



1 **Structural divergence between satellite and surface CO observations across** 2 **China, North America, and Europe (2015–2024)**

3
4 Zhaojun Tang¹, Zhe Jiang^{1*}, Panpan Yang¹, Jiaqi Chen², Pingqing Fu¹ and Yanan Shen³

5
6 ¹Institute of Surface–Earth System Science, School of Earth System Sciences, Tianjin
7 University, Tianjin, 300072, China.

8 ²Anhui Institute of Meteorological Sciences, Hefei, Anhui, 230031, China.

9 ³School of Earth and Space Sciences, University of Science and Technology of China, Hefei,
10 Anhui, 230026, China.

11
12 *Correspondence to: Zhe Jiang (zhejiang@tju.edu.cn)

13 14 15 **Abstract**

16 Satellite and surface observations of atmospheric carbon monoxide (CO) yield divergent
17 emission estimates, yet the underlying causes of this discrepancy remain poorly understood.
18 Here we present a multi-region analysis spanning 2015–2024 over China, North America, and
19 Europe, assimilating MOPITT satellite retrievals and ground-based CO measurements from
20 over 3300 monitoring stations. Our results reveal that the systematic underestimation of
21 modeled surface CO is a common feature across all three continents, with median observation-
22 to-model ratios of 1.82 (China), 1.65 (North America), and 2.25 (Europe). This bias
23 progressively diminishes from urban to rural sites, by 44% in North America and 58% in
24 Europe, and almost disappears at remote sites, demonstrating that representation error is an
25 important driver of the surface-model mismatch. In contrast, comparisons of column CO
26 between MOPITT and model show no systematic underestimation. Assimilation experiments
27 further illuminate the structural nature of this divergence: while surface-data assimilation
28 greatly improves agreement with in situ observations, it introduces a positive column bias
29 relative to MOPITT. This bias is most pronounced over China and Europe but remains modest
30 over North America, and accordingly the decadal trends from surface CO show a sharp decline
31 in China ($-5.92\% \text{ yr}^{-1}$) vs. plateaus after 2018 and 2016 in North America and Europe, while



32 satellite-derived column trends are substantially weaker. Our multi-region analysis
33 demonstrates that the satellite-surface discrepancy is a structurally embedded feature of the
34 observing system, and advocates for joint utilization of both observation platforms in future
35 inversion studies to achieve robust and self-consistent CO emission estimates.

36

37 **1. Introduction**

38 Atmospheric carbon monoxide (CO) is a key trace gas affecting tropospheric chemistry
39 and air quality, with major sources including fossil fuel combustion, biomass burning, and
40 hydrocarbon oxidation. Through its role as a major sink of hydroxyl radicals (OH) and a
41 precursor to tropospheric ozone, CO exerts significant influence on the atmosphere's oxidizing
42 capacity and secondary pollutant formation. Satellite observations, with global coverage, have
43 been widely used to investigate atmospheric CO changes (Han et al., 2018; Jeffery et al., 2024)
44 and constrain CO emissions (Zheng et al., 2019; Fortems-Cheiney et al., 2024; Tang et al.,
45 2026). Meanwhile, surface measurements from ground-based monitoring networks provide
46 high temporal resolution and direct measurements of CO concentrations at the surface level,
47 enabling detailed assessments of local emission changes and air quality impacts (Zhou et al.,
48 2017; Feng et al., 2020b; Xie et al., 2025). Recent advances in computational techniques further
49 enabled the integration of satellite and surface CO observations with machine learning models
50 (Han et al., 2022; Wei et al., 2023), thereby deepening our understanding of CO sources and
51 spatiotemporal variations.

52 However, a growing body of inverse modeling studies reveals a notable discrepancy
53 between CO emission estimates derived from satellite and surface observations. Emission
54 estimates based on satellite data generally find limited adjustments relative to a priori
55 inventories: for instance, Zheng et al. (2019) and Fortems-Cheiney et al. (2024) reported a
56 posteriori CO emission estimates in China and Europe about 1-5% lower than the respective a



priori estimates, while Tang et al. (2026) suggested a upward adjustment of 5-9% for China. By contrast, inversions using surface observations indicate much larger underestimations, for example, Feng et al. (2020a) and Jia et al. (2025) reported that CO emissions in China could be underestimated by 180-250%. This divergence points to a potential systematic discrepancy between satellite and surface CO observations, particularly over China, underscoring the need for a thorough understanding of the drivers of this discrepancy to ensure accurate use and interpretation of CO datasets.

Understanding this discrepancy requires recognizing the inherent limitations of each observation type. Limited vertical resolution of satellite measurements means that retrieved lower-tropospheric CO signals are inevitably mixed with contributions from the free troposphere (Jiang et al., 2013; Tang et al., 2024), and thus satellite-derived CO fields are more susceptible to influences from long-range transport, making it difficult to isolate local source signals. On the other hand, surface CO measurements suffer from representation errors when compared to grid-based model simulations. Monitoring stations, particularly in urban areas, sample air masses that may not be representative of the model grid average (Fang et al., 2024; Fang et al., 2026), and uncertainties in planetary boundary layer mixing further complicate the interpretation (Castellanos et al., 2011; Du et al., 2020). These complementary strengths and weaknesses imply that the discrepancies between their respective inversion results of CO emissions may reflect fundamental differences in what each measurement actually represents. As a preliminary attempt to explore this issue, Tang et al. (2022) compared assimilated CO fields from MOPITT and surface measurements over China (2015-2020) and found that surface observations showed persistently higher concentrations and a steeper declining trend than those inferred from satellite data.

In this study, we move beyond single-region analyses by constructing a unified multi-region assimilation framework spanning China, North America, and Europe for the period



82 2015-2024, assimilating both MOPITT satellite retrievals and ground-based CO measurements
83 from over 3300 stations. This design directly tests a central hypothesis: whether the satellite-
84 surface discrepancy is a region-specific artifact tied to Chinese emissions and monitoring
85 networks, or instead reflects a fundamental structural difference between the two observation
86 platforms. A key methodological innovation is the systematic stratification of surface sites into
87 urban, suburban, rural, and remote categories, enabled by independent mapping rules for each
88 monitoring network, which allows us to quantitatively disentangle the contribution of
89 representation error from systematic biases in emission inventories. Through this lens, we
90 pursue three interconnected objectives: (1) to diagnose the magnitude and spatial structure of
91 the model-observation mismatch in surface CO across continents, and to attribute its drivers
92 via the urban-to-rural gradient; (2) to conduct cross-validation assimilation experiments that
93 test how surface-data assimilation improves in situ agreement while potentially altering column
94 CO consistency with MOPITT; and (3) to assess decadal trends (2015-2024) in CO
95 concentrations, evaluating how the choice of observation platforms shapes the inferred rate of
96 CO change and its sensitivity to background transport vs. local emissions. By integrating these
97 lines of evidence, this study aims to deliver a robust diagnostic for reconciling multi-platform
98 CO observations, offering actionable guidance for future inversion systems that seek to harness
99 the strengths of both satellite and surface measurements while mitigating their respective
100 limitations.

101 The paper is organized as follows: Section 2 describes the multi-platform observations
102 (MOPITT satellite retrievals and ground-based measurements from three continents), the
103 GEOS-Chem model configuration, and the suboptimal Kalman filter assimilation method.
104 Section 3 presents the comparative results and discussion, focusing on the systematic
105 underestimation of modeled surface CO and the role of representation error, the contrasting
106 impacts of satellite vs. surface data assimilations, and the decadal trends in CO concentrations.



107 Conclusions are given in Section 4.

108

109 **2. Data and Methodology**

110 **2.1 MOPITT CO Retrievals**

111 The MOPITT was launched aboard the NASA Terra spacecraft on December 18, 1999.

112 Terra operates in a sun-synchronous polar orbit at an altitude of approximately 705 km and

113 crosses the equator at about 10:30 local time. MOPITT has a cross-track scan width of

114 approximately 612 km, with an individual pixel footprint of about 22 km × 22 km. The

115 MOPITT CO data used in this study are from the version 9J joint retrieval product. This product

116 retrieves vertical CO profiles using an optimal estimation approach that combines radiance

117 information from the thermal infrared channel (TIR; 4.7 μm) and the near-infrared channel

118 (NIR; 2.3 μm) (Worden et al., 2010; Deeter et al., 2022). The retrieved CO volume mixing

119 ratios are reported as layer averages at 10 pressure levels, including the surface, 900, 800, 700,

120 600, 500, 400, 300, 200, and 100 hPa. The relationship between the retrieved CO profile and

121 the true atmospheric state can be expressed as $\mathbf{z}_{\text{ret}} = \mathbf{z}_a + \mathbf{A}(\mathbf{z} - \mathbf{z}_a) + \mathbf{G}_\epsilon$, where $\mathbf{z}_{\text{ret}}$ is the

122 retrieved CO profile, $\mathbf{z}_a$ is the MOPITT a priori profile, $\mathbf{z}$ is the true atmospheric state, $\mathbf{A}$

123 is the averaging kernel matrix, which characterizes the sensitivity of the retrieval to the true

124 vertical distribution of CO, and $\mathbf{G}_\epsilon$ represents the retrieval error term. In the data screening

125 procedure, we only retained cloud-free observations with Cloud Description = 2. Since the NIR

126 channel relies on reflected solar radiation, nighttime observations were not included in the

127 analysis.

