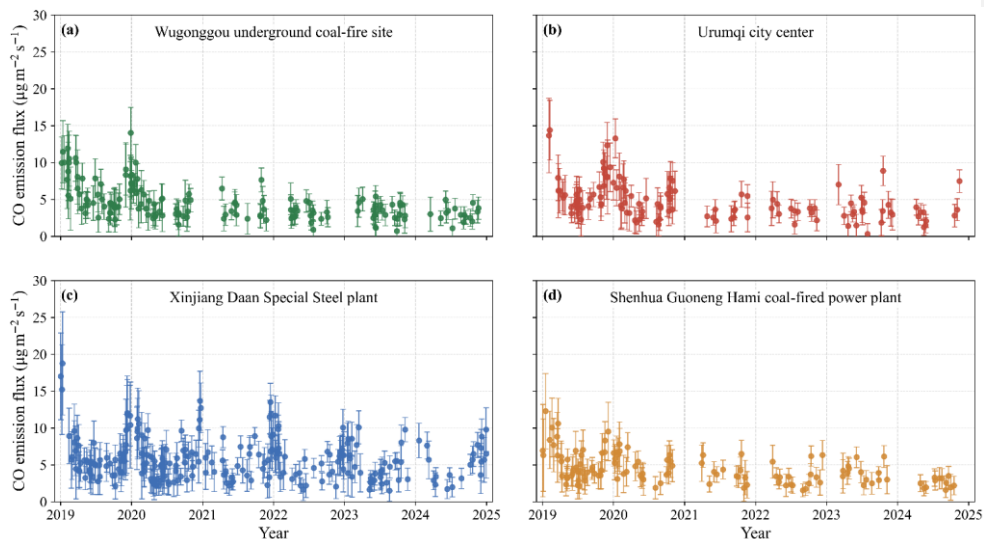


Figure 8. Temporal variations in MFIEF-derived CO emission fluxes at four representative locations from January 2019 to December 2024: (a) the Wugonggou underground coal-fire site, (b) Urumqi city center, (c) the Xinjiang Daan Special Steel plant, and (d) the Shenhua Guoneng Hami coal-fired power plant. Markers represent the mean emission fluxes, and error bars indicate ± 1 standard deviation. The values were extracted from the 0.05° grid cell containing each location.

Table 1 includes industrial facilities that operated continuously throughout 2019-2024. The screened sample contains 53 coal-fired power plants and 4 steel plants; the city-center and underground coal-fire categories each contain 8 locations. Based on grid-scale estimates from this fixed sample, mean emissions decreased from 6.47 to $3.74 \mu\text{g m}^{-2} \text{s}^{-1}$ at city-center locations (42.2%), from 7.09 to $4.18 \mu\text{g m}^{-2} \text{s}^{-1}$ at underground coal fire locations (41.0%), from 6.22 to $4.08 \mu\text{g m}^{-2} \text{s}^{-1}$ at steel-plant locations (34.4%), and from 4.94 to $3.02 \mu\text{g m}^{-2} \text{s}^{-1}$ at coal-fired power-plant locations (38.9%) between 2019 and 2024. The 90th-percentile values also decreased, including from 11.00 to $6.58 \mu\text{g m}^{-2} \text{s}^{-1}$ at steel-plant locations and from 8.20 to $4.69 \mu\text{g m}^{-2} \text{s}^{-1}$ at coal-fired power-plant locations.

903
904

Table 1. Annual statistics of grid-scale CO emission fluxes for selected source categories from 2019 to 2024. Steel-plant and power-plant samples include only facilities operating continuously across all six years (unit: $\mu\text{g m}^{-2} \text{s}^{-1}$).

