

1 *Remote Sensing of Environment*

2 Supporting Information for

3 **Fossil fuel CO<sub>2</sub> emissions over metropolitan areas from space: a multi-model analysis of**  
4 **OCO-2 data over Lahore, Pakistan**

5  
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8  
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28

29 **Text S1 More details of the FCSI method**

30 **Text S1.1 Background estimation**

31 Two methods were used to calculate the background:

- 32 1. ‘median’: following Schwander et al., (2017) we used the median of the selected  
33 sounding as a background value. This method can be seen as conservative as it may  
34 overestimate the plume size when the plume width is around (or bigger) half the track  
35 length.
- 36 2. ‘coordFit’ : following the idea developed in Ye et al., (2020) we made a planar fit of the  
37 track in two step : a first planar fit is done  $((a_1, b_1, c_1) = \operatorname{argmin}_{(a_1, b_1, c_1)} (X_{\text{CO}_2}(\text{lat}, \text{lon}) -$   
38  $a_1 * \text{lat} - b_1 * \text{lon} - c_1)^2)$ , the retrieved background ( $\text{bckg1} = a_1 * \text{lat} + b_1 * \text{lon} + c_1$ ) is subtracted  
39 to the track, we select the soundings under the mean corrected signal plus one standard  
40 deviation ( $\text{selec} = \{\text{lat}, \text{lon}\} / (X_{\text{CO}_2} - \text{bckg1}) < (\langle \text{bckg1} \rangle + \sigma_{\text{bckg1}})$ ) and make a planar fit on  
41 the selected soundings  $((a_2, b_2, c_2) = \operatorname{argmin}_{(a_2, b_2, c_2)} (X_{\text{CO}_2}(\text{lat}, \text{lon}) - a_2 * \text{lat} - b_2 * \text{lon} - c_2)^2_{(\text{lat}, \text{lon}) \text{ in}}$   
42  $\text{selec})$ . This second planar interpolation is used as background ( $\text{bckg}(\text{lon}, \text{lat}) =$   
43  $a_2 * \text{lat} + b_2 * \text{lon} + c_2$ ) .

44 **Text S1.2 Plume limit detection**

45 Plume limit detection in 2 steps:

- 46 1. smoothing (5\*6 methods): 5 smoothing methods (using a uniform, a median, a wiener, a  
47 gaussian, and a low pass filter) were used on 6 smoothing sizes (from 10 to 25 km wide,  
48 with a 3 km step). For the Wiener filter, the noise was set as the mean of the  $X_{CO_2}$   
49 uncertainty (provided at each sounding in the L2 Lite files) in the track. The detail of the  
50 methods can be found in SciPy documentation  
51 (<https://docs.scipy.org/doc/scipy/reference/> chapter signal for wiener method, ndimage  
52 for uniform, Gaussian and median methods, and fftpack for the low pass filter).
- 53 2. peak identification (2 methods): maximal peak in the smoothed background subtracted  
54 track or closest peak to the estimated effective wind direction – track axis intersection in  
55 the smoothed background subtracted track.

56 All the positive smoothed values surrounding the selected peak are set as a part of the plume.

57

### 58 **Text S1.3 Emission zone definition**

59 The emission zone is defined as the zone upwind from the track. To set the limits of the emission  
60 zone in the cross-wind direction, we report the plume axis on the plume limits and take the  
61 emission within this boundary. The plume axis is set as the axis going from the city center  
62 (defined as the maximum if emission in ODIAC) and the middle of the detected plume. Only the  
63 ODIAC cells above the median ODIAC cell emission of the targeted zone (corresponding to  
64 WRF domain 3) are kept. An illustration can be found in Figure 4. On average, a third of the  
65 targeted zone is captured in the emission zone (28% when using all data subsets and 35% when  
66 using high-quality tracks).

67

68 **Text S1.4 Subset selection**

69 We check manually the selected plume and the coherence between the plume axis (plume center  
70 – city center axis) and the wind direction to define the criterion of acceptance specific to each  
71 date. The criterion is based on two variables: the plume size and the angle between the plume  
72 and the estimated effective wind (see Table S1).

73

74 **Text S1.5 Discussion on the method uncertainty**

75 The error in our methods comes from different steps: the wind evaluation, the background  
76 evaluation, the plume detected limits, the emission zone definition, the measurement error, the  
77 method representativity error, and the spatial representativity of the used inventory.

78

79 Wind angle error: the error related to the wind angle will depend on the angle between the wind  
80 and the track normal vector. For an angle of  $0^\circ$  between the track normal vector and the wind, an  
81 error of  $10^\circ$  on the wind angle will give an error of 1.5% on the result. For an angle of  $60^\circ$   
82 between the track normal vector and the wind, an error of  $10^\circ$  on the wind angle will give an  
83 error of 30% on the result. The error increases with the absolute difference between the wind and  
84 the track normal vector and with the wind error.

85

86 Wind speed error: the emission error should be linearly related to this error (an error of 10% on  
87 the wind modulus will result in an error of 10% on the estimate).

88

89 Background calculation error: this error is proportional to the plume size and the wind speed. An  
90 error of 0.2 ppm (mean difference between the two retrieved background for the good quality  
91 data) on the background for a 30 km plume (classic retrieved plume size) with a 2 m/s wind  
92 speed (mean wind in the PBL for the selected dates) and a 1000 hPa dry surface pressure will  
93 give an error of approximately 180kg/s on the result, which correspond to 30% of the emission  
94 usually detected (600kg/s).

95  
96 Emission zone error: this error is related to the wind angle error and the plume limits error; we  
97 evaluated its spread using 2 masks: a large one (upwind enlarging limits) and the one used in the  
98 study. It gives us an error of approximately 10% of variation in the emission.

99  
100 Method representativity error: our equation is designed for steady state conditions (constant and  
101 homogeneous wind, constant emissions) The deviation of the overpass conditions from this  
102 steady state is the cause of this error. We applied our method for different pseudo OCO-2 track  
103 generated with the WRF simulations a day of highly turning plume (suggesting far from steady  
104 state conditions) and found a variation of 30% in the ratio given the track due to this turning  
105 shape, whereas the uncertainty is only 5% when the plume has a straight shape (suggesting close  
106 from steady state conditions).

107  
108 Plume detection error: using a pseudo track on a WRF simulation, we made the plume size vary  
109 randomly from -15% to +15% (+/- 6km on each side of a 35km plume) to estimate the variation  
110 due to error on the plume limits. It gives a spread of 25% of the mean for the emission

111 estimation. Figure S2 shows the standard deviation of the selected subset for each date. Using a  
112 subset should reduce this source of uncertainty.

113

114 Measurement error: this error is hopefully unbiased and quantified with the  $X_{CO_2}$  uncertainty.

115

116 Spatial representativity of the inventory: to evaluate the emission of Lahore, we use a ratio  
117 calculated on a fraction of the city (or a fraction of the city and its suburbs). Our estimation relies  
118 on the representativity of the inventory. Our subset (by evaluating the emission on different  
119 plume limits and thus different emission zone sizes) should reduce this error.

120

#### 121 **Text S2 OCO-2 two-level quality inspection**

122 The OCO-2  $X_{CO_2}$  soundings are subject to a two-level quality inspection. The inspection results  
123 of each criterion are marked by the variable named “sounding\_flag\_sel” for the level 1 (L1)  
124 product and “xco2\_qf\_bitflag” for the level 2 (L2) product. The final results are marked by the  
125 variable named “xco2\_quality\_flag” in OCO-2 v9r L2 lite files. The “xco2\_quality\_flag” is set to  
126 0 for high-quality soundings which meet all selection criteria. The “xco2\_quality\_flag” is set to 1  
127 for low-quality soundings that failed to meet one or more criteria.