128 **2.2 Ground-Based Air Quality Observations**

129 This study used hourly ground-based CO concentration observations from China, North

130 America, and Europe. The data were obtained from the China Ministry of Ecology and

131 Environment (MEE) national air quality monitoring network, the United States (US)



132 Environmental Protection Agency (EPA) Air Quality System (AQS), the Canadian National
133 Air Pollution Surveillance (NAPS) program, and the European Environment Agency (EEA)
134 air quality monitoring database. To ensure cross-regional comparability, all CO concentration
135 data units were converted to ppb. Specifically, US and Canadian data in ppm were multiplied
136 by 1000 to convert to ppb, while mg m^{-3} unit data from China and Europe were converted based
137 on the ideal gas law. Considering that reference temperature for observations in China changed
138 from 273 K to 298 K on September 1, 2018, observations from that date onward were converted
139 back to 273 K reference state to ensure consistency in the 2015-2024 time series.

140 After unit conversion, abnormal observations with values ≤ 0 or > 6000 ppb were removed
141 and treated as missing. On this basis, two annual quality control checks were applied at the
142 site-year level. A given year was flagged as an unqualified year if the fraction of valid
143 observation days was below 75% or if the maximum consecutive missing-data gap within that
144 year was ≥ 10 days. To ensure the representation and temporal stability of sites for the long-
145 term analysis over 2015-2024, an additional cross-year consistency check was performed at the
146 site level. A site was included in the long-term analysis only if at least 80% of its 10 years were
147 qualified years, and the full 10-year observation record was retained for such sites. After quality
148 control, the site observations were spatiotemporally paired with the corresponding GEOS-
149 Chem grids. Finally, 1931 long-term stable sites in China, 430 in North America, and 984 in
150 Europe were retained. To characterize the spatial representation of ground monitoring sites for
151 CO concentrations, this study assigned a representation scale parameter L in kilometers to
152 each site based on the native metadata fields of each monitoring network. This parameter
153 represents the approximate spatial range over which the observation is considered
154 representative. Due to differences in the classification systems adopted by various monitoring
155 networks, independent mapping rules were specifically constructed based on study in Elbern
156 et al. (2007) (see details in Table S1).



157 **2.3 GEOS-Chem Model**

158 This study used the GEOS-Chem (v14.6.2) three-dimensional global atmospheric
159 chemistry transport model to simulate CO concentrations. The model is driven by MERRA-2
160 reanalysis meteorological data. The global default anthropogenic emissions were taken from
161 the Community Emissions Data System for CMIP6 (CEDS-CMIP6) inventory (Hoesly et al.,
162 2018). For Asia, these emissions were replaced by MIX v2 (Li et al., 2024). Over the US,
163 anthropogenic emissions were taken from the 2016 National Emissions Inventory (NEI2016),
164 while over Europe the global CEDS-CMIP6 inventory was retained. Biomass burning
165 emissions were obtained from the Global Fire Emissions Database version 5 (GFED5) (van der
166 Werf et al., 2025). Biogenic volatile organic compound (BVOC) emissions were calculated
167 using the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN v2.1)
168 (Guenther et al., 2006). The simulations were conducted using a nested-domain configuration
169 with a horizontal resolution of $0.5^\circ \times 0.625^\circ$. A CO-only simulation was performed, in which
170 the chemistry, emissions, transport, and deposition processes of CO were calculated. The
171 chemical loss of CO is primarily controlled by oxidation by OH, and the OH fields used here
172 were obtained from archived monthly mean concentrations from a full-chemistry GEOS-Chem
173 simulation. The lateral boundary conditions for the nested domain were provided by a $4^\circ \times 5^\circ$
174 global GEOS-Chem simulation and updated every 3 h to represent the influence of transport
175 from outside the regional domain on CO concentrations within the simulation region.

176 **2.4 Kalman Filter Assimilation Method**

177 This study assimilated MOPITT and ground-based CO observations using the suboptimal
178 Kalman filter method to optimize CO concentration fields, as described by Tang et al. (2022).
179 Briefly, the model first advances the analysis field from the previous time step to obtain the
180 background field, $\mathbf{x}_t^b = \mathbf{M}(\mathbf{x}_{t-1}^a)$, and then corrects the CO concentration through the Kalman
181 update, $\mathbf{x}_t^a = \mathbf{x}_t^b + \mathbf{G}_t(\mathbf{y}_t - \mathbf{K}_t\mathbf{x}_t^b)$, where $\mathbf{y}_t$ is the observation vector, $\mathbf{K}_t$ is the



182 observation operator, and $\mathbf{G}_t$ is the Kalman gain. The Kalman gain is defined as $\mathbf{G}_t =$
183 $\mathbf{S}_t^b \mathbf{K}_t^T (\mathbf{K}_t \mathbf{S}_t^b \mathbf{K}_t^T + \mathbf{S}_\epsilon)^{-1}$, where $\mathbf{S}_t^b$ and $\mathbf{S}_\epsilon$ are the model background error covariance and
184 observation error covariance, respectively.

185 The assimilation experiment started on July 1, 2014, to generate optimized initial
186 conditions for January 1, 2015. Thereafter, a 1 h assimilation window was used to update the
187 model CO concentration field hourly and advance it to the next time step. The MOPITT
188 observation error was determined from the error covariance matrix provided in the product.
189 The ground-based observation error included both measurement error and representation error.
190 The measurement error was calculated as $\epsilon_0 = \epsilon_{\max} + 0.0075\Pi_0$, where $\epsilon_{\max} = 6$ ppb and
191 Π_0 is the observed CO concentration. The representation error was calculated as $\epsilon_r =$
192 $\gamma\epsilon_0\sqrt{\Delta l/L}$, where $\gamma = 0.5$, Δl is the model horizontal resolution, and L is the site
193 representation scale (see details in Table S1). The total ground-based observation error was
194 defined as $\epsilon = \sqrt{\epsilon_0^2 + \epsilon_r^2}$. To reduce single-site representation errors, multiple ground-based
195 observations within the same model grid cell were combined into grid-scale observations using
196 the super-observation approach (Miyazaki et al., 2017; Feng et al., 2020b).

197

198 **3. Results and Discussions**

199 **3.1 Discrepancy between modeled and observed CO concentrations**

200 **3.1.1 Systematic underestimation in modeled surface CO**

201 We first diagnose the magnitude and spatial structure of the model-observation mismatch
202 in surface CO concentrations across the three continents. As a starting point, we evaluate the
203 spatial distribution of CO concentrations from the GEOS-Chem a priori simulations (Figs. 1a-
204 c). The model successfully reproduces large-scale spatial gradients of CO concentrations,
205 revealing contrasts in emission intensity across the three continents. In China (Fig. 1a), the a
206 priori CO field exhibits a pronounced pollution dome, with peak concentrations exceeding 400



207 ppb concentrated over the North China Plain (NCP), Yangtze River Delta (YRD), and Sichuan
208 Basin (SCB). In contrast, North America (Fig. 1b) shows a more subdued pollution pattern.
209 Localized enhancements occur along the West Coast and in the Northeastern megalopolises,
210 where modeled CO concentrations can reach 200 ppb. Europe (Fig. 1c) presents a complex,
211 multi-center distribution driven by its dense network of urban and industrial centers. The
212 highest a priori CO concentrations, exceeding 150 ppb, are found over the Po Valley, Benelux
213 region, and parts of Central Europe.

214 Despite these seemingly reasonable large-scale patterns, a dramatic discrepancy emerges
215 when the a priori CO fields are evaluated against surface observations (Figs. 1d-f). The median
216 of observation-to-model (Obs/Mod) ratios reaches 1.82 (mean: 2.08) in China (Fig. 2a),
217 consistent with the suggested underestimation of CO emissions by approximate 180-250%
218 (Feng et al., 2020b; Jia et al., 2025). Particularly high ratios (>4.0) appear in western,
219 northeastern, and some background stations (Fig. 1d). This spatial pattern of bias is further
220 quantified in the scatter relationship between simulations and observations (Figs. 3a-d), which
221 shows a correlation coefficient (R) of 0.31, a regression slope (k) of 0.22 and mean bias of -
222 331.4 ppb over the entire China domain (Table 1.1); as well as 0.47 (R), 0.24 (k) and -399.8
223 ppb (bias) in the NCP; 0.28 (R), 0.43 (k) and -139.6 ppb (bias) in the YRD; and 0.43 (R), 0.28
224 (k) and -257.8 ppb (bias) in the SCB (Table S2.1), respectively.

225 Moving to North America, the median Obs/Mod ratio is 1.65 (mean: 1.92), lower than
226 that in China (Fig. 2b). Extreme underestimation is rare, with only six sites showing ratios >5.0 .
227 Moderate underestimation (ratios $\sim 2-4$) prevails across most of the continent, with slightly
228 elevated ratios along the West Coast and in some urbanized areas. Over the entire North
229 American domain, the R is 0.23, the k is 0.07, and the mean bias is -198.9 ppb (Table 1.1). A
230 similar yet more severe pattern of underestimation characterizes Europe. The Obs/Mod ratio
231 (Fig. 2c) is the highest among the three continents: the median is 2.25 (mean: 2.52), with 57



232 sites ($\sim 5.7\%$) having ratios above 5.0, predominantly located in the Apennine Peninsula. The
233 consistency for Europe is the lowest among the three regions, with $R = 0.29$, $k = 0.03$, and a
234 mean bias of -244.7 ppb (Table 1.1). While China exhibits the largest absolute negative mean
235 bias (-331.4 ppb) due to its higher ambient CO concentrations, it also demonstrates the largest
236 slope ($k = 0.22$). This is significantly larger than the slopes observed in North America ($k =$
237 0.07) and Europe ($k = 0.03$).