type	Number of locations	Year	Mean $\pm$ std	10th	20th	50th	80th	90th	Annual change (%)
City Center	8	2019	6.47 $\pm$ 2.93	2.73	3.43	5.30	9.47	12.03	
		2020	5.26 $\pm$ 2.20	2.45	3.09	4.76	7.08	9.08	-19%
		2021	4.35 $\pm$ 1.75	2.16	2.72	4.01	5.74	7.11	-17%
		2022	3.95 $\pm$ 1.59	1.94	2.44	3.57	5.21	6.32	-9%
		2023	3.92 $\pm$ 1.65	2.07	2.65	3.80	5.09	6.01	-1%
		2024	3.74 $\pm$ 1.53	1.96	2.38	3.40	4.96	6.08	-5%
Coal fire site	8	2019	7.09 $\pm$ 3.00	3.35	4.06	5.81	9.93	12.27	
		2020	5.41 $\pm$ 2.23	2.69	3.04	5.07	6.87	9.74	-24%
		2021	4.84 $\pm$ 1.76	2.31	3.15	4.33	6.45	7.34	-10%
		2022	4.54 $\pm$ 1.74	1.92	2.58	4.10	6.41	6.82	-6%
		2023	4.30 $\pm$ 1.76	2.08	2.62	3.82	5.91	7.68	-5%
		2024	4.18 $\pm$ 1.71	1.73	2.18	3.99	5.70	7.22	-3%
Steel plants	4	2019	6.22 $\pm$ 2.77	2.61	3.56	5.36	8.89	11.00	
		2020	4.96 $\pm$ 2.07	2.24	2.95	4.34	6.62	8.37	-20%
		2021	4.33 $\pm$ 1.77	2.25	2.71	3.79	5.54	7.48	-13%
		2022	4.33 $\pm$ 1.56	1.74	2.18	3.41	5.59	8.11	0%
		2023	4.23 $\pm$ 1.68	1.96	2.51	3.84	5.78	7.16	-2%
		2024	4.08 $\pm$ 1.47	1.89	2.48	3.49	5.28	6.58	-4%
Power plants	53	2019	4.94 $\pm$ 2.18	2.34	2.80	4.33	6.85	8.20	
		2020	4.10 $\pm$ 1.78	2.01	2.43	3.80	5.43	6.69	-17%
		2021	3.32 $\pm$ 1.38	1.79	2.20	3.10	4.30	5.17	-19%
		2022	2.99 $\pm$ 1.25	1.37	1.80	2.81	4.01	4.69	-10%
		2023	3.13 $\pm$ 1.33	1.51	2.09	2.92	4.26	4.64	5%
		2024	3.02 $\pm$ 1.21	1.53	1.91	2.88	4.02	4.69	-4%

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Fig. 9 compares estimated coal consumption with satellite-derived grid-scale CO emissions for 11 selected coal-fired power plants in 2023. Coal consumption ranged from approximately 57 to 206 kt coal yr⁻¹, while the mean grid-scale CO emissions ranged from approximately 2.8x10³ to 4.0x10³ t CO yr⁻¹. A moderate positive association was observed between coal consumption and the inferred CO emissions ($r = 0.59$, $p = 0.058$, $N = 11$; Fig. 9a), indicating that plants with higher coal consumption generally tended to be located in grid cells with higher inferred emissions. However, the relationship did not reach the conventional significance level of $p < 0.05$. The relatively broad 25th – 75th percentile ranges also indicate substantial temporal variability in the grid-scale emission estimates.

The grid-scale CO emission-to-coal-consumption ratio, calculated as the satellite-derived CO emission from the 0.05° x 0.05° grid cell containing each power plant divided by the reported coal consumption of that plant, ranged from approximately 14 to 53 g CO kg⁻¹ coal. This ratio is used only as a comparative metric rather than a facility-specific emission factor, because

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the satellite-derived emissions represent the combined emissions from all sources within the corresponding grid cell rather than actual stack emissions. The ratio showed a strong inverse relationship with coal consumption ($r = -0.93$, $p < 0.001$, $N = 11$; Fig. 9b). However, because coal consumption is included in the denominator, the relationship is partly affected by mathematical coupling. Contributions from other sources within the grid cell may further influence the ratio, particularly for plants with lower coal consumption.