128

129 We extracted “sounding\_flag\_sel” from sounding selection (L2Sel) files. The value of  
130 “sounding\_flag\_sel” could be 0, 1, 2, 32. Soundings marked as “sounding\_flag\_sel = 0” are run  
131 through the L2 full-physics retrieval algorithm. Note that “sounding\_flag\_sel = 0” does not  
132 necessarily mean the sounding will show up in L2 data. If it fails to converge, or the code crashes  
133 on a sounding in some rare cases, it does not show up in L2 data. Also, soundings over water

134 that are not in glint mode sometimes run through L2, but they are never included in L2 files. We  
135 focus on cities on land in this study. Thus, we exclude the cases that “sounding\_flag\_sel” does  
136 not show up in L2 data. The definitions of selection criteria when “sounding\_flag\_sel” equaling  
137 to 1, 2, 32 are listed as follows:

- 138 • flag\_1: The cloud cover exceeded max warn level
- 139 • flag\_2: The cloud cover exceeded warn level cutoff
- 140 • flag\_32: The sounding has invalid geolocation

141  
142 The variable “xco2\_qf\_bitflag” in OCO-2 v9r L2 lite files uses 28 out of the 32 bits of a long  
143 integer to report the result of each test. A bit value of 0 means passed, and 1 means failed. If  
144 they all pass, the value of this variable will be 0, just as for the normal quality flag variable.  
145 Regardless of surface type, the currently-defined 28 bits within this long integer correspond to  
146 the following 28 selection criteria (from bit 0 to 27): defined\_mode, eof3\_3\_rel,  
147 max\_declocking\_wco2, max\_declocking\_sco2, albedo\_slope\_sco2, rms\_rel\_wco2, h2o\_ratio,  
148 co2\_ratio, dp\_o2a, dp\_sco2, co2\_grad\_del, windspeed, dp\_abp, aod\_ice, xco2\_uncertainty,  
149 chi2\_wco2, albedo\_slope\_wco2, surface\_type\_flipped, altitude\_stddev, aod\_total, dws,  
150 albedo\_sco2, rms\_rel\_sco2, aod\_water, ice\_height, aod\_strataer, aod\_oc, aod\_seasalt. In OCO-2  
151 Data Product User’s Guide (JPL, 2018). Only 24 variables are listed at its Page 25. But  
152 definitions of all 28 variables can be found in the user guide. We list definitions of 28 selection  
153 criteria when as follows in alphabet sequence:

- 154
- 155 • albedo\_sco2: Over-land retrievals: Surface reflectance at a reference wavelength in band  
156 3 (2.06  $\mu\text{m}$ ) in the primary scattering geometry (sun->ground->sensor) derived from the

157           retrieved BRDF. Over-water retrievals: Retrieved Lambertian albedo at the band 3  
158           reference wavelength.

- 159           • aod\_seasalt: Retrieved Extinction Optical Depth of sea salt aerosol at 0.755  $\mu\text{m}$ .
- 160           • albedo\_slope\_sco2: Slope of the albedo\_sco2 term with respect to wavenumber.
- 161           • albedo\_slope\_wco2: Slope of the albedo\_wco2 term with respect to wavenumber.
- 162           • altitude\_stddev: The standard deviation of the surface elevation in the target field of  
163           view, in meters.
- 164           • aod\_ice: Retrieved Extinction Optical Depth of cloud ice at 0.755  $\mu\text{m}$ .
- 165           • aod\_oc: Retrieved Extinction Optical Depth of organic carbon at 0.755  $\mu\text{m}$ .
- 166           • aod\_seasalt: Retrieved Extinction Optical Depth of sea salt aerosol at 0.755  $\mu\text{m}$ .
- 167           • aod\_strataer: Retrieved Extinction Optical Depth of stratospheric aerosol at 0.755  $\mu\text{m}$ .
- 168           • aod\_total: Retrieved Extinction Optical Depth of cloud+aerosol at 0.755  $\mu\text{m}$ .
- 169           • aod\_water: Retrieved Extinction Optical Depth of cloud water at 0.755  $\mu\text{m}$ .
- 170           • chi2\_wco2: Reduced chi-squared value of the L2 fit residuals for band 2.
- 171           • co2\_grad\_del: Change (between the retrieved profile and the prior profile) of the co2 dry  
172           air mole fraction difference from the surface minus that at level 13, measured in ppm.  
173           Level 13 is at a pressure  $P = 0.631579 P_{\text{surf}}$ . This variable is used in the  $X_{\text{CO}_2}$  bias  
174           correction over both land and water surfaces.
- 175           • co2\_ratio: Contains the ratio of the retrieved CO2 column from the weak Co2 band  
176           relative to that from the strong CO2 band. This ratio should be near unity. Significant  
177           departure from unity is currently used as a way to flag bad soundings (usually cloud or  
178           aerosol-contaminated). This value has also been footprint corrected using

179 co2\_ratio\_offset\_per\_footprint, and further it has been bias corrected to remove a small  
180 feature-dependent bias.

- 181 • dp\_abp: This is the retrieved surface pressure minus the “best-guess” surface pressure  
182 from the ECMWF forecast model. This has been adjusted for a clear-sky bias as well as  
183 the local surface elevation of the observed footprint. A value of this greater than about 50  
184 hPa absolute value typically indicates cloud or aerosol contamination.
- 185 • dp\_o2a: The difference  $psurf - psurf\_apriori\_sco2$ , in hPa.
- 186 • dp\_sco2: The difference  $psurf - psurf\_apriori\_sco2$ , in hPa. This variable is used in the  
187  $X_{CO_2}$  bias correction over both land and water surfaces.
- 188 • dws: Given by  $aod\_dust + aod\_water + aod\_seasalt$ . This is used in the  $X_{CO_2}$  bias  
189 correction for soundings over land.
- 190 • eof3\_3\_rel: Relative amplitude of the 3rd EOF in the strong CO2 band.
- 191 • h2o\_ratio: Contains the ratio of the retrieved H2O column from the weak CO2 band  
192 relative to that from the strong CO2 band. This ratio should be near unity. Significant  
193 departure from unity is currently used as a way to flag bad soundings (usually cloud or  
194 aerosol-contaminated).
- 195 • ice\_height: Retrieved central pressure of the ice layer, relative to the retrieved surface  
196 pressure.
- 197 • max\_declocking\_sco2: An estimate of the absolute value of the clocking error in the  
198 strong co2 band (used in the clocking correction algorithm that attempts to correct the  
199 L1b radiances for clocking errors). Expressed in percent. Typical values range from 0 to  
200 10%.
- 201 • max\_declocking\_wco2: Same as max\_declocking\_sco2, but for the weak co2 band.

- 202 • rms\_rel\_sco2: RMS of the L2 fit residuals for band 3, relative to the continuum signal, in  
203 percent.
- 204 • rms\_rel\_wco2: RMS of the L2 fit residuals for band 2, relative to the continuum signal,  
205 in percent.
- 206 • surface\_type\_flipped: This field is 0 if the surface type is still maintains the same surface  
207 type designation as in version 8, and 1 otherwise. As a reminder, the L2 algorithm defines  
208 “land” to be  $\geq 80\%$  sounding\_land\_fraction, and “ocean” (or “water”) to be  $\leq 20\%$  for  
209 sounding\_land\_fraction.
- 210 • windspeed: Retrieved surface wind speed (in m/s) from the L2 algorithm, over water  
211 surfaces only.
- 212 • xco2\_uncertainty: The posterior uncertainty in  $X_{CO_2}$  calculated by the L2 algorithm, in  
213 ppm. This is generally 30-50% smaller than the true retrieval uncertainty.

214

### 215 **Text S3 Main causes for L1/L2 OCO-2 data loss due to quality flag filtering**

216 To investigate the causes behind the OCO-2 L1/L2 data losses, we calculated the failing  
217 occurrences of the selection criteria used in quality flags. Figure S6 shows the failing  
218 occurrences of 3 criteria in L1 and 27 criteria in the L2 product. The “defined\_mode” is not  
219 shown in the figure because all soundings pass it over Lahore. See Section Text S2 for the  
220 definitions of the selection criteria. Note that a sounding in L2 data a sounding could fail  
221 multiple criteria simultaneously, hence the total percentage of L2 quality flags is greater than  
222 100%. Similar to Figure 2, we calculated the percentages in boxes with 25-, 50-, 75-, 100-, and  
223 200-km border-to-center distances around cities. The discrepancy in percentages between  
224 selection criteria is significantly greater than the discrepancy between box sizes. For the L1

225 products, about 70% of the low-quality soundings are caused by cloud-related issues. For the L2  
226  $X_{CO_2}$  products, cloud- and aerosol-related criteria cause the greatest loss of data, followed by  
227 topography-related criteria.

228

#### 229 **Text S4 Wind nudging and evaluation**

230 To minimize the  $X_{CO_2}$  errors caused by model transport error, the analysis nudging in Four-  
231 Dimensional Data Assimilation (FDDA) for the outer domain is open ( $grid\_fdda = 1$ ). Figure S7  
232 shows a sample comparison of  $X_{CO_2ff}$  plume locations between nudged and non-nudged  
233 simulations. In the sample case, the  $X_{CO_2ff}$  plume crossed the OCO-2 track with nudging on while  
234 it missed track with nudging off.