238 **3.1.2 Role of site representation error: stratification by station type**

239 To further investigate the underlying causes of model underestimation, we stratified
240 surface sites by type, i.e., urban, suburban, rural, and remote, across the three continents. This
241 classification allows us to assess how representation error, arising from the coarse resolution
242 of the model relative to local emission gradients, contributes to the systematic bias. Notably,
243 China's monitoring network includes only urban and suburban station types, whereas North
244 America and Europe offer a more complete spectrum of site classifications (urban, suburban,
245 rural, and remote). This contrast motivates our extension of the analysis to the latter two
246 continents, enabling a more comprehensive evaluation of the role of site representativeness. In
247 China, both urban and suburban sites exhibit nearly twofold underestimation (Table 2): the
248 median and mean ratios are 1.80 and 2.05 for urban sites, and 1.96 and 2.27 for suburban sites,
249 respectively.

250 By contrast, a clear gradient emerges in North America with median ratios of 1.77, 1.64,
251 1.43, and 1.21 across urban, suburban, rural, and remote sites, respectively (Table 2), which
252 means that from urban sites to rural sites, the model's negative bias was reduced by 44%; while
253 from urban sites to remote sites, the model's negative bias was reduced by 73%. Similarly, the
254 median ratios are 2.30, 2.17, 1.55, and 0.94 across urban, suburban, rural, and remote sites in
255 Europe, respectively (Table 2), which means that from urban sites to rural sites, the model's
256 negative bias was reduced by 58% (comparable to that of North America); while the model's



negative bias completely disappeared at remote sites (due to the limited number of sites, the reliability of the analysis at remote sites is limited in Europe). Since this site type sequence typically corresponds to decreasing influence from local anthropogenic emissions, this result suggests that model-observation discrepancies in surface CO concentrations may diminish as local emission impacts weaken. It is consistent with the smoothing effect of coarse-resolution, implying that representation error is an important source of underestimation in model surface CO concentrations.

3.1.3 Evidence from model-observation discrepancy in column CO

We have suggested that the model-observation discrepancy in surface CO concentrations could be partially attributed to representation error, here we further compare the modeled CO column concentrations against satellite retrievals to provide an independent, large-scale evaluation. Figs. 1g-i show the ratios between MOPITT and modeled CO column concentrations (MOP/Mod) across three continents. In China, the ratios of MOP/Mod are greater than 1 in the NCP and Northeast China, reaching approximately 1.2-1.4 in some areas, while being less than 1 in other regions. In North America, the ratios are greater than 1 in northern regions. In Europe, the ratios are greater than 1 in the eastern and northern regions. Overall, the R is 0.73, the k is 0.62, and the mean bias is 6.8 ppb in China (Table 1.2), as well as 0.30 (R), 0.14 (k) and 3.1 ppb (bias) in North America; and 0.22 (R), 0.08 (k) and 1.8 ppb (bias) in Europe. Similar to surface CO concentrations, the modeled CO column concentrations show larger R and slope when compared with satellite observations over China than those over North America and Europe.

Comparing the findings of Section 3.1.2 with those of this section yields a coherent picture. Ground-based observations indicate that modeled surface CO is strongly biased low (-198.9 to -331.4 ppb across three continents, Table 1.1). In contrast, the column CO comparison reveals no systematic underestimation; instead, biases are slightly positive (1.8-6.8 ppb across



three continents, Table 1.2). Because column CO integrates the entire atmosphere, it is less sensitive to local emission gradients that dominate the signal at surface stations but occupy only a small fraction of a model grid cell. This dichotomy thus provides evidence supporting the important role of site representation error in the underestimation of modeled surface CO concentrations.

Aside from the overall positive bias of modeled CO column concentrations relative to satellite observations, the simulation-observation discrepancies exhibit noticeable regional variations: In China's highly industrialized NCP, modeled CO column concentrations show a negative bias of -16.6 ppb compared to satellite observations (Table S2.2); Similarly, the bias declines from continental averages to near-zero values in the Northeast US (3.1 to -0.6 ppb) and Central Europe (1.8 to 0.4 ppb) (Tables S3.2, S4.2). That is, in regions where local anthropogenic emissions exert strong control on surface and column CO concentrations (e.g., the NCP domain in China), the column bias tends to converge toward the surface bias. Conversely, in most other regions, the column comparison appears to be more influenced by large-scale signals (Figs. 1g-i).

3.2 Impacts of assimilations on atmospheric CO concentrations

3.2.1 Cross-validation of satellite and surface data assimilation

The preceding analysis suggests that the discrepancy between surface and column CO stems from a structural mismatch between point-like surface measurements and grid-scale satellite retrievals. To test this hypothesis directly, we now perform separate assimilation experiments using surface and satellite observations, and cross-validate their impacts on the resulting CO fields. As we expected, assimilation of MOPITT CO column data significantly improved the consistency between simulations and MOPIT observations: the a posteriori bias is close to zero in most regions, with slight positive biases remaining in a few remote areas, such as the Tibetan Plateau and low-latitude ocean areas (Figs. 5g-i). Scatter statistics also



307 indicate a marked improvement in consistency (Fig. 4): the R value increased from 0.22-0.73
308 to 0.72-0.87, and bias decreased from 1.8-6.8 ppb to 1.0-3.9 ppb across the three continents
309 (Table 1.5). The R value showed even more substantial increases in various subregions: in the
310 highly industrialized NCP, it rose from 0.11 to 0.71 (Table S2.5); in the Northeast US and
311 Middle US, it increased from negative 0.06-0.02 to positive 0.67-0.75 (Table S3.5). Compared
312 to assimilating satellite data, assimilating surface observation data brings a greater
313 improvement in the agreement between the analysis field and surface observations (Figs. 5a-
314 c). The R value increased from 0.23-0.31 to 0.94-0.95, and bias decreased from negative 331.4-
315 198.9 ppb to negative 80.1-60.7 ppb across the three continents (Table 1.3).

316 However, an analysis field closer to surface observations does not necessarily imply
317 better agreement with satellite observations. Cross-validation reveals that surface-based station
318 assimilation introduces positive biases relative to MOPITT data (Figs. 5d-f). Compared to
319 MOPITT satellite observations (sampled at locations of surface stations), assimilating surface
320 observations causes the bias of modeled CO column concentrations to increase from 1.8-6.8
321 ppb to 5.4-27.1 ppb across three continents (Table 1.4). This result is consistent with the
322 important role of site representation error in the difference between modeled and observed
323 surface CO concentrations: the assimilations add positive CO increments to surface layer to
324 reduce negative bias at surface level, and these positive CO increments are transported
325 vertically into the free troposphere, ultimately leading to an overall positive bias relative to
326 satellite column observations.

327 **3.2.2 Regional mechanisms behind surface assimilation impacts on CO columns**

328 The impact of surface observation assimilation on CO columns exhibits noticeable
329 regional differences: in China and Europe, assimilating surface observations leads to a
330 significant increase in the bias relative to satellite observations (Table 1.4), from 6.8 ppb to
331 27.1 ppb (China) and from 1.8 ppb to 10.5 ppb (Europe). However, in North America,



332 assimilating surface observations results in only a slight increase in the bias, from 3.1 ppb to
333 5.4 ppb. Why is there such a difference between North America and China/Europe? To
334 understand the source of this discrepancy, Fig. 6 illustrates the impact of surface observation
335 assimilation on atmospheric CO concentrations at different altitudes. In China and Europe,
336 surface observation assimilation results in widespread and significant enhancements in surface
337 CO concentrations (Figs. 6a, 6k), whereas in North America, both the magnitude and spatial
338 extent of these enhancements are notably smaller (Fig. 6f). This weaker surface enhancement
339 in North America can be attributed to two interrelated factors. First, the model's systematic
340 underestimation of surface CO is less severe in North America than in China or Europe (Fig.
341 2), reducing the magnitude of positive increments needed. Second, the sparser surface
342 observation network in North America (430 stations vs. 1931 in China and 984 in Europe)
343 further limits the total positive CO increments introduced by assimilation.

344 The weaker column impact over North America arises from additional factors beyond
345 those controlling surface increments. As noted by Jiang et al. (2015), the vertical transport of
346 surface CO into the free troposphere occurs slower in North America than in East Asia,
347 suppressing the upward propagation of these increments. Furthermore, transboundary transport
348 further modulates the regional imprint of surface assimilation. North America and China lie on
349 similar latitudinal bands, meaning that the free tropospheric CO over North America is strongly
350 influenced by the Asian outflow, a dominant global source of CO. In this context, locally
351 emitted CO from North American surface sources competes with a strong imported background,
352 diluting the vertical propagation signal of positive increments added by surface assimilation.
353 Conversely, European surface CO competes primarily with the North American outflow, where
354 the two source strengths are more comparable. Therefore, in Europe, the additional CO from
355 surface assimilation is less diluted by the background flow, allowing a more effective upward
356 propagation. The combined effect of these factors is that the impact of positive CO increments



357 resulting from surface observation assimilation can effectively propagate upward to the 400
358 hPa level in East Asia (Fig. 6d) and to the 600 hPa level in Europe (Fig. 6m), but are largely
359 confined to the lower troposphere in North America (Figs. 6f-g).