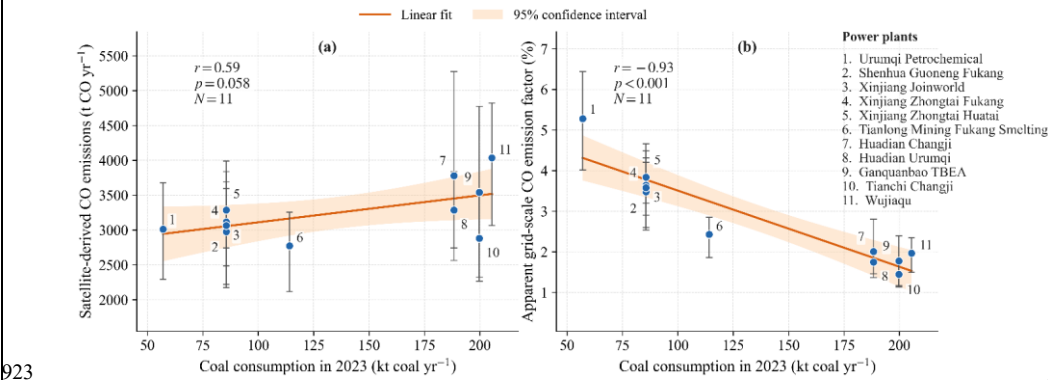


Figure 9. Relationship between satellite-derived CO emissions and coal consumption at selected coal-fired power plants.
(a) Comparison between annual coal consumption and MFlEF-derived CO emissions for individual power-plant grid cells. (b) Relationship between coal consumption and the grid-scale CO emission-to-coal-consumption ratio. Circles and vertical error bars represent the means and 25th–75th percentile ranges, respectively.

4. Discussion

Fig. 10 compares the spatial distributions of the MFlEF-derived grid-scale CO emission estimates and EDGAR anthropogenic CO emissions for their overlapping period from 2019 to 2022. Datasets show relatively high emissions in major urban and industrialized regions in northern Xinjiang, as well as in some regions of Kazakhstan, which may have high CO emissions due to industrial activities, transportation and home heating. In contrast, low emission values are mainly found in remote areas of Xinjiang, such as steppes, deserts, and mountainous regions, as well as in some sparsely populated and less industrialized regions of Kazakhstan. Overall, the geographical distribution of CO emissions shows obvious unevenness, with the emission intensity in urban and industrial areas being significantly higher than that in the surrounding rural and natural areas, especially in the urban centers in northern Xinjiang and some industrial areas in Kazakhstan, where high emissions may be related to the local level of economic development, the structure of energy consumption, and the industrial layout. This broad spatial correspondence indicates that both approaches capture several major anthropogenic emission regions. Nevertheless, local differences are evident around individual urban and industrial areas, reflecting actual differences between

删除了: A comparison between this emission and EDGAR is presented in
删除了: 9a, illustrating
删除了: distribution
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the bottom-up and top-down approaches in the observational record in terms of source, spatial allocation, and temporal sampling.

Fig. 10b shows the grid-cell difference calculated as MFIEF minus EDGAR for the common analysis period. The MFIEF-derived fluxes are higher than the EDGAR anthropogenic emissions across most of the study domain, whereas urban centers such as Urumqi and Almaty show locally lower MFIEF values, with differences reaching approximately $-5 \mu\text{g m}^{-2} \text{s}^{-1}$. Madrazo et al. (2018) compared local Cuban emission inventories with EDGAR for CO emissions and mentioned that EDGAR overestimates the stationary source emission factors by a factor of three, while Li et al. (2025) indicates that both EDGAR and MEIC estimates of well-established urban areas and large factories overestimate CO emissions in Shanxi, while newer and smaller urban sources are underestimated or missing. The distributions of the grid-cell differences are shown in Fig. 10c. Across 2019-2022, differences are positive across most matched grid cells. The annual mean and median differences decrease from 3.95 and 3.64 $\mu\text{g m}^{-2} \text{s}^{-1}$ in 2019 to 2.50 and 2.43 $\mu\text{g m}^{-2} \text{s}^{-1}$ in 2022, respectively, while the distribution peak shifts from 3.20 to 2.20 $\mu\text{g m}^{-2} \text{s}^{-1}$. These statistics characterize the numerical differences between the datasets. EDGAR estimates anthropogenic emissions using sector-specific activity data and emission factors on a monthly time scale and locates them based on a priori information, whereas MFIEF derives a grid-scale signal constrained by atmospheric observations. These differences may arise from additional or missing emission sources, inaccuracies in the spatial allocation of sources, or differences between actual emissions and bottom-up estimates. Differences in atmospheric transport, satellite sampling and retrievals, and inversion assumptions including chemistry, physics, and long-range-transport may all contribute to the observed variability and uncertainty in the top-down estimate. For additional magnitude context, Wang et al. (2025) reported observation-constrained CO emission fluxes ranging approximately from 0 to 30 $\mu\text{g m}^{-2} \text{s}^{-1}$, with values in relatively clean regions generally below 6 $\mu\text{g m}^{-2} \text{s}^{-1}$. The regional mean fluxes obtained in this study ranged from 3.06 to 4.93 $\mu\text{g m}^{-2} \text{s}^{-1}$, with peak values exceeding 10 $\mu\text{g m}^{-2} \text{s}^{-1}$, and therefore are of a comparable order of magnitude and spread. This numerical comparison provides a broad reference for the inferred emission magnitudes.