235

236 To evaluate the uncertainty of WRF-Chem transport, we compared the model wind speed and  
237 direction outputs with two datasets, which are NCEP ADP Global Surface Observational  
238 Weather Data (NCEP., n.d.) and NCAR Upper Air Database (NCAR n..d). Note that Lahore,  
239 Pakistan is growing fast, but still a less-developing area compared to developed countries. The  
240 meteorological observation data are still lacking. Only one site in WRF-Chem inner domain can  
241 be found in each of the two datasets. The wind speed and direction residuals of WRF-Chem  
242 outputs compared to monitor data are shown in Figure S6. The wind speed residual is  $2.11 \pm 0.38$   
243 m/s ( $1-\sigma$ ) and the wind direction residual is  $10.20 \pm 66.04$  deg ( $1-\sigma$ ). Shahid et al., (2015)  
244 evaluated WRF-Chem simulated surface-layer meteorological parameters with a horizontal  
245 resolution of 1 degree against the NCEP reanalyzed datasets over Pakistan. The mean bias of U-  
246 and V-wind ranged in  $-0.2 - 0.2$  m/s and the normalized mean bias ranged  $-42.9\%$  to  $58.5\%$ . Due  
247 to the lack of studies on wind profile evaluation over Lahore or Pakistan, we also compared our

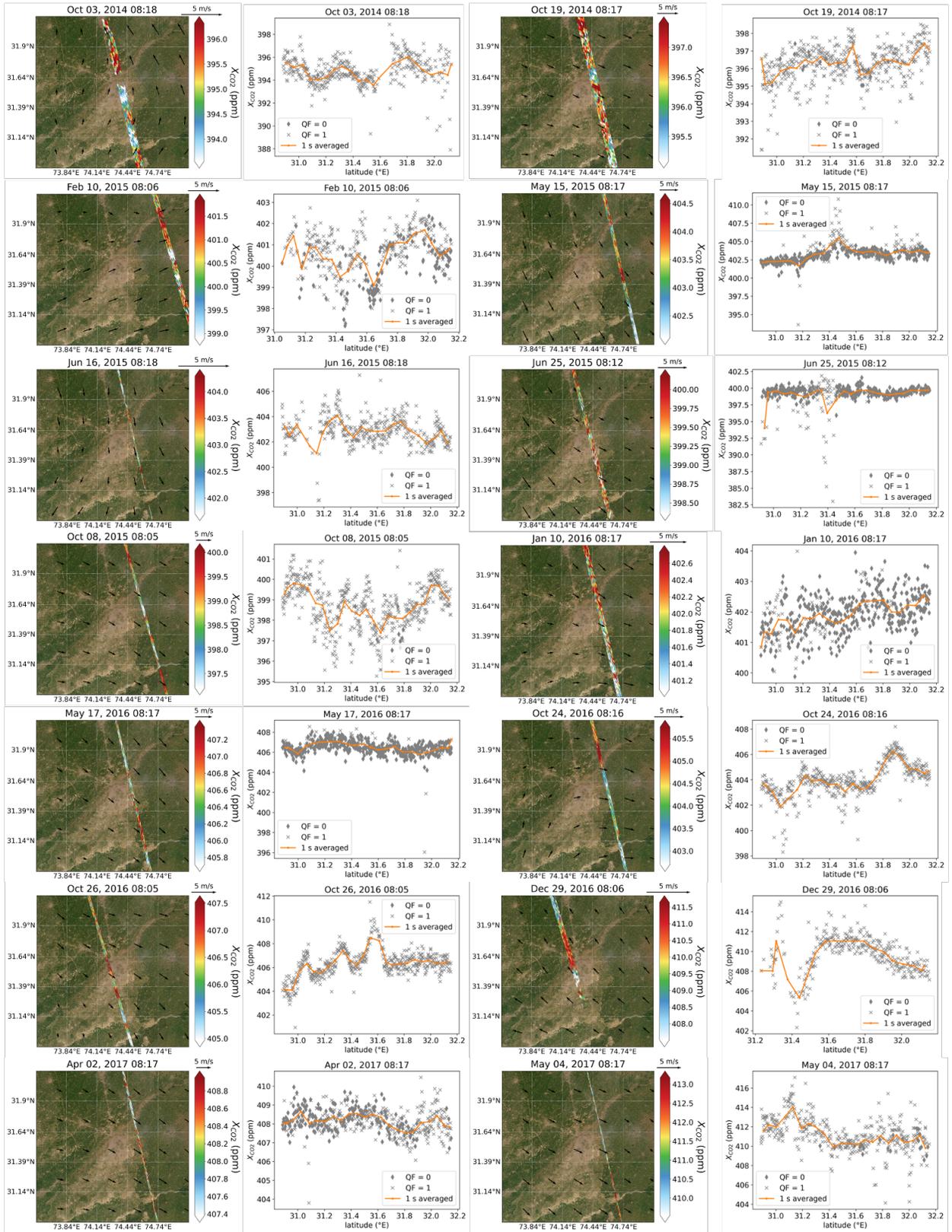
248 results with WRF studies over Los Angeles (LA). Angevine et al. (2012) evaluated WRF at 4 km  
249 for California Nexus (CalNex) field campaign over LA using a radar wind profiler operated by  
250 the South Coast Air Quality Management District near Los Angeles International Airport (LAX).  
251 They found showed  $1.1 \pm 2.7$  m/s bias in wind speed and  $-2.6 \pm 67^\circ$  in wind direction near the  
252 surface. Feng et al., 2016 evaluated a set of WRF planetary boundary layer (PBL) schemes  
253 performance over LA. They found the biases of the MYNN\_UCM scheme are  $1.4 \pm 2.0$  m/s in  
254 wind speed and  $-1.3 \pm 20.0^\circ$  in wind direction at the 4 km model resolution.

255

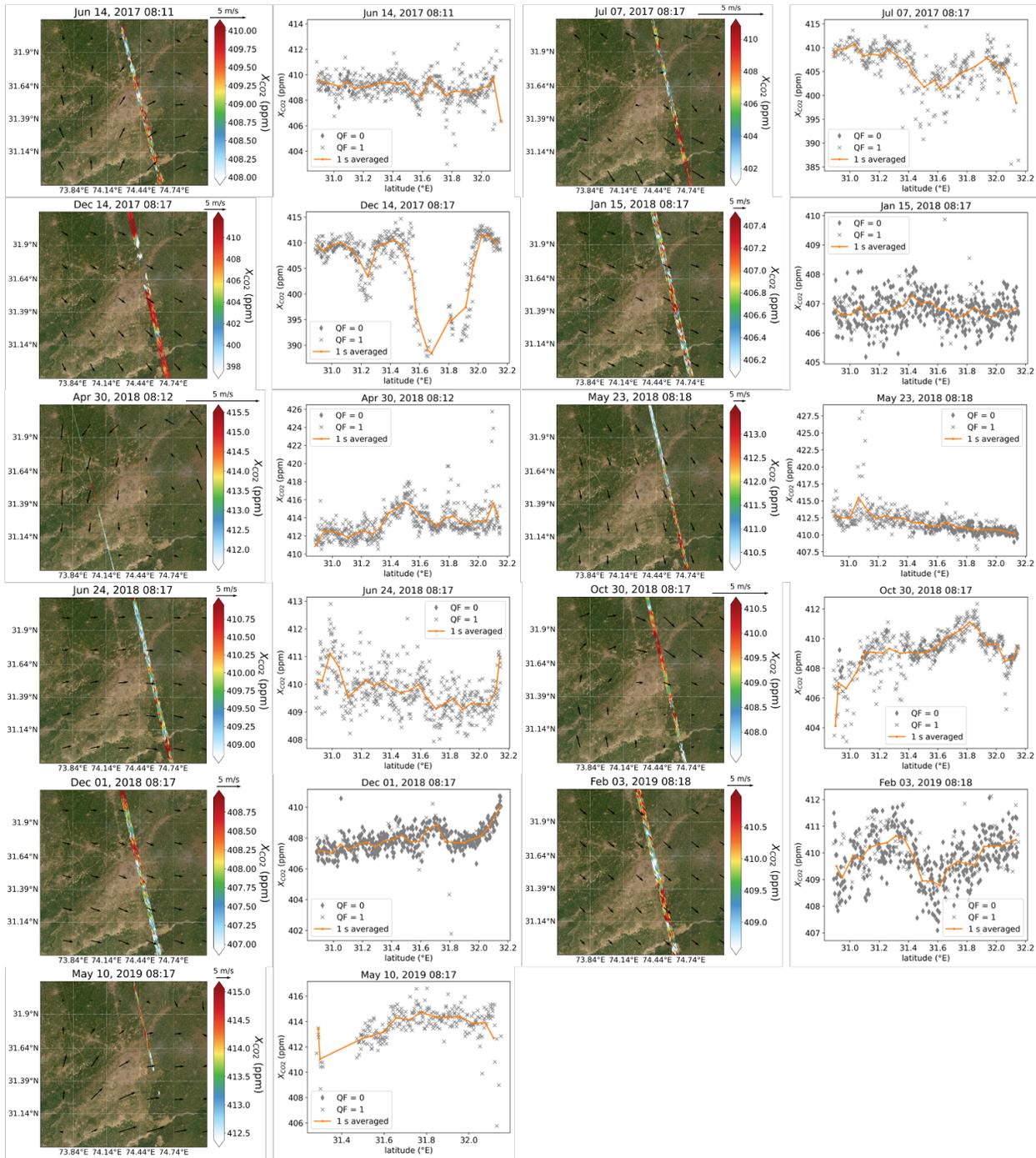
#### 256 **Text S5 Optimize the OCO-2 quality flag selection criteria**