360 **3.3 Decadal trends in CO concentrations in observations and assimilations**

361 Given the structural divergence revealed by the assimilation experiments, we next
362 examine whether this divergence also manifests in the decadal trends of CO concentrations.
363 Surface observations indicate a significant decreasing trend in CO concentrations: in China,
364 surface CO concentration decreased at a rate of $-43.6 \text{ ppb yr}^{-1}$ ($-5.92\% \text{ yr}^{-1}$) (Table 3), with the
365 highly polluted NCP showing a stronger decline of $-68.3 \text{ ppb yr}^{-1}$ ($-8.25\% \text{ yr}^{-1}$) (Table S5). In
366 contrast, weaker trends were observed in North America and Europe: -5.3 ppb yr^{-1} ($-1.51\% \text{ yr}^{-1}$)
367 ¹⁾ in North America and -3.9 ppb yr^{-1} ($-1.02\% \text{ yr}^{-1}$) in Europe. Beyond the overall downward
368 trends, distinct interannual variations in CO concentrations exist across the three regions (Fig.
369 7). Surface CO concentrations in China have generally exhibited a sustained decline.
370 Correspondingly, the declining trends in North America and Europe are largely driven by
371 earlier reductions in CO concentrations: in North America, the overall downward trend in
372 surface CO concentrations leveled off after 2018 (Fig. 7e); in Europe, it plateaued after 2016
373 (Fig. 7i). The interannual evolution of surface CO concentrations broadly aligns with recent
374 CO emission estimates derived from MOPITT CO retrievals: Tang et al. (2026) reported that
375 anthropogenic CO emissions in China continuously decreased between 2015 and 2022, while
376 the declining trends in anthropogenic CO emissions in the US and Europe have stalled in recent
377 years.

378 The Kalman filter assimilation based on surface CO observations effectively captures the
379 observed decreasing trends: the declining trends in assimilated surface CO fields are -5.38%
380 yr^{-1} , $-1.36\% \text{ yr}^{-1}$, and $-1.18\% \text{ yr}^{-1}$ for China, North America, and Europe, respectively. As
381 shown in Figures 8a-c, assimilated surface CO concentrations exhibit an overall decline across



382 China; correspondingly, more pronounced regional differences appear in North America and
383 Europe: scattered hotspots of increasing CO concentrations are found in the south-central US,
384 while Europe displays more country-level variability: regions with rising CO concentrations
385 are clustered in the Iberian Peninsula, Slovakia, Hungary, Serbia, and Turkey, with sporadic
386 occurrences in Czechia and the Apennine Peninsula.

387 MOPITT observations also show a decreasing trend in CO column concentrations from
388 2015 to 2024, but the rate of decrease is significantly lower than that of surface observations.
389 In China, the CO column concentration decreases at a rate of -0.79 ± 0.40 ppb yr⁻¹ (-0.77% yr⁻¹)
390 ¹⁾ (Table 3), with the highly polluted NCP reaching a trend of -2.65 ± 0.64 ppb yr⁻¹ (-1.85% yr⁻¹)
391 ¹⁾ (Table S5). North America and Europe show only insignificant decreasing trends: $-0.11 \pm$
392 0.70 ppb yr⁻¹ (-0.11% yr⁻¹) in North America and -0.21 ± 0.65 ppb yr⁻¹ (-0.21% yr⁻¹) in Europe.
393 Furthermore, CO column concentrations in North America and Europe exhibit substantial
394 interannual variability during 2020-2024 (Fig. 9), with notably low values occurring in 2022.
395 As shown in Fig. S1, compared to 2021, CO column concentrations at high northern latitudes
396 in 2022 were significantly lower, and the extreme low-value areas occurred over high-latitude
397 North America and eastern Siberia, which overlap with high-latitude boreal fire regions (Zheng
398 et al., 2023; Tang et al., 2026). Therefore, the interannual variability of CO column
399 concentrations during 2020-2024 reflects the influence of wildfire emissions.

400 The Kalman filter assimilation (KF-MOPITT) captures the decreasing trend in CO
401 column concentrations reasonably well: the assimilated CO column concentration trends are -
402 0.50% yr⁻¹, -0.11% yr⁻¹, and -0.10% yr⁻¹ for China, North America, and Europe, respectively.
403 As shown in Figs. 8g-i, the assimilated CO column concentration field still shows an overall
404 decreasing trend in China, but the declining trends are markedly reduced in the US and Europe.
405 Correspondingly, the a posteriori CO column concentrations derived from KF-Surface exhibit
406 a significantly higher decreasing trend in China (-0.97% yr⁻¹) than that from satellite



407 assimilation. Although the a posteriori CO column concentration trends in North America and
408 Europe are numerically similar to those from KF-MOPITT, i.e., $-0.15\% \text{ yr}^{-1}$ (US) and -0.14%
409 yr^{-1} (Europe) in KF-Surface, their spatial distributions (Figs. 8d-f) differ markedly from KF-
410 MOPITT: they show a more pronounced decreasing trend in China (Fig. 8d) and also exhibit
411 overall decreasing trends in North America and Europe (Figs. 8e-f). This suggests that the
412 assimilation results from KF-Surface reflect more strongly the influence of local CO emissions.
413 This is also reflected in their interannual evolution: as shown in Fig. 9, the a posteriori CO
414 column concentrations generated by KF-Surface remain roughly stable during 2020-2024,
415 unaffected by high-latitude wildfire emissions.

416

417 **4. Conclusion**

418 This study presents a comprehensive multi-region analysis of atmospheric CO over
419 China, North America, and Europe from 2015 to 2024, leveraging both MOPITT satellite
420 retrievals and ground-based observations. Our results confirm that the model's systematic
421 underestimation of surface CO concentrations is a common feature across all three regions.
422 The median observation-to-model ratio is 1.82 in China, 1.65 in North America, and 2.25 in
423 Europe, indicating a widespread negative bias in modeled surface CO. This bias diminishes
424 progressively from urban to rural sites, by 44% in North America and 58% in Europe, and
425 almost disappears at remote sites, suggesting that representation error is an important driver of
426 the surface-model discrepancy. In contrast, comparisons of column CO between MOPITT and
427 the model reveal no systematic underestimation; biases are slightly positive across all
428 continents (1.8-6.8 ppb).

429 Assimilation experiments illuminate the nature of this divergence. While surface-data
430 assimilation greatly improves agreement with in situ observations ($R > 0.94$), it introduces a
431 positive bias in column CO relative to MOPITT, especially over China (27.1 ppb) and Europe



432 (10.5 ppb). In North America, however, surface assimilation increases the column bias slightly
433 to 5.4 ppb, owing to a combination of weaker model bias, sparser observational networks,
434 slower vertical transport, and dilution by strong transboundary inflow from Asia. These
435 regional differences underscore that the satellite-surface discrepancy is a structurally embedded
436 feature of the observing system, modulated by local meteorology, network density, and
437 background transport. Consistent with this structural divergence, decadal trend analyses show
438 that surface CO concentrations declined sharply in China ($-5.92\% \text{ yr}^{-1}$), while trends in North
439 America and Europe were plateaued after 2018 and 2016, respectively. Satellite-derived
440 column trends were substantially weaker, particularly in North America and Europe, where
441 recent interannual variability was dominated by boreal wildfire emissions. Thus, assimilated
442 fields from surface observations captured local emission-driven trends, while satellite-based
443 assimilations reflected broader, transport-influenced signals.

444 Taken together, our multi-region analysis clarifies the complementary roles of surface
445 and satellite CO observations. Surface measurements capture local emission signals and their
446 temporal trends with high fidelity, yet their quantitative use for emission strength estimation is
447 hampered by unresolved representation errors. Satellite retrievals, while insensitive to such
448 representativity issues, exhibit dampened temporal trends due to the influence of broad-scale
449 background transport. These intrinsic differences imply that neither observation type alone may
450 not be able to fully characterize CO emissions. We therefore advocate for future inversion
451 studies to jointly utilize both observation platforms, systematically comparing and reconciling
452 the emission estimates derived from surface and satellite data. Such a synergistic framework
453 will enable a more robust and self-consistent assessment of CO emissions, leveraging the
454 strengths of each observation type while mitigating their respective limitations.

455

456 **Data availability:** The MEE data can be downloaded from <https://quotsoft.net/air/>. The AQS



457 data can be downloaded from https://aqsweb.airdata/download_files.html. The
458 NAPS data can be downloaded from <https://donnees.azurewebsites.net/>.
459 The EEA data can be downloaded from [https://eeadmz1-downloads-](https://eeadmz1-downloads-webapp.azurewebsites.net/)
460 [webapp.azurewebsites.net/](https://eeadmz1-downloads-webapp.azurewebsites.net/). The MOPITT data can be downloaded from
461 <https://asdc.larc.nasa.gov/data/MOPITT/>. The GEOS-Chem model (version 14.6.2) can be
462 downloaded from <https://github.com/geoschem/geos-chem/releases#release-14.6.2>.
463

464
465 **Author Contributions:** Z.J. designed the research. Z.T. performed the research. Z.J. and Z.T.
466 wrote the manuscript. All authors contributed to discussions and editing the manuscript.