删除了: Fig. 9b shows the mean difference between the CO emissions estimated in this study and the EDGAR emission inventory across the study area from 2019 to 2022. While the emissions from this study are higher than EDGAR in most regions, urban centers such as Urumqi and Almaty show the opposite trend, with values up to 5 $\mu\text{g}/\text{m}^2/\text{s}$ lower than EDGAR. In the figure, the blue areas of the figure are mainly distributed. Madrazo et al (2018)

删除了: Li et al (2025)

删除了: 9(c) shows the PDF of the differences between our CO emission estimates and the EDGAR inventory over the study region. Across 2019-2022, the differences are predominantly positive, with 99.9% of grid points showing higher emissions in our estimates. The annual mean and median differences gradually decrease from 3.95 and 3.64 $\mu\text{g}/\text{m}^2/\text{s}$ in 2019 to 2.50 and 2.43 $\mu\text{g}/\text{m}^2/\text{s}$ in 2022, indicating a narrowing gap over time. The PDF peaks shift similarly from 3.20 $\mu\text{g}/\text{m}^2/\text{s}$ in 2019 to 2.20 $\mu\text{g}/\text{m}^2/\text{s}$ in 2022, while the overall range remains largely positive. These statistics suggest that our inversion generally yields higher CO emissions than EDGAR, with differences decreasing over time and concentrated in high-emission areas

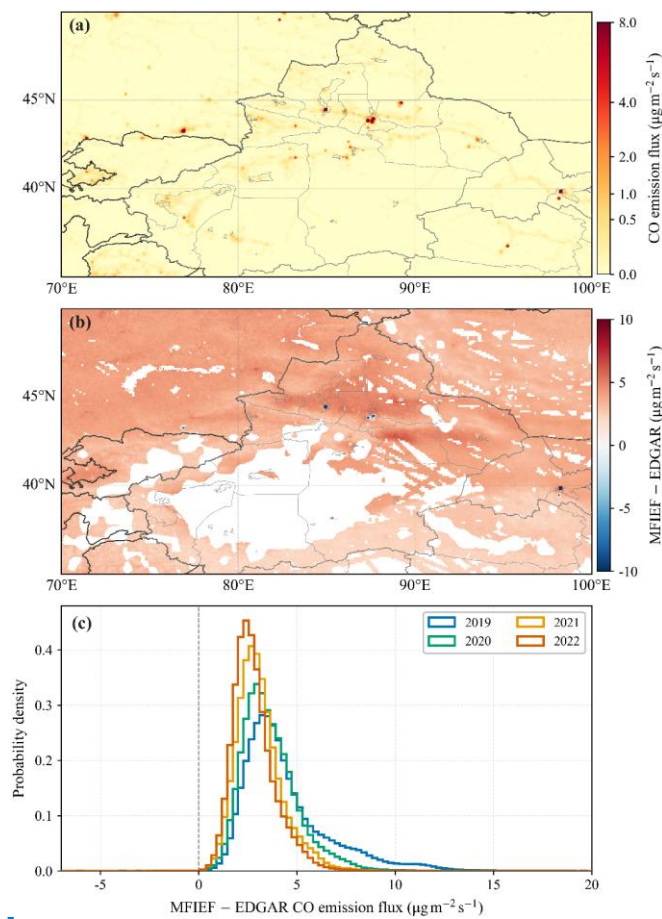


Figure 10. Comparison of CO emission estimates from MFIEF and EDGAR during their overlapping period from 2019 to 2022. (a) Mean EDGAR anthropogenic CO emission flux. (b) Mean grid-cell difference calculated as MFIEF minus EDGAR. Positive values indicate larger MFIEF-derived fluxes, whereas negative values indicate larger EDGAR emissions. (c) Probability density distributions of the monthly grid-cell differences. The differences reflect the distinct source coverage, spatial allocation, temporal representation, and estimation methods of MFIEF and EDGAR.