257 The OCO-2 quality flags are originally designed for global-scale studies. Although we evaluated  
258 their effectiveness at the city level in Section 3.5, the valid tracks over Lahore are too few to  
259 capture the  $\text{CO}_{2\text{ff}}$  emission trend without relying on the trend of prior (Section 3.6). We tried to  
260 optimize the OCO-2 selection criteria based on  $X_{\text{CO}_2}$  residuals between OCO-2 and WRF-Chem.  
261 We found that the default thresholds triggering the selection criteria are slightly over-strict  
262 (Figure S12). By adequately enlarging the range of thresholds of 8 criteria, we obtained more  
263 high-quality soundings (Table S3) but failed to obtain more valid tracks. Further research on  
264 more tracks and over more cities is required to confirm if optimization of quality flag selection  
265 criteria over cities helps recover valid tracks.

266



269 Figure S1 OCO-2 tracks over Lahore. The first and the third columns: The OCO-2 all-data track  
270 locations. The colors represent the  $X_{CO_2}$  mixing ratios. The vectors represent 10-m wind, with the  
271 reference vector standing for the wind speed of 5 m/s. The background is the map of Lahore  
272 from the ArcGIS REST API service. The second and the fourth columns: The OCO-2  $X_{CO_2}$   
273 soundings with quality flags (QF = 0 and QF = 1) and 1-s averaged  $X_{CO_2}$  along the track  
274 latitudes. Note: Tracks filtered with quality flag (QF = 0) on May 15, 2015, June 25, 2015,  
275 January 10, 2016, May 17, 2016, April 04, 2017, January 15, 2018, December 01, 2018,  
276 February 03, 2019 were selected as high-quality tracks.  
277

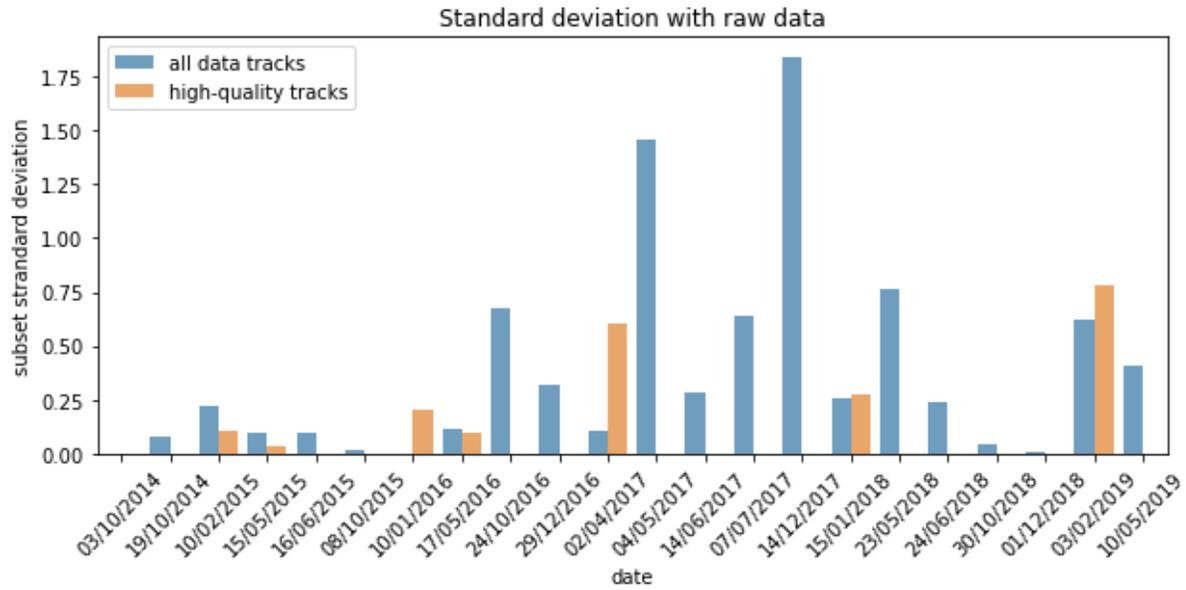


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279 Figure S1 (Cont.)

280

281

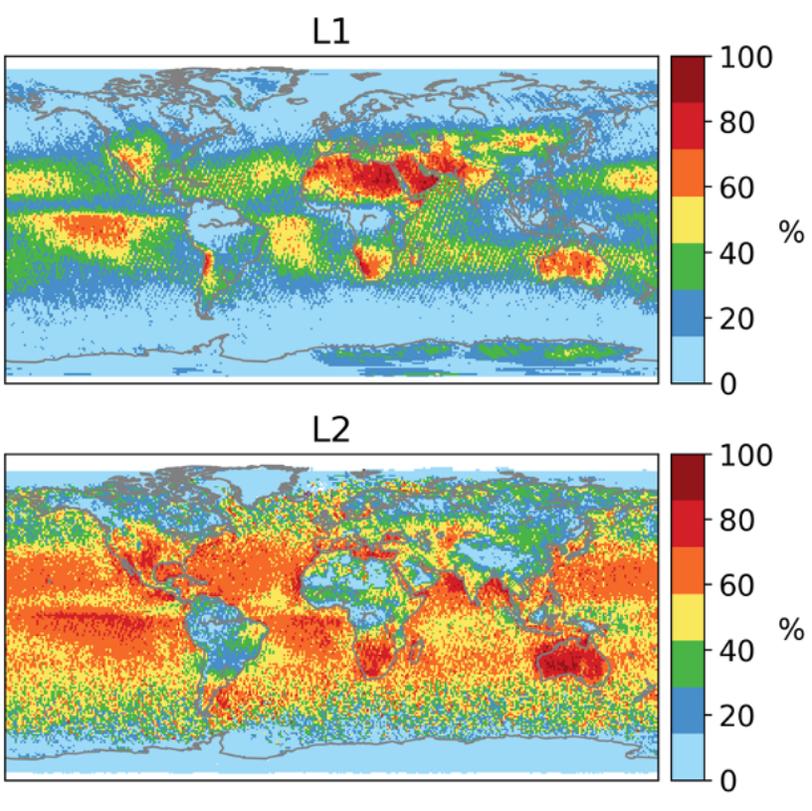


282

283 Figure S2 Standard deviation of the selected subset for each date, using all (blue) and only good  
 284 quality (orange) data from L2Lite\_v9r version of OCO2 files. This standard deviation gives an  
 285 estimation of only a fraction of the estimation uncertainty.