467
468 **Competing interests:** The authors declare that they have no conflict of interest.
469

470 **Acknowledgments:** We thank the providers of the MOPITT CO data. We thank the Ministry
471 of Ecology and Environment of China for providing the MEE surface measurements; US
472 Environmental Protection Agency for providing the AQS surface observations; the
473 Environment and Climate Change Canada for providing the NAPS data, and the European
474 Environment Agency for providing the EEA data. This work was supported by the National
475 Natural Science Foundation of China (42277082).
476

477 **References**

478 Castellanos, P., Marufu, L. T., Doddridge, B. G., Taubman, B. F., Schwab, J. J., Hains, J. C.,
479 Ehrman, S. H., and Dickerson, R. R.: Ozone, oxides of nitrogen, and carbon monoxide during
480 pollution events over the eastern United States: An evaluation of emissions and vertical
481 mixing, *J. Geophys. Res.-Atmos.*, 116, D16307, 10.1029/2010jd014540, 2011.
482 Deeter, M., Francis, G., Gille, J., Mao, D., Martínez-Alonso, S., Worden, H., Ziskin, D.,
483 Drummond, J., Commane, R., Diskin, G., and McKain, K.: The MOPITT Version 9 CO



- 484 product: sampling enhancements and validation, *Atmos. Meas. Tech.*, 15, 2325-2344,
485 10.5194/amt-15-2325-2022, 2022.
- 486 Du, Q., Zhao, C., Zhang, M., Dong, X., Chen, Y., Liu, Z., Hu, Z., Zhang, Q., Li, Y., Yuan, R.,
487 and Miao, S.: Modeling diurnal variation of surface PM_{2.5} concentrations over East China
488 with WRF-Chem: impacts from boundary-layer mixing and anthropogenic emission, *Atmos.*
489 *Chem. Phys.*, 20, 2839-2863, 10.5194/acp-20-2839-2020, 2020.
- 490 Elbern, H., Strunk, A., Schmidt, H., and Talagrand, O.: Emission rate and chemical state
491 estimation by 4-dimensional variational inversion, *Atmos. Chem. Phys.*, 7, 3749-3769,
492 10.5194/acp-7-3749-2007, 2007.
- 493 Fang, C., Jiang, Z., Wang, M., Chen, X., Han, W., He, T.-L., and Shen, Y.: Quantifying and
494 correcting systematic discrepancies in the comparison between surface CO observations and
495 simulations, *Atmos. Environ.*, 368, 121769, 10.1016/j.atmosenv.2025.121769, 2026.
- 496 Fang, L., Jin, J., Segers, A., Li, K., Xia, J., Han, W., Li, B., Lin, H. X., Zhu, L., Liu, S., and
497 Liao, H.: Observational operator for fair model evaluation with ground NO₂ measurements,
498 *Geosci. Model Dev.*, 17, 8267-8282, 10.5194/gmd-17-8267-2024, 2024.
- 499 Feng, S., Jiang, F., Wang, H., Wang, H., Ju, W., Shen, Y., Zheng, Y., Wu, Z., and Ding, A.:
500 NO_x Emission Changes Over China During the COVID-19 Epidemic Inferred From Surface
501 NO₂ Observations, *Geophys. Res. Lett.*, 47, e2020GL090080, 10.1029/2020GL090080,
502 2020a.
- 503 Feng, S., Jiang, F., Wu, Z., Wang, H., Ju, W., and Wang, H.: CO Emissions Inferred From
504 Surface CO Observations Over China in December 2013 and 2017, *J. Geophys. Res.-Atmos.*,
505 125, e2019JD031808, 10.1029/2019jd031808, 2020b.
- 506 Fortems-Cheiney, A., Broquet, G., Potier, E., Plauchu, R., Berchet, A., Pison, I., Denier van
507 der Gon, H., and Dellaert, S.: CO anthropogenic emissions in Europe from 2011 to 2021:
508 insights from Measurement of Pollution in the Troposphere (MOPITT) satellite data, *Atmos.*
509 *Chem. Phys.*, 24, 4635-4649, 10.5194/acp-24-4635-2024, 2024.
- 510 Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of
511 global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and
512 Aerosols from Nature), *Atmos. Chem. Phys.*, 6, 3181-3210, 10.5194/acp-6-3181-2006, 2006.
- 513 Han, H., Liu, J., Yuan, H., Jiang, F., Zhu, Y., Wu, Y., Wang, T., and Zhuang, B.: Impacts of
514 Synoptic Weather Patterns and their Persistency on Free Tropospheric Carbon Monoxide
515 Concentrations and Outflow in Eastern China, *J. Geophys. Res.-Atmos.*, 123, 7024-7046,
516 10.1029/2017jd028172, 2018.
- 517 Han, W., He, T.-L., Tang, Z., Wang, M., Jones, D., and Jiang, Z.: A comparative analysis for
518 a deep learning model (hyDL-CO v1.0) and Kalman filter to predict CO concentrations in
519 China, *Geosci. Model Dev.*, 15, 4225-4237, 10.5194/gmd-15-4225-2022, 2022.
- 520 Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T.,
521 Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N.,
522 Kurokawa, J. I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.:
523 Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the
524 Community Emissions Data System (CEDS), *Geosci. Model Dev.*, 11, 369-408,
525 10.5194/gmd-11-369-2018, 2018.



- 526 Jeffery, P. S., Drummond, J. R., Zou, J., and Walker, K. A.: Identifying episodic carbon
527 monoxide emission events in the MOPITT measurement dataset, *Atmos. Chem. Phys.*, 24,
528 4253-4263, 10.5194/acp-24-4253-2024, 2024.
- 529 Jia, M., Jiang, F., Evangeliou, N., Eckhardt, S., Stohl, A., Huang, X., Shen, Y., Feng, S., He,
530 W., Wang, J., Wang, H., Wu, M., Ju, W., and Ding, A.: Anthropogenic Carbon Monoxide
531 Emissions During 2014–2020 in China Constrained by In Situ Ground Observations, *J.*
532 *Geophys. Res.-Atmos.*, 130, e2024JD042371, 10.1029/2024jd042371, 2025.
- 533 Jiang, Z., Jones, D. B. A., Worden, H. M., Deeter, M. N., Henze, D. K., Worden, J., Bowman,
534 K. W., Brenninkmeijer, C. A. M., and Schuck, T. J.: Impact of model errors in convective
535 transport on CO source estimates inferred from MOPITT CO retrievals, *J. Geophys. Res.-*
536 *Atmos.*, 118, 2073-2083, 10.1002/jgrd.50216, 2013.
- 537 Jiang, Z., Jones, D. B. A., Worden, H. M., and Henze, D. K.: Sensitivity of top-down CO source
538 estimates to the modeled vertical structure in atmospheric CO, *Atmos. Chem. Phys.*, 15, 1521-
539 1537, 10.5194/acp-15-1521-2015, 2015.
- 540 Li, M., Kurokawa, J., Zhang, Q., Woo, J. H., Morikawa, T., Chatani, S., Lu, Z., Song, Y., Geng,
541 G., Hu, H., Kim, J., Cooper, O. R., and McDonald, B. C.: MIXv2: a long-term mosaic
542 emission inventory for Asia (2010–2017), *Atmos. Chem. Phys.*, 24, 3925-3952, 10.5194/acp-
543 24-3925-2024, 2024.
- 544 Miyazaki, K., Eskes, H., Sudo, K., Boersma, K. F., Bowman, K., and Kanaya, Y.: Decadal
545 changes in global surface NO_x emissions from multi-constituent satellite data assimilation,
546 *Atmos. Chem. Phys.*, 17, 807-837, 10.5194/acp-17-807-2017, 2017.
- 547 Tang, W., Gaubert, B., Emmons, L., Ziskin, D., Mao, D., Edwards, D., Arellano, A., Raeder,
548 K., Anderson, J., and Worden, H.: Advantages of assimilating multispectral satellite retrievals
549 of atmospheric composition: a demonstration using MOPITT carbon monoxide products,
550 *Atmos. Meas. Tech.*, 17, 1941-1963, 10.5194/amt-17-1941-2024, 2024.
- 551 Tang, Z., Chen, J., and Jiang, Z.: Discrepancy in assimilated atmospheric CO over East Asia
552 in 2015–2020 by assimilating satellite and surface CO measurements, *Atmos. Chem. Phys.*,
553 22, 7815-7826, 10.5194/acp-22-7815-2022, 2022.
- 554 Tang, Z., Yang, P., Miyazaki, K., Worden, J., Worden, H., Henze, D. K., Jones, D. B. A., and
555 Jiang, Z.: Global CO emissions and drivers of atmospheric CO trends constrained by
556 MOPITT satellite measurements, *Atmos. Chem. Phys.*, 26, 5531-5551, 10.5194/acp-26-
557 5531-2026, 2026.
- 558 van der Werf, G. R., Randerson, J. T., van Wees, D., Chen, Y., Giglio, L., Hall, J., Vernooij,
559 R., Mu, M., Binte Shahid, S., Barsanti, K. C., Yokelson, R., and Morton, D. C.: Landscape
560 fire emissions from the 5th version of the Global Fire Emissions Database (GFED5),
561 *Scientific Data*, 12, 1870, 10.1038/s41597-025-06127-w, 2025.
- 562 Wei, J., Li, Z., Wang, J., Li, C., Gupta, P., and Cribb, M.: Ground-level gaseous pollutants
563 (NO₂, SO₂, and CO) in China: daily seamless mapping and spatiotemporal variations, *Atmos.*
564 *Chem. Phys.*, 23, 1511-1532, 10.5194/acp-23-1511-2023, 2023.
- 565 Worden, H. M., Deeter, M. N., Edwards, D. P., Gille, J. C., Drummond, J. R., and Nédélec, P.:
566 Observations of near-surface carbon monoxide from space using MOPITT multispectral
567 retrievals, *J. Geophys. Res.-Atmos.*, 115, D18314, 10.1029/2010jd014242, 2010.