Fig. 11 compares MFIEF-derived grid-scale CO emission fluxes with EDGAR anthropogenic CO emissions at selected coal-fire sites and urban centers. At the four persistent underground coal-fire sites, MFIEF identifies enhanced CO emissions, whereas EDGAR shows limited emissions in the corresponding grid cells. This difference is expected because persistent

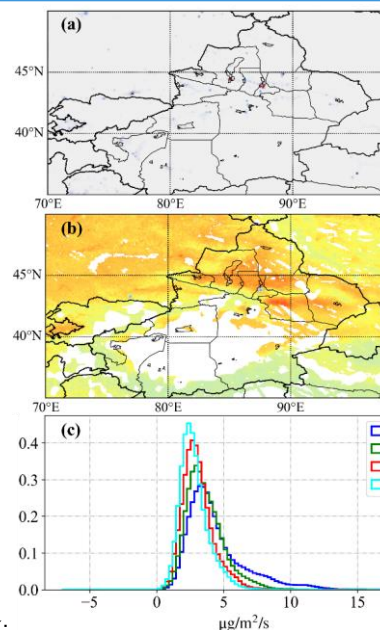
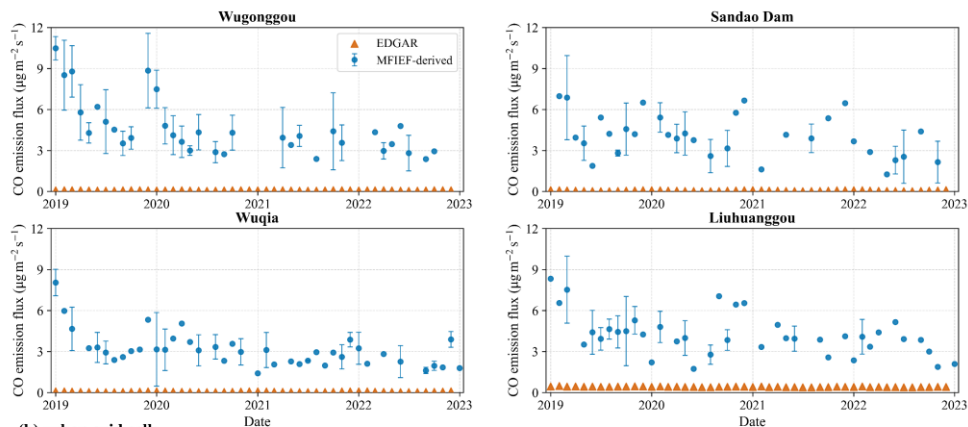


Figure 9: (a) EDGAR CO emissions; and (b) difference between this study's results and EDGAR; and (c) monthly difference PDF.

A fundamental methodological divergence between satellite observations and model-based inventories is demonstrated in Fig.10. At four confirmed coal fire sites, TROPOMI unambiguously detects strong CO emission, whereas the EDGAR inventory shows negligible emissions. This discrepancy arises because the bottom-up EDGAR inventory, by design, excludes non-industrial sources such as coal fires. Furthermore, the comparison in urban regions (e.g., Urumqi, Almaty) reveals a pattern of partial agreement and quantitative divergence. While the datasets show consistent temporal trends, EDGAR exhibits systematic biases, overestimating winter peaks in some cities and displaying spatial inaccuracies in others. These discrepancies are attributable to limitations in the inventory's activity data, the regional applicability of its emission factors, and its coarse spatial resolution. Collectively, these findings underscore the value of top-down satellite data. This approach not only identifies emission sources missing from conventional inventories but also provides an independent means for validating urban emission estimates, proving indispensable for developing

1057 [underground coal fires are not explicitly represented as an independent source category in EDGAR and may therefore be absent](#)
1058 [or only indirectly represented within existing anthropogenic sectors. At the selected urban locations, the differences between](#)
1059 [MFIEF and EDGAR were strongly site dependent. The mean EDGAR emissions at Urumqi and Almaty were approximately](#)
1060 [2.0 and 2.4 times higher than the corresponding MFIEF estimates, respectively, whereas the discrepancy was much larger at](#)
1061 [Jiayuguan, where EDGAR emissions were approximately 63 times higher. The two datasets also exhibited distinct temporal](#)
1062 [characteristics. For example, at Jiayuguan, the inter-month variability of MFIEF monthly emissions was substantially larger](#)
1063 [than that of EDGAR \(coefficient of variation: 41.5% versus 3.7%\). Moreover, daily MFIEF estimates showed considerable](#)
1064 [within-month variability, with median intra-month coefficients of variation of approximately 22–35% across the selected urban](#)
1065 [sites. These differences indicate that discrepancies between bottom-up inventories and satellite-derived estimates are](#)
1066 [associated not only with emission magnitude but also with differences in spatial allocation and prescribed temporal profiles of](#)
1067 [emission sources \(Zheng et al., 2021b\).](#)