286

287



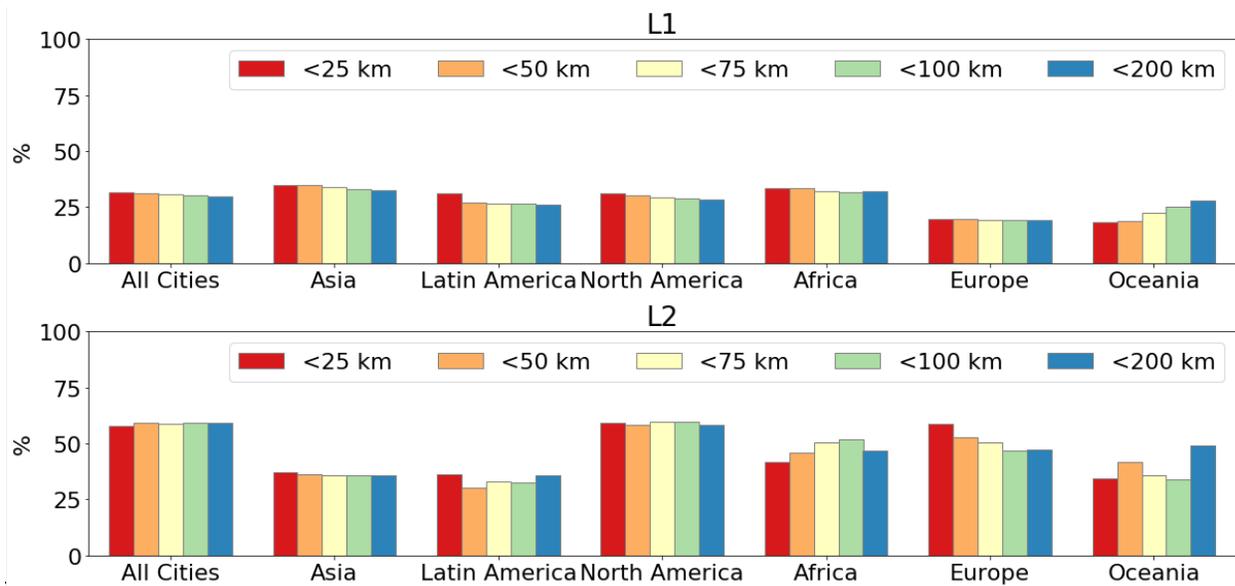
288

289 Figure S3 Global 1° x 1° OCO-2 high-quality soundings ratios in L1 and L2 products.



290

291 Figure S4 Names and locations of 70 cities.

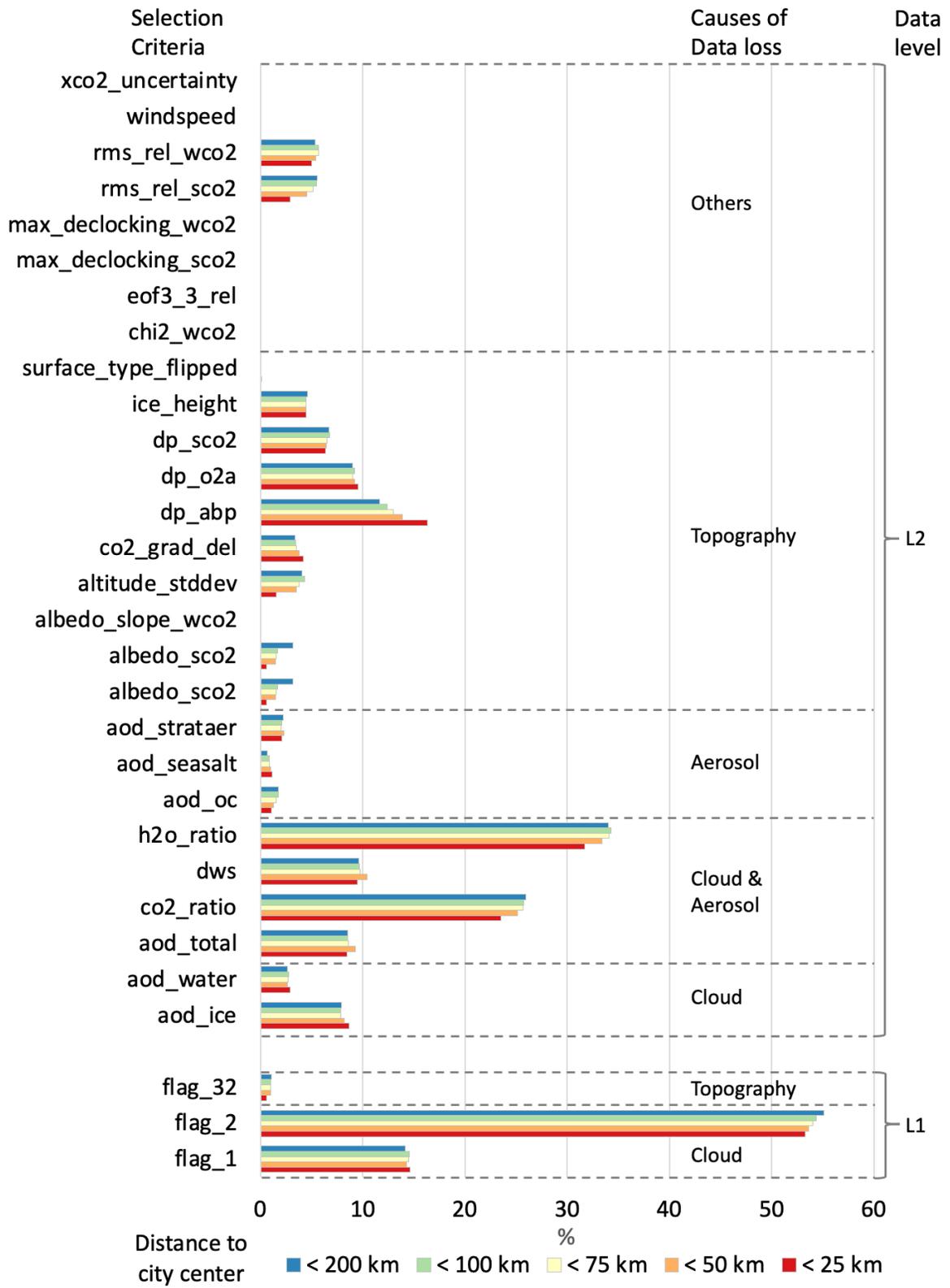


292

293 Figure S5 Regional ratios of OCO-2 high-quality soundings in L1 and L2 product over the most  
 294 populated 70 cities within boxes of 25-, 50-, 75-, 100-, and 200-km border-to-center distance.

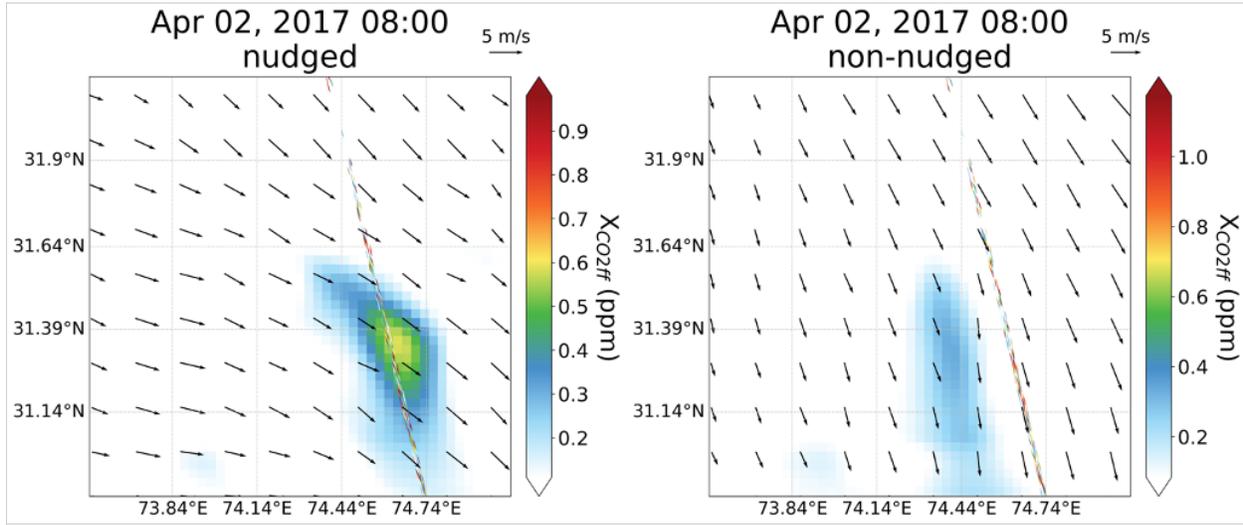
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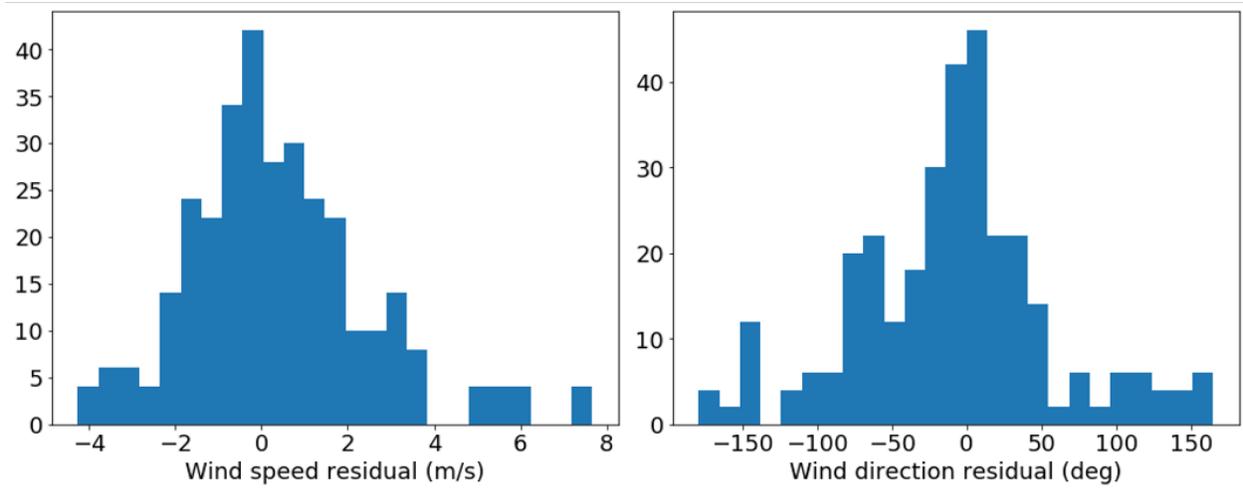
298 Figure S6 Percentages of data loss due to each sounding selection criterion over global 70 cities.



