Supplementary information

1 Upwind background selection

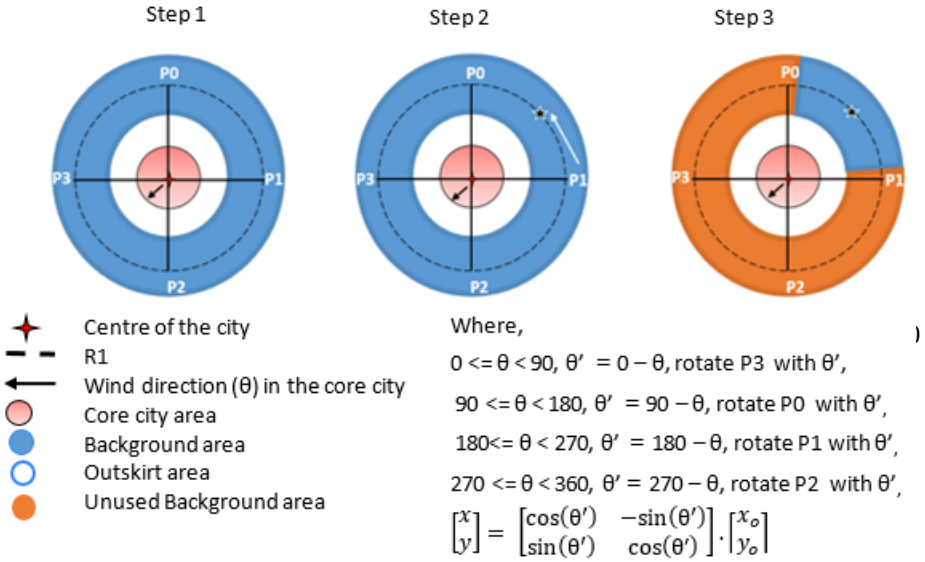

Figure S1. Schematic representation of the procedure used for selecting the upwind background. The centre of the city is represented by red star. The city, outskirt and background radii (see Table 1) are used to divide the city into three parts i.e. the core city (red circle), outskirt (white circle) and background region (blue circle), respectively. Step 1. Selection of radius R1, the mean of outskirt and background radii. Select the points P0, P1, P2, P3 where the north, east south and west wind directions ($\boldsymbol{\theta}$) intersect at the outer rim of the dashed circle with radius R1. The black arrow symbolises an average wind direction over the core city region, which is used to derive θ'. Step 2. The rotation of the corresponding point (P1) with θ' in reference to the city centre and generate the new point. Step 3. Select the fraction of the upwind region. The Δlat and Δlon from the new point vary with the size of the city and are provided in Table1.

15 **2 CO and NO2 enhancements over megacities from a single TROPOMI overpass**

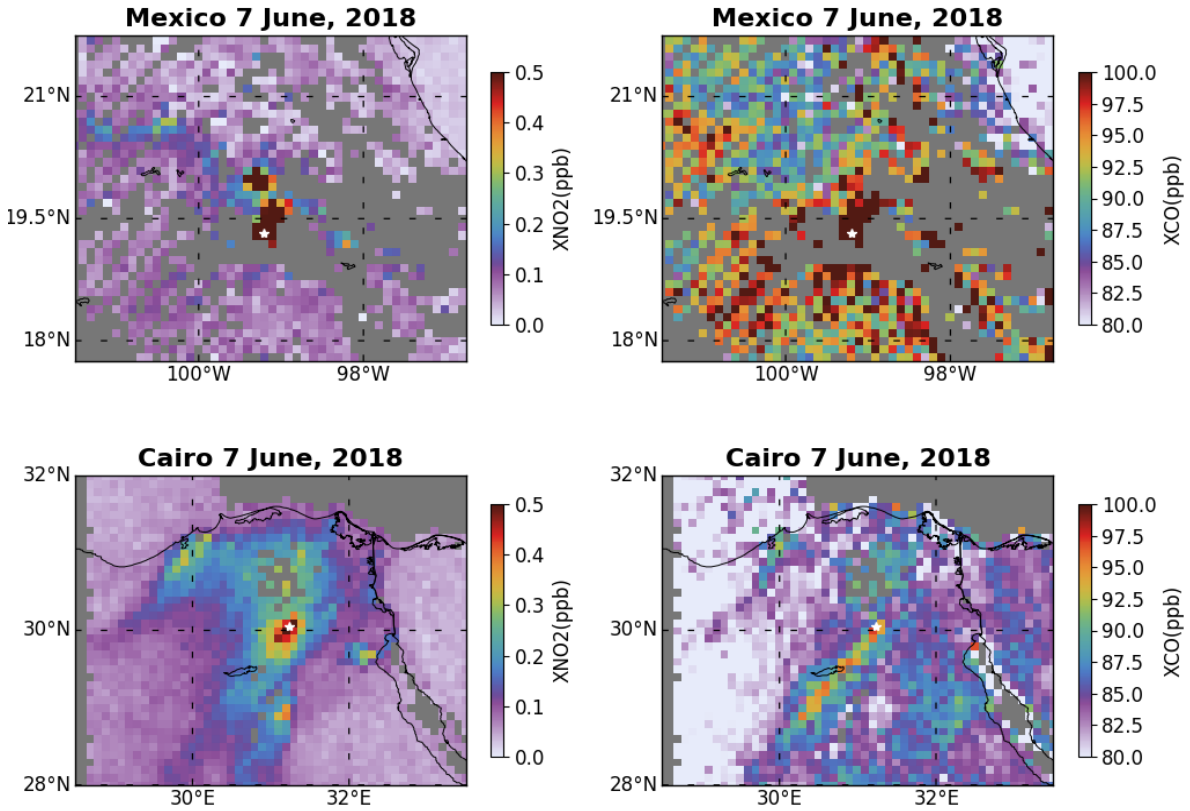

Figure S2. Collocated XCO and XNO2 for 7 June, 2018 over Mexico (top) and Cairo (bottom). The white star represents the city centre. CO and NO2 retrievals are gridded at 0.1˚x0.1˚ resolution.