(a) coal fire sites



(b) urban grid cells

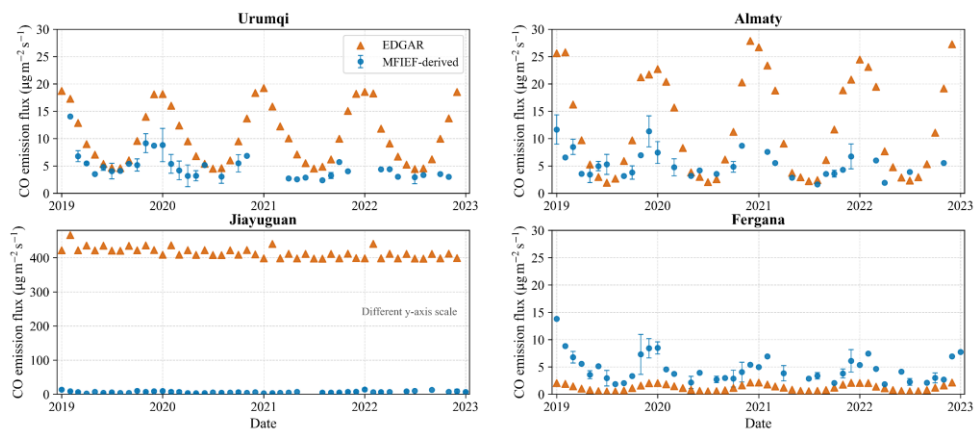


Figure 11. Comparison of MFIEF-derived grid-scale CO emission fluxes and EDGAR anthropogenic CO emissions at selected locations. (a) Persistent underground coal-fire sites, which are not explicitly represented as a separate source category in EDGAR. (b) Selected urban centers. MFIEF-derived emissions are presented as monthly means, with vertical bars indicating the within-month variability (standard deviation) of daily estimates, whereas EDGAR values represent monthly inventory estimates. The comparison illustrates differences in source coverage and temporal representation between the two datasets.

1075 **5. Conclusion**

1076 In this study, we applied the Model-Free Inversion Estimation Framework to derive gridded CO emission estimates over
1077 Central Asia using TROPOMI CO and HCHO total-column retrievals together with meteorological reanalysis data.
1078 Propagating observational uncertainty through the inversion quantifies the corresponding range of derived emissions and
1079 identifies the grid cells where the estimates meet the adopted stability criteria.

1080 Regional mean CO emissions decreased from 4.93 $\mu\text{g m}^{-2} \text{s}^{-1}$ in 2019 to 3.06 $\mu\text{g m}^{-2} \text{s}^{-1}$ in 2024, corresponding to a 37.9%
1081 endpoint reduction. The regional decline remains significant after accounting for seasonality. The identified emission hotspots
1082 weakened over the study period. Higher emissions during winter and early spring were observed at several urban and industrial
1083 locations. These temporal patterns may reflect a combination of changes in industrial activity, residential heating, energy use,
1084 emission-control measures, meteorological conditions, satellite sampling, and persistent underground coal-fire activity.
1085 Interestingly, peak emissions at the underground coal-fire sites occurred one year later than those at the industrial and urban
1086 sites, further suggesting that underground coal fires exhibit distinct temporal behavior and may be inadequately represented in
1087 conventional bottom-up emission inventories. Together, these changes characterize the principal regional evolution of CO
1088 emissions during 2019–2024.

1089 These findings demonstrate the value of observation-driven inversion for regional emission characterization. The
1090 uncertainty analysis quantifies the observational contribution to emission uncertainty and identifies grid-cell estimates that do
1091 not meet the adopted stability criteria. The retained estimates provide complementary constraints on regional emission patterns
1092 and source environments, particularly for characterizing emerging, spatially variable, and temporally dynamic emission
1093 sources that may be inadequately represented in conventional inventories. Future applications to other satellite-retrieved
1094 species will require species-specific evaluation of retrieval sensitivity, chemistry, transport, and independent validation data.