300

301 Figure S7 A sample comparison of modeled and observed  $X_{CO2f}$  enhancements between nudged  
302 (left) and non-nudged (right) simulations.

303

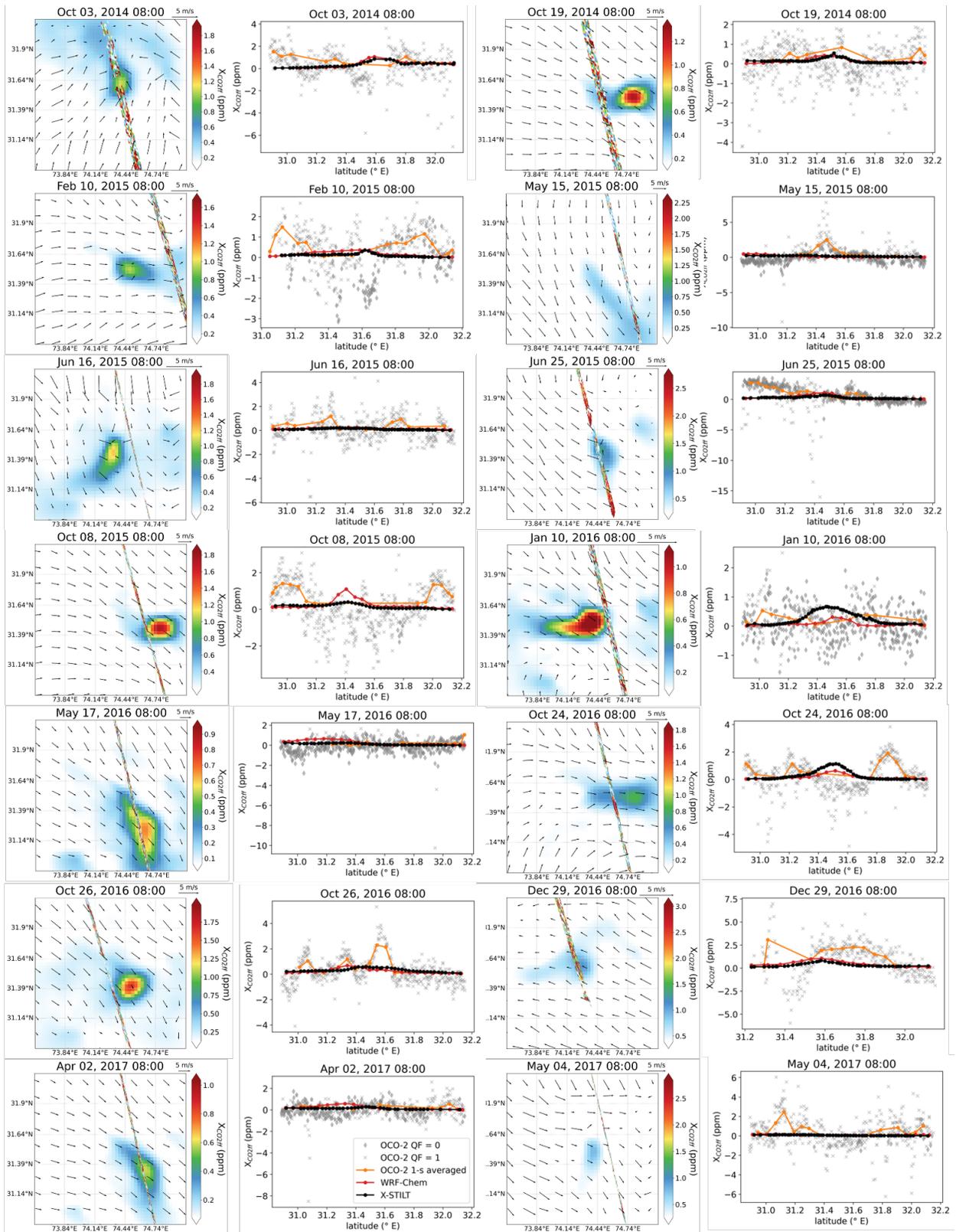


304

305 Figure S8 Histograms of wind speed (left) and direction (right) residuals of WRF-Chem outputs  
306 compared to monitor data.