- 568 Xie, Y., Han, H., and Liu, J.: Spatial and seasonal variations and trends in carbon monoxide
569 over China during 2013–2022, *Atmos. Environ.*, 350, 121163,
570 10.1016/j.atmosenv.2025.121163, 2025.
- 571 Zheng, B., Chevallier, F., Yin, Y., Ciais, P., Fortems-Cheiney, A., Deeter, M. N., Parker, R. J.,
572 Wang, Y., Worden, H. M., and Zhao, Y.: Global atmospheric carbon monoxide budget 2000–
573 2017 inferred from multi-species atmospheric inversions, *Earth Syst. Sci. Data*, 11, 1411–
574 1436, 10.5194/essd-11-1411-2019, 2019.
- 575 Zheng, B., Ciais, P., Chevallier, F., Yang, H., Canadell, J. G., Chen, Y., van der Velde, I. R.,
576 Aben, I., Chuvieco, E., Davis, S. J., Deeter, M., Hong, C., Kong, Y., Li, H., Li, H., Lin, X.,
577 He, K., and Zhang, Q.: Record-high CO₂ emissions from boreal fires in 2021, *Science*, 379,
578 912–917, 10.1126/science.ade0805, 2023.
- 579 Zhou, Y., Mao, H., Demerjian, K., Hogrefe, C., and Liu, J.: Regional and Hemispheric
580 Influences on Temporal Variability in Baseline Carbon Monoxide and Ozone over the
581 Northeast US, *Atmos Environ* (1994), 164, 309–324, 10.1016/j.atmosenv.2017.06.017, 2017.
582



Experiments		Dataset for evaluation	Domain	R	Slope	Bias (ppb)	RMSE (ppb)
T1.1	a priori simulation	Surface	China	0.31	0.22	-331.4	512.8
			North America	0.23	0.07	-198.9	282.2
			Europe	0.29	0.03	-244.7	378.7
T1.2	a priori simulation	MOPITT	China	0.73	0.62	6.8	18.8
			North America	0.30	0.14	3.1	13.7
			Europe	0.22	0.08	1.8	11.4
T1.3	KF-Surface	Surface	China	0.94	0.80	-80.1	157.0
			North America	0.95	0.72	-68.8	102.3
			Europe	0.95	0.71	-60.7	128.9
T1.4	KF-Surface	MOPITT (sampled)	China	0.76	0.98	27.1	33.9
			North America	0.44	0.27	5.4	14.8
			Europe	0.46	0.33	10.5	14.9
T1.5	KF-MOPITT	MOPITT	China	0.87	0.74	3.9	13.0
			North America	0.80	0.55	1.6	8.6
			Europe	0.72	0.45	1.0	8.0

Table 1. Statistical metrics comparing simulations and assimilations against surface and MOPITT data across China, North America and Europe. Three different experiments are performed, namely the GEOS-Chem a priori simulation (T1.1, T1.2), the Kalman Filter assimilation of surface CO observations (T1.3, T1.4), and the Kalman Filter assimilation of MOPITT CO column observations (T1.5). The evaluation employs three distinct datasets: surface measurements, MOPITT retrievals and a sampled MOPITT dataset matching locations of surface stations. Please check Tables S2-S4 for the statistical information of each subregion.

Domain	Variable	All sites	Urban	Suburban	Rural	Remote
China	Number of sites	1931	1703	228	/	/
	Obs/Mod (mean)	2.08	2.05	2.27	/	/
	Obs/Mod (median)	1.82	1.80	1.96	/	/
North America	Number of sites	430	228	130	52	20
	Obs/Mod (mean)	1.92	1.98	2.03	1.62	1.26
	Obs/Mod (median)	1.65	1.77	1.64	1.43	1.21
Europe	Number of sites	984	742	159	70	3
	Obs/Mod (mean)	2.52	2.64	2.33	1.82	0.95
	Obs/Mod (median)	2.25	2.30	2.17	1.55	0.94

Table 2. Obs/Mod ratios of surface CO concentrations over China, North America and Europe in 2015-2024. Surface observation sites are categorized into five types: all sites, as well as urban (including both traffic and urban sites in this Table), suburban, rural and remote sites. Data for China are presented for urban and suburban sites only, as rural and remote data are unavailable.



Experiments		China		North America		Europe	
		Mean	Trend	Mean	Trend	Mean	Trend
Surface (ppb or ppb/year)	a priori simulation	401.3	-6.84 ± 5.27	172.1	-0.20 ± 1.42	140.3	-0.26 ± 0.33
	surface obs	735.3	-43.55 ± 7.51	349.4	-5.29 ± 3.12	385.0	-3.91 ± 4.01
	KF-surface	654.6	-35.21 ± 6.56	287.6	-3.91 ± 2.17	324.3	-3.82 ± 2.59
Column (Xco ppb or ppb/year)	a priori simulation	109.3	-0.28 ± 0.14	98.9	-0.10 ± 0.10	98.9	-0.02 ± 0.07
	MOPITT column	102.4	-0.79 ± 0.40	95.7	-0.11 ± 0.70	97.1	-0.21 ± 0.65
	KF-surface	120.9	-1.17 ± 0.18	100.2	-0.15 ± 0.13	103.6	-0.14 ± 0.13
	KF-MOPITT	106.4	-0.53 ± 0.24	97.4	-0.11 ± 0.42	98.1	-0.10 ± 0.32

Table 3. Averages and trends of surface and column CO concentrations from different experiments and datasets in 2015-2024.

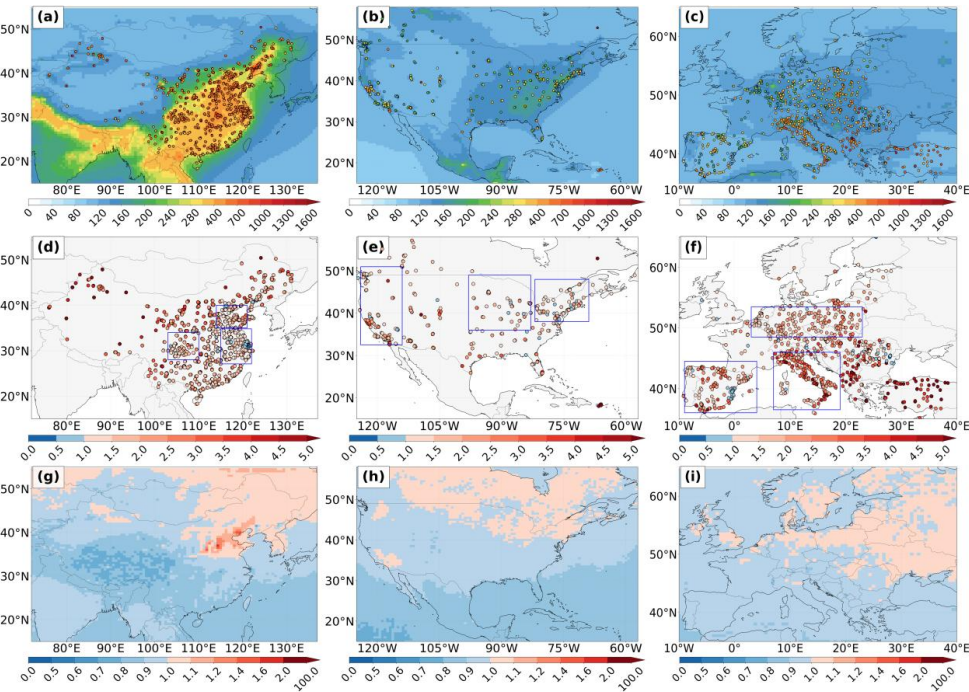


Fig. 1. (a-c) Modeled (contour, a priori simulation) and observed (dotted) surface CO concentrations in 2015-2024 with unit ppb; (d-f) Ratios between observed and modeled surface CO concentrations in 2015-2024. The blue boxes define the subregions of North China Plain (NCP), Yangtze River Delta (YRD), and Sichuan Basin (SCB) for China; Northeast, Middle and West Coast for North America; and Iberian Peninsula, Apennine Peninsula and Central Europe for Europe. (g-i) Ratios between MOPITT and modeled CO column concentrations in 2015-2024.

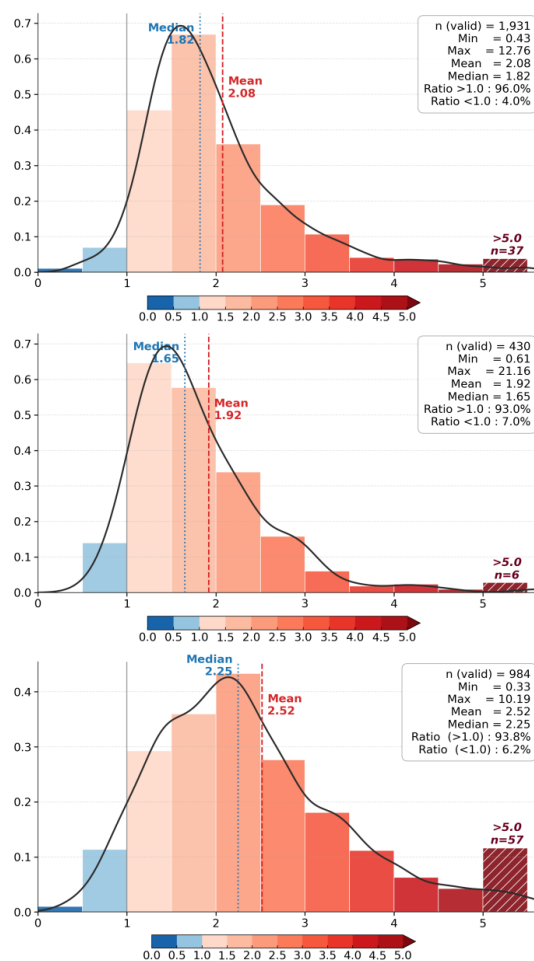


Fig. 2. Distribution of Obs/Mod ratios of surface CO concentrations over China, North America and Europe in 2015-2024. Please check the statistical metrics for different site types in Table 2.