1095 Several limitations should be considered when interpreting the MFIEF-derived CO emissions. First, independent ground-
1096 based or facility-level CO emission measurements with spatial and temporal coverage comparable to the satellite-derived
1097 estimates are unavailable across the study region. The comparisons with EDGAR and reported coal consumption provide
1098 contextual assessments of spatial correspondence and physical plausibility. Second, the derived emissions remain sensitive to
1099 uncertainties in the satellite CO and HCHO retrievals, meteorological fields, temporal sampling, and the assumptions used to
1100 separate transport, chemical production, and surface emissions. In particular, vertical transport can alter the relationship
1101 between surface CO concentrations and satellite-observed column loadings (Wang et al., 2026). Third, the grid-scale emission
1102 flux estimated for an industrial facility includes the combined influence of sources within the corresponding grid cell. The
1103 present results are most appropriate for characterizing broad spatial patterns and relative temporal variability at the grid scale.
1104 Future integration of ground-based observations, facility-level measurements, aircraft or UAV observations, and multi-species
1105 satellite measurements would provide additional constraints on emission magnitude and source attribution.

删除了: In this study, we applied the MFIEF to quantify CO emissions across Central Asia by integrating TROPOMI CO and HCHO retrievals with meteorological reanalysis data. By explicitly accounting for satellite observational uncertainties, our approach provides a more reliable representation of emission dynamics in a region where ground-based measurements and inventory data remain scarce. This is especially important in this region of the world, where the economic growth is very large and industries and urban areas are both expanding rapidly. Furthermore, there is rapid development of both the chemical industry due to the production of many rare earths in this area, coupled with the rapid implementation of solar and wind power, all of which leads to substantial CO emissions during the production process. The results reveal a pronounced interannual decline of approximately 38% in CO emissions from 2019 to 2024, indicating that although not directly targeted by various emissions rules and regulations, that it is never the less being mitigated as a co-emitted substance along with targeted species. Furthermore, distinct seasonal variations are observed with stronger emissions during winter and early spring, reflecting increases in energy demand through the colder periods of the year, industrial production increases in the lead up to the Chinese New Year, and local meteorological influences in part due to the unique snow and desert environment. Attribution analysis further highlights the contribution of major industrial centers, power plants, and urban agglomerations, while also pointing to previously underrepresented sources such as underground coal fires. Comparison with the commonly used EDGAR emissions database underscores both the limitations of global inventories in capturing local emission characteristics and the value of satellite-based approaches for identifying missing or mischaracterized sources. Beyond providing an updated regional emission assessment, this work demonstrates the critical importance of explicitly treating observational uncertainties, which contribute substantially to the fact that about 69% of grids are found to not have sufficiently reliable CO retrievals to be useful for successful emissions inversion. This study advances the understanding of CO emissions in Central Asia by integrating TROPOMI observations with the MFIEF framework and explicitly computing both emissions uncertainties, as well as regions when and where emissions cannot be reliably inverted. The results not only provide valuable constraints for regional inventories and emission source characterization but also strengthen the foundation for retrieving CH₄, CO₂, and other SWIR- and NIR-absorbing species. Beyond regional relevance, the approach offers broader implications for improving bottom-up inventories, supporting air quality modeling, and informing climate and carbon mitigation strategies. Limitations of the current work come from three different directions. First, there are no on-the-ground or direct observations of emissions from CEMS or other networks for

1227 **Data availability**

1228 The authors would like to thank the TROPOMI, ERA-5, and EDGAR products for making their data available. The
1229 satellite CO and HCHO datasets used in this study are available at <https://disc.gsfc.nasa.gov/datasets>. The ERA-5 reanalysis
1230 product is available at <https://doi.org/10.24381/cds.bd0915c6>. The EDGAR product can be accessed from
1231 <https://edgar.jrc.ec.europa.eu/>. The data that support the findings of this study are openly available at the following URL/DOI:
1232 <https://doi.org/10.6084/m9.figshare.28462817>.

1233 **Author contribution**

1234 This work was conceptualized by Jason Blake Cohen and Ye Feng. The methods were developed by Jason Blake Cohen
1235 and Kai Qin. Xiaolu Li and Lingxiao Lu provided insights on methodology. Visualizations were made by Ye Feng and Jason
1236 Blake Cohen. Writing of the original draft was done by Ye Feng and Jason Blake Cohen. Writing at the review and editing
1237 stages were done by Xiaolu Li, Lingxiao Lu, [Shuo Wang](#), Zhewen Liu, Lei Wang, Jian Liu and Jason Blake Cohen.

1238 **Competing Interests**

1239 At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

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