307

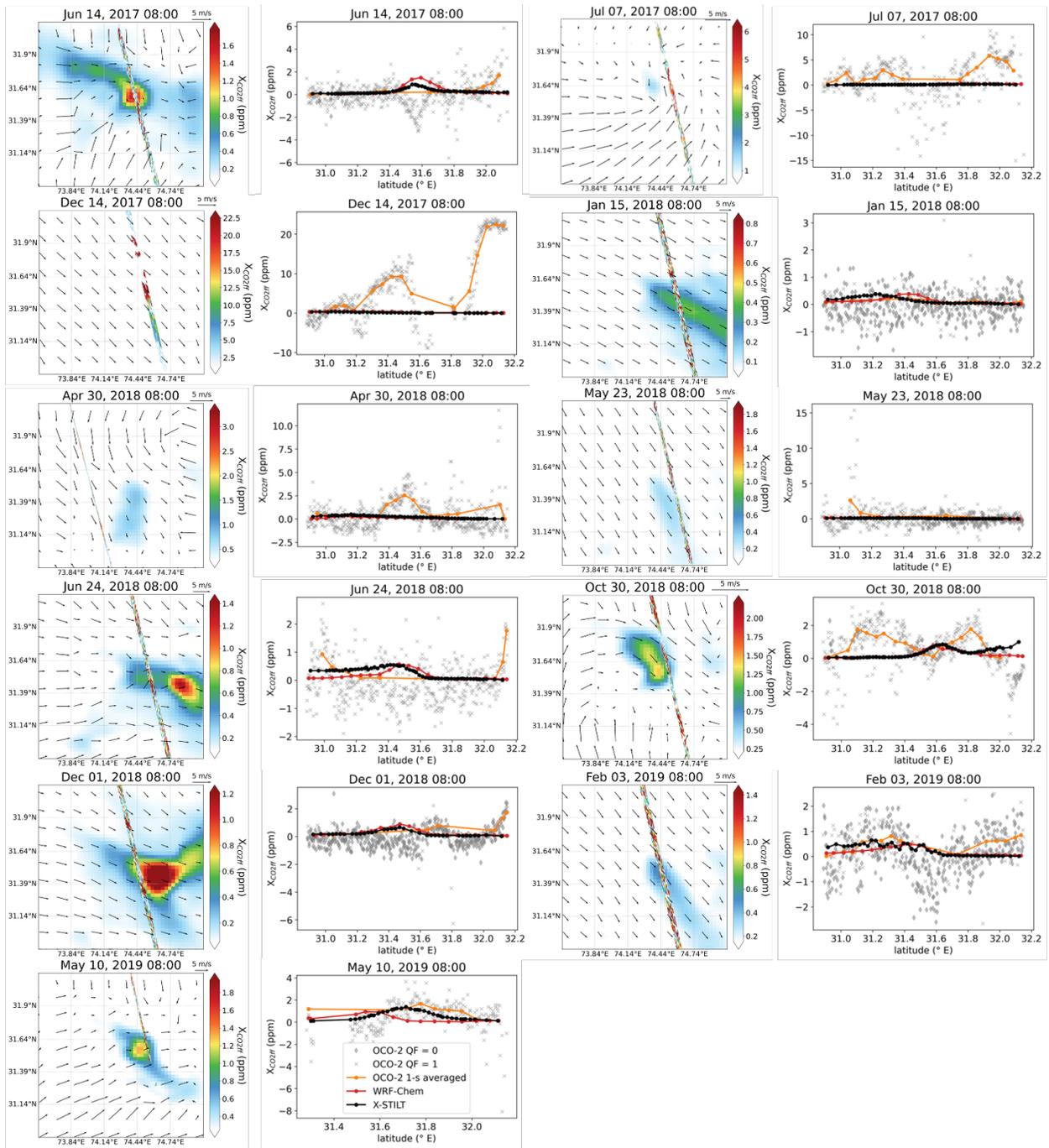
308



310 Figure S9 Comparison of modeled and observed  $X_{CO_2ff}$  enhancements by the OCO-2 data for 25  
311 all-data tracks (QF = 0&1) over Lahore. The first and third columns show the simulated  $X_{CO_2ff}$  by  
312 WRF-Chem and the observed  $X_{CO_2ff}$  obtained from the OCO-2 data (background and biosphere  
313  $X_{CO_2}$  have been subtracted). The vectors represent 10-m wind from WRF-Chem with the  
314 reference vector standing for the wind speed of 5 m/s. The second and fourth columns show the  
315 OCO-2  $X_{CO_2ff}$  (grey diamond marks represent high-quality (QF = 0) soundings, grey cross marks  
316 represent high-quality (QF = 1) soundings, background and biosphere  $X_{CO_2}$  have been  
317 subtracted), 1-s averaged OCO-2  $X_{CO_2ff}$  (Orange dotted line), simulated  $X_{CO_2ff}$  by WRF-Chem  
318 (red dotted line) and X-STILT (black dot line).

319

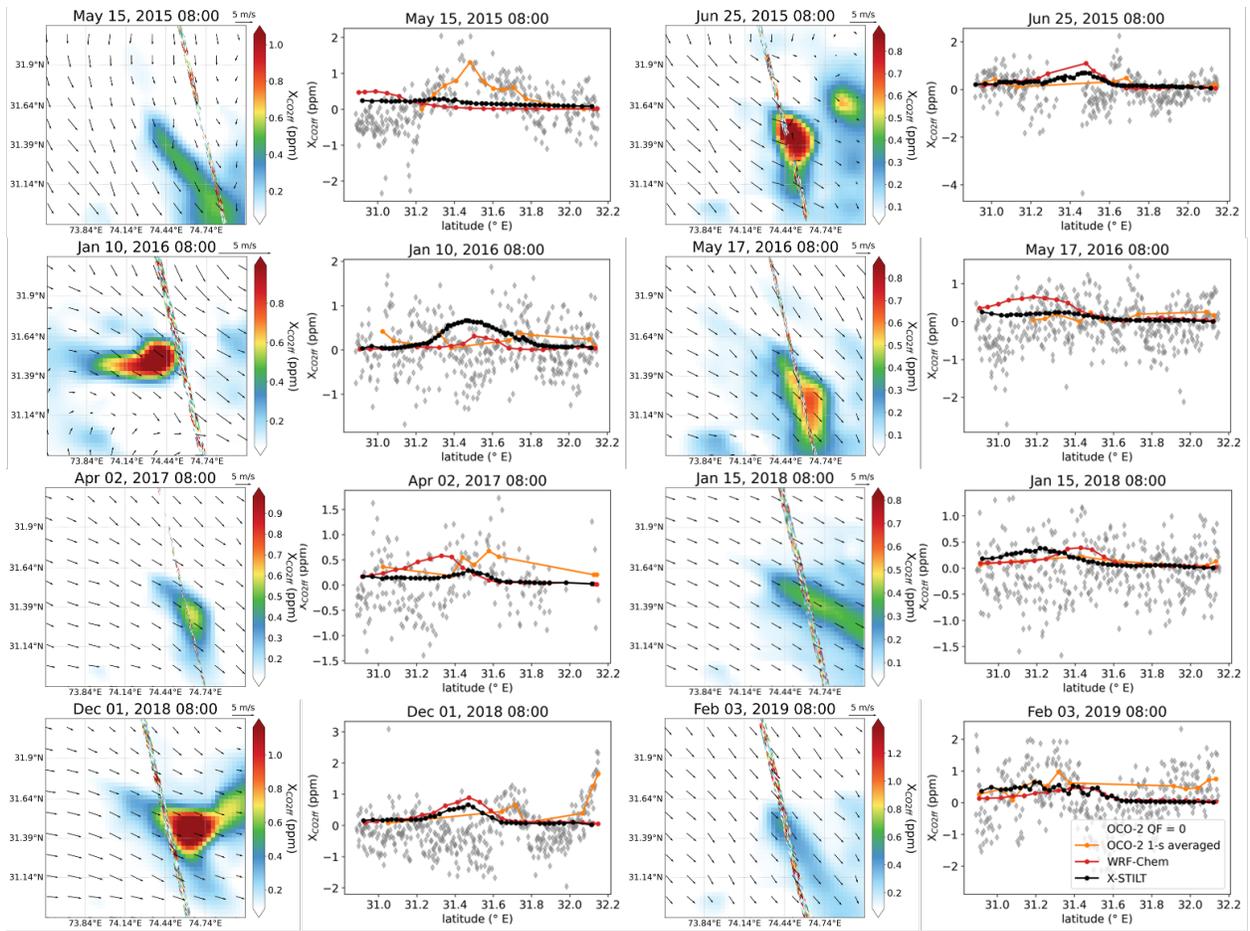
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322 Figure S9 (Cont.)

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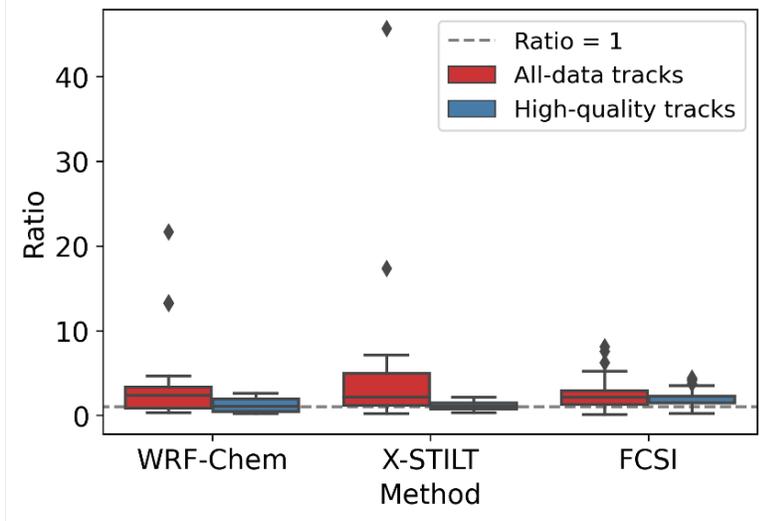


324

325 Figure S10 Same as Figure S9 but for 8 high-quality tracks. Low-quality (QF = 1) soundings are  
 326 filtered out and only keep high-quality (QF = 0) soundings.

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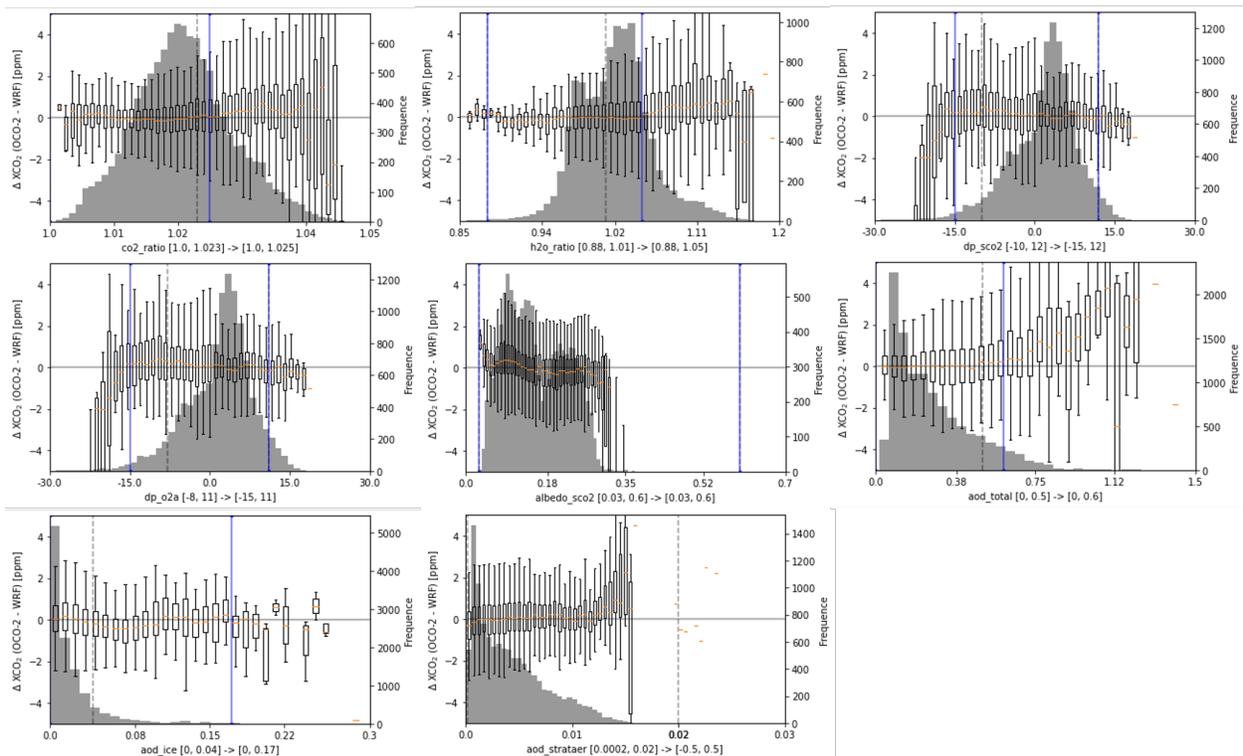