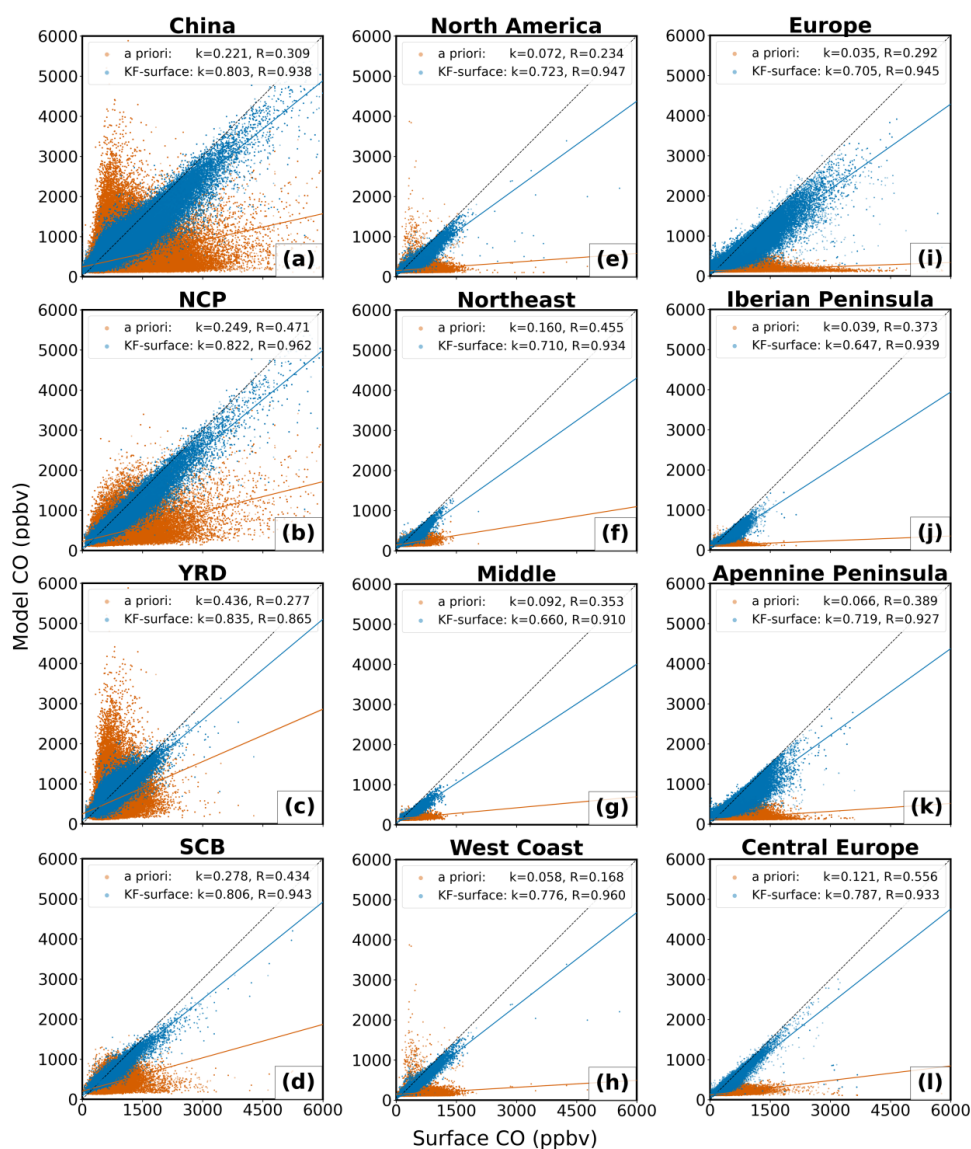


Fig. 3. Relationships of simulations (red) and assimilations (KF-Surface, blue) against surface CO observations across China, North America, and Europe, along with their respective subregions in 2015-2024.

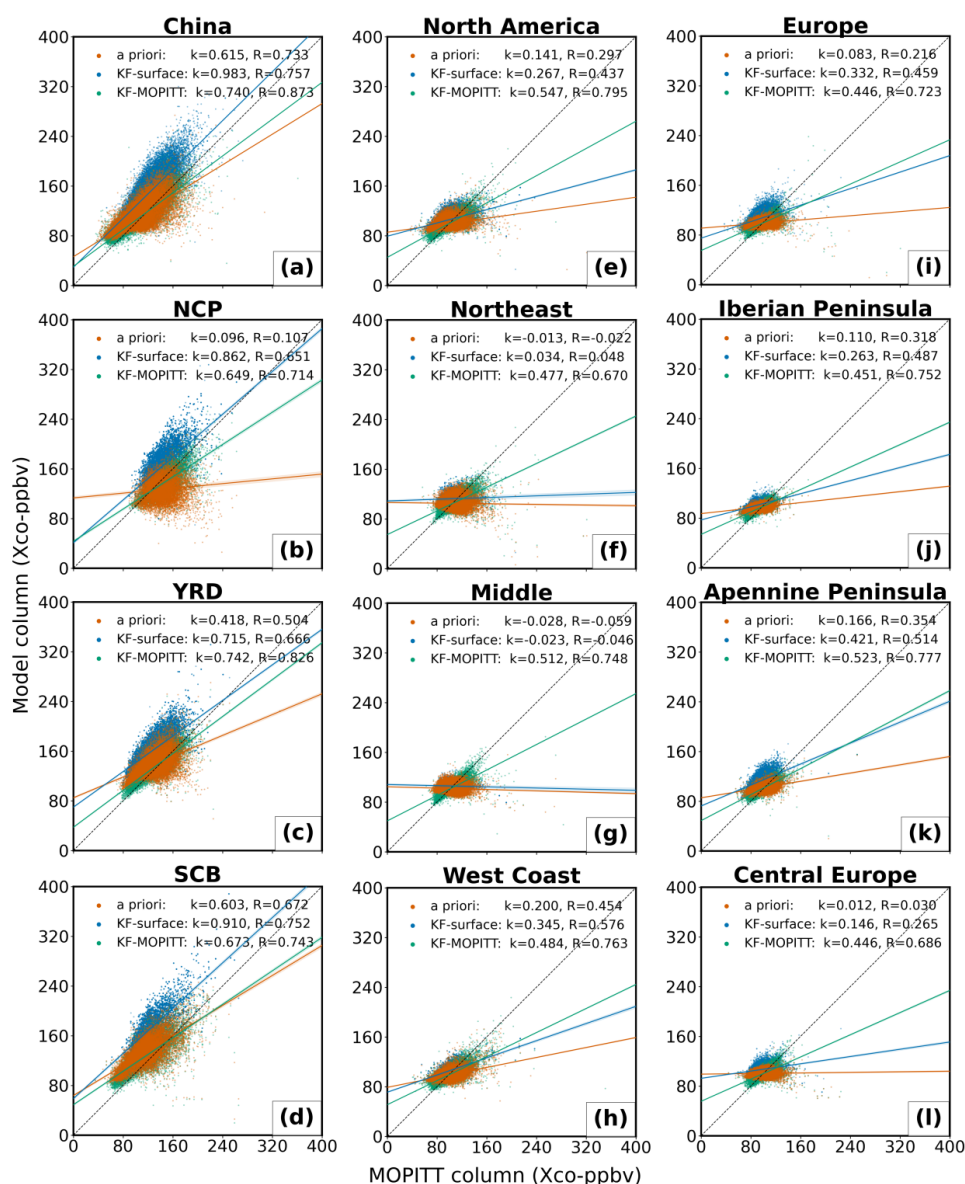


Fig. 4. Relationships of simulations (red) and assimilations against MOPITT CO observations across China, North America, and Europe, along with their respective subregions in 2015-2024. The assimilation here includes the KF-MOPITT (green) and KF-Surface (blue). Note sampled MOPITT dataset matching the locations of surface stations is compared with KF-Surface.