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330 Figure S11 Similar to Figure 5 but show outliers as black diamonds.

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334 Figure S12 Adjusting thresholds of OCO-2 quality flag selection criteria. The boxes represent the

335  $X_{CO_2}$  residuals between OCO-2 and WRF-Chem. For each box, the central line indicates the

336 median, and the bottom and top edges of the box indicate the 25th and 75th percentiles (q1 and  
337 q3), respectively. The outliers are omitted for simplicity. The whiskers extend to the most  
338 extreme value that is not an outlier. The grey dashed vertical lines represent the default  
339 thresholds of the OCO-2 selection criteria. The blue vertical lines represent the adjusted  
340 thresholds based on  $X_{CO_2}$  residuals. Labels on x-axes show the names of OCO-2 selection criteria  
341 and the default and adjusted thresholds triggering the quality flags in the square brackets.

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352 Table S1 Subset selection criteria for each date and data type.  $\theta_{\text{plume}}-\theta_{\text{wind}}$  is the angle between the  
 353 plume axis and the wind direction.

	All-data tracks		High-quality tracks	
	Criterion	Subset size after filtering	Criterion	Subset size after filtering
10/03/2014	$ \theta_{\text{plume}}-\theta_{\text{wind}} <15^\circ$	1	-	0
19/10/2014	$ \theta_{\text{plume}}-\theta_{\text{wind}} <10^\circ$	5	-	0
10/02/2015	$ \theta_{\text{plume}}-\theta_{\text{wind}} <7^\circ$	13	$ \theta_{\text{plume}}-\theta_{\text{wind}} <30^\circ$	5
15/05/2015	$ \theta_{\text{plume}}-\theta_{\text{wind}}+35^\circ <5^\circ$	25	$ \theta_{\text{plume}}-\theta_{\text{wind}}+35^\circ <5^\circ$	21
16/06/2015	$ \theta_{\text{plume}}-\theta_{\text{wind}} <15^\circ$	22	-	0
25/06/2015	-	0	-	0
08/10/2015	$ \theta_{\text{plume}}-\theta_{\text{wind}} <6.5^\circ$ & (plume size)/(track size) > 1/10	2	-	
10/01/2016	$ \theta_{\text{plume}}-\theta_{\text{wind}} <30^\circ$	1	$ \theta_{\text{plume}}-\theta_{\text{wind}} <26^\circ$	3
17/05/2016	$ \theta_{\text{plume}}-\theta_{\text{wind}} <7^\circ$	9	$ \theta_{\text{plume}}-\theta_{\text{wind}} <7^\circ$	5
24/10/2016	$(\theta_{\text{plume}}-\theta_{\text{wind}})<0^\circ$	38	-	
26/10/2016	-	0	-	0
29/12/2016	$ \theta_{\text{plume}}-\theta_{\text{wind}} <17^\circ$	10	-	0
02/04/2017	$ \theta_{\text{plume}}-\theta_{\text{wind}} <10^\circ$	3	$ \theta_{\text{plume}}-\theta_{\text{wind}} <5^\circ$	6
04/05/2017	$ \theta_{\text{plume}}-\theta_{\text{wind}} <25^\circ$	42	-	0
14/06/2017	$ \theta_{\text{plume}}-\theta_{\text{wind}} <36^\circ$	2	-	0
07/07/2017	$ \theta_{\text{plume}}-\theta_{\text{wind}} <15^\circ$	22	-	0
14/12/2017	$ \theta_{\text{plume}}-\theta_{\text{wind}} <5^\circ$	37	-	0
15/01/2018	$ \theta_{\text{plume}}-\theta_{\text{wind}} <2^\circ$	8	$ \theta_{\text{plume}}-\theta_{\text{wind}} <4^\circ$	14
30/04/2018	-	0	-	0
23/05/2018	$ \theta_{\text{plume}}-\theta_{\text{wind}} <15^\circ$	16	No soundings downwind	0
24/06/2018	$ \theta_{\text{plume}}-\theta_{\text{wind}} <15^\circ$	8	-	0
30/10/2018	$ \theta_{\text{plume}}-\theta_{\text{wind}} <5^\circ$	7	-	0
01/12/2018	$ \theta_{\text{plume}}-\theta_{\text{wind}} <10^\circ$	3	$ \theta_{\text{plume}}-\theta_{\text{wind}} <7^\circ$	1
03/02/2019	$ \theta_{\text{plume}}-\theta_{\text{wind}} <15^\circ$	10	$ \theta_{\text{plume}}-\theta_{\text{wind}} <15^\circ$	14
10/05/2019	all	50	-	0

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358 Table S2 Inversion inputs of 8 high-quality tracks over Lahore

Date	Prior total emission (Mt C/year)	Satellite integral $X_{CO_2ff} (y_o) \pm$ Measurement uncertainty ( $\sigma_{\text{measurement}}$ ) (unit: ppm)	Model integral $X_{CO_2ff} (y_m) \pm$ Transport model uncertainty ( $\sigma_{\text{model}}$ ) (unit: ppm)
May 15, 2015	8.52E+06	$0.37 \pm 0.26$	$0.15 \pm 0.23$
June 25, 2015	8.55E+06	$0.16 \pm 0.26$	$0.44 \pm 0.50$
January 10, 2016	8.90E+06	$0.25 \pm 0.22$	$0.10 \pm 0.22$
May 17, 2016	9.04E+06	$0.17 \pm 0.20$	$0.33 \pm 0.21$
April 2, 2017	9.57E+06	$0.31 \pm 0.43$	$0.26 \pm 0.16$
January 15, 2018	1.03E+07	$0.15 \pm 0.28$	$0.17 \pm 0.16$
December 1, 2018	1.10E+07	$0.07 \pm 0.26$	$0.36 \pm 0.66$
February 3, 2019	1.06E+07	$0.42 \pm 0.28$	$0.24 \pm 0.31$

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363 Table S3 Comparison of high-quality soundings (QF = 0) before and after adjusting the  
 364 thresholds OCO-2 quality flag selection criteria.

Date	Good soundings (QF = 0)		All soundings (QF = 0&1)
	Before adjusting	After adjusting	
Oct 03, 2014	1	76	411
Oct 19, 2014	19	95	481
Feb 10, 2015	133	284	418
May 15, 2015	338	373	461
Jun 16, 2015	0	243	408
Jun 25, 2015	346	357	449
Oct 08, 2015	8	235	533
Jan 10, 2016	410	456	494
May 17, 2016	434	456	491
Oct 24, 2016	0	79	463
Oct 26, 2016	0	33	498
Dec 29, 2016	0	3	320
Apr 02, 2017	214	385	498
May 04, 2017	24	162	423
Jun 14, 2017	13	166	479
Jul 07, 2017	2	35	347
Dec 14, 2017	18	61	380
Jan 15, 2018	395	459	489
Apr 30, 2018	1	80	535
May 23, 2018	136	209	488
Jun 24, 2018	1	242	487
Oct 30, 2018	45	297	448
Dec 01, 2018	443	478	486
Feb 03, 2019	386	438	487
May 10, 2019	0	16	264

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