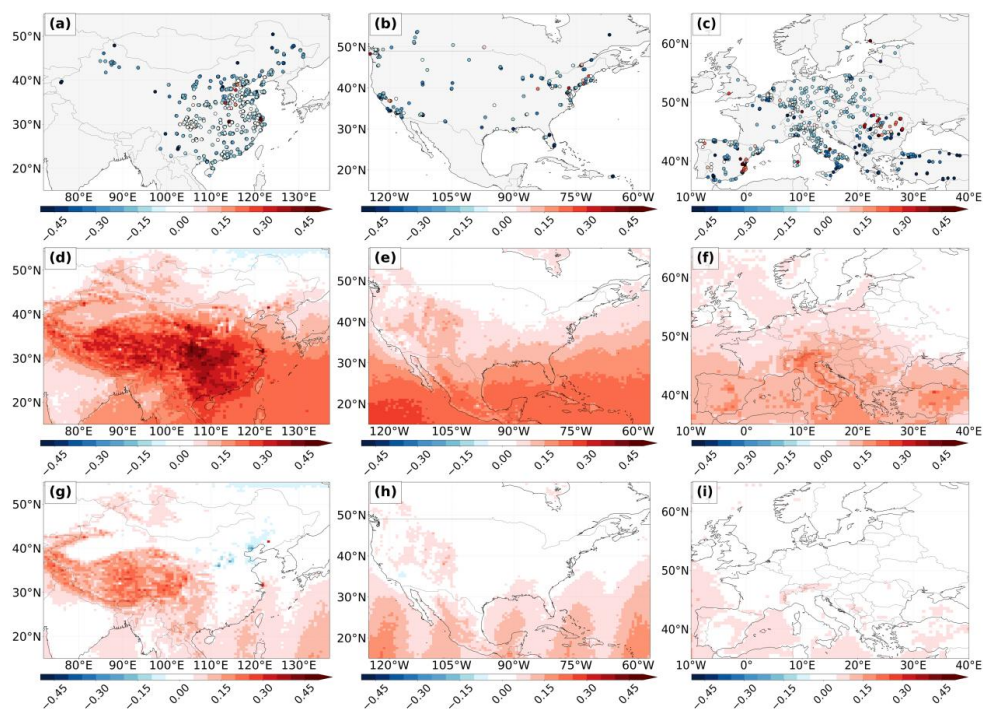


Fig. 5. (a-c) Relative difference of CO concentrations from KF-Surface to surface observations. (d-f) Relative difference of CO concentrations from KF-Surface to MOPITT observations. (g-i) Relative difference of CO concentrations from KF-MOPITT to MOPITT observations. The calculation method for relative difference is $(\text{model} - \text{observation}) / \text{observation}$.

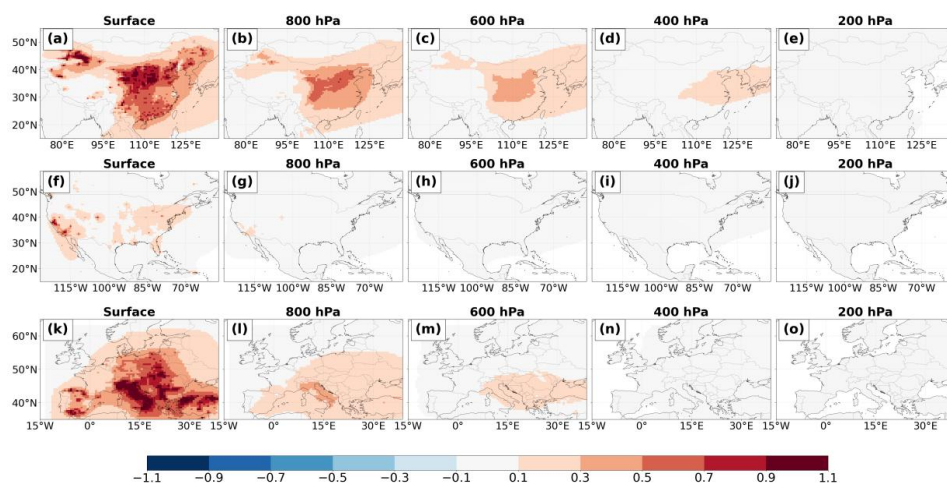


Fig. 6. Relative difference of CO concentrations from KF-Surface to the GEOS-Chem a priori simulation at different vertical levels over (a–e) East Asia, (f–j) North America, and (k–o) Europe. From left to right, the results are shown at the surface, 800 hPa, 600 hPa, 400 hPa, and 200 hPa, respectively. The calculation method for relative difference is $(a \text{ posteriori} - a \text{ priori}) / a \text{ priori}$.

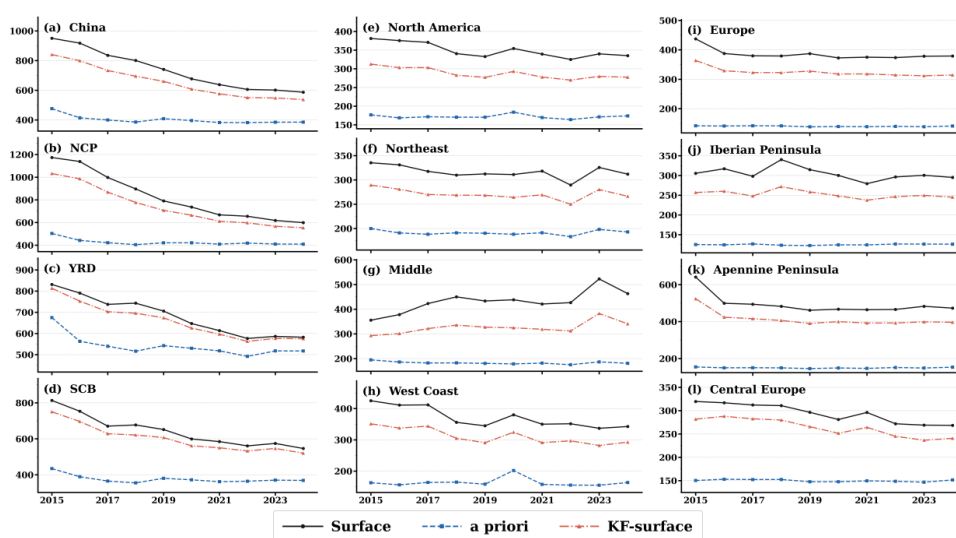


Fig. 7. Surface CO concentrations from surface observations (black), GEOS-Chem a priori simulations (blue) and assimilations of KF-Surface (red).

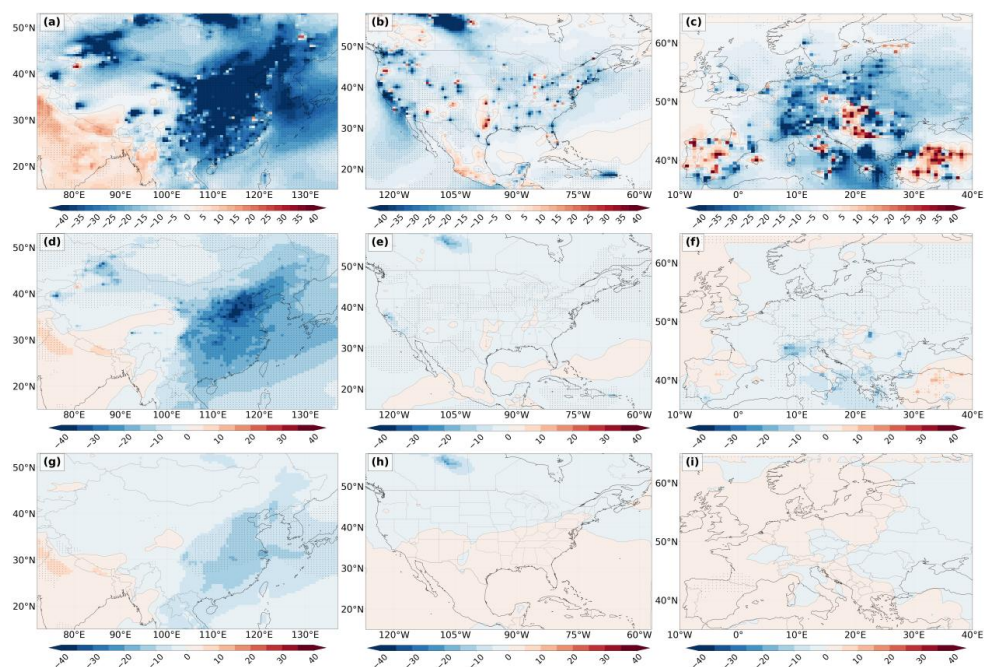


Fig. 8. Trends of surface CO concentrations from KF-Surface (a-c), as well as trends of CO columns from KF-Surface (d-f) and KF-MOPITT (g-i) in 2015-2024.

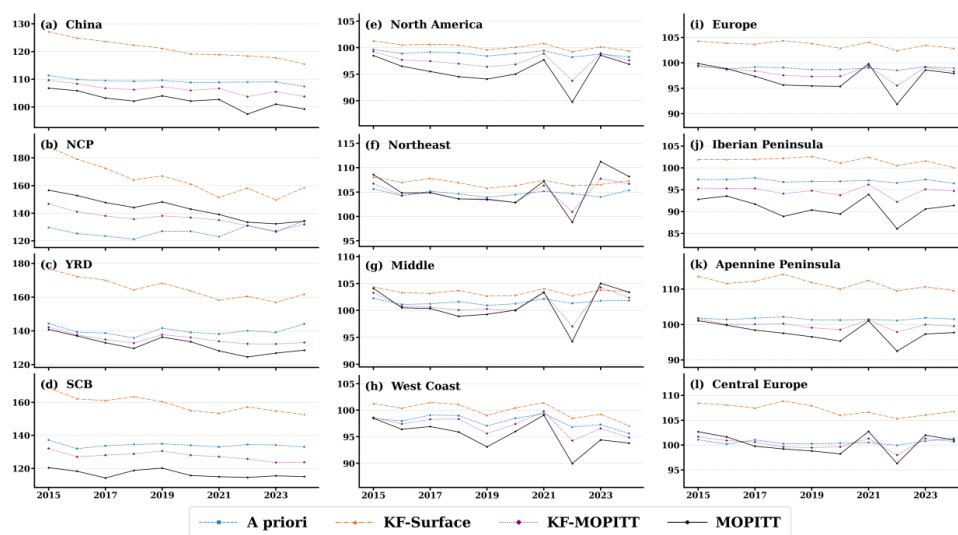


Fig. 9. CO columns from MOPITT observations (black), GEOS-Chem a priori simulations (blue) as well as assimilations of KF-Surface (red) and KF-MOPITT (magenta).