3 Correlation between TROPOMI, EDGAR and MACCity

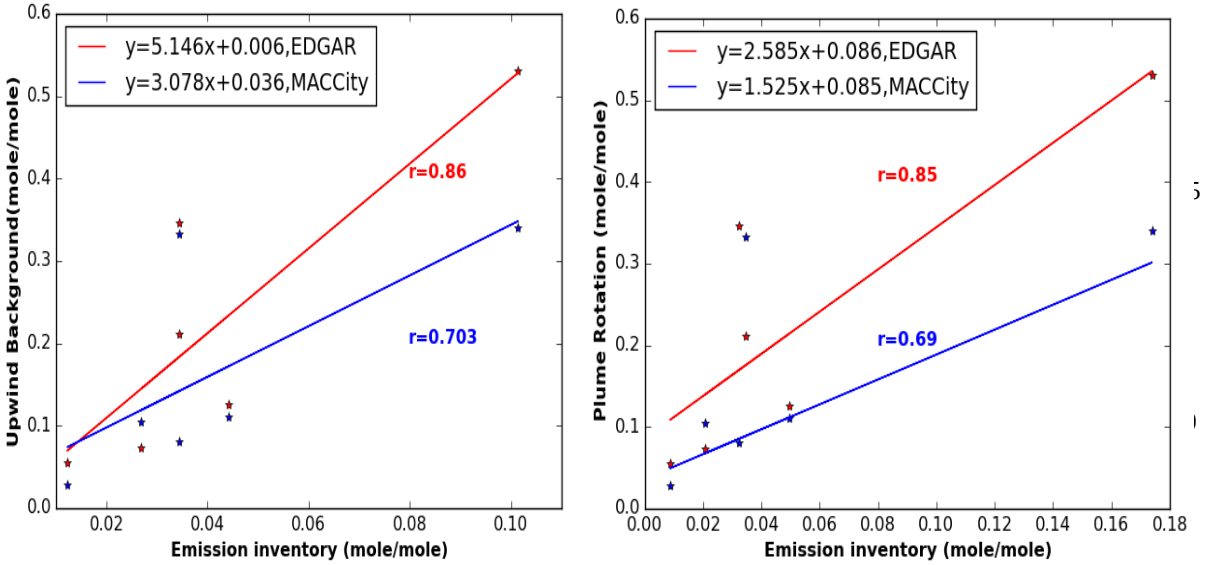

Figure S3. TROPOMI derived ($\Delta X_{NO2}/\Delta X_{CO}$) column enhancement ratio using the Upwind Background (left) and Plume rotation method (right) versus inventory NO2/CO emission ratio and corresponding regression lines for MACCity (blue) and TROPOMI (red).

Table S1. Summary of TROPOMI and inventory derived NO2/CO ratios. Estimates for TROPOMI are given using the upwind background and plume rotation methods, with and without corrections.

Cities	Upwind ratio ± error	Upwind ratio (OH corrected ratio)	Upwind ratio (OH +Ak)	Plume rotation ratio ± error	Plume rotation ratio (OH corrected ratio)	Plume rotation ratio (OH +Ak)	MACCity ratio	EDGAR ratio
Tehran	0.044± 0.0031	0.09	0.1	0.049± 0.0074	0.10	0.11	0.11	0.126
Mexico city	0.027± 0.0019	0.076	0.084	0.021± 0.0040	0.062	0.068	0.10	0.073
Cairo	0.034± 0.005	0.058	0.063	0.032 ± 0.0046	0.056	0.060	0.08	0.345
Riyadh	0.10± 0.03	0.25	0.29	0.15± 0.034	0.35	0.39	0.34	0.53
Lahore	0.012± 0.003	0.054	0.064	0.009± 0.0007	0.037	0.045	0.028	0.055
Los Angeles	0.034± 0.008	0.074	0.087	0.034± 0.0032	0.073	0.086	0.33	0.211

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Table S2. The CAMS derived influence of the averaging kernel on the total column NO2/CO ratio

City	Days	NO2/CO Ak =1 (A)	NO2/CO TROPOMI Ak (B)	Relative difference ((A-B)/A)*100 (%)
Tehran	July 03, 04 August 20	0.0158 ±0.002	0.0143 ±0.0019	9.5
Mexico City	June-August	0.024361 ±0.0018	0.022251± 0.00173	9.0
Cairo	August,09, 15,16,21,22,31	0.053813 ± 0.0062	0.050061 ±0.00861	7.1
Riyadh	July 06, 12 August 18,22,27,28	0.147859 ±0.08	0.133242 ±0.07	11.0
Lahore	June 06 July 06 August 19, 29, 30	0.004024±0.000985	0.003352 ±0.00080	16.6
Los Angeles	June-August	0.065387 ±0.0090	0.055326 ±0.00752	15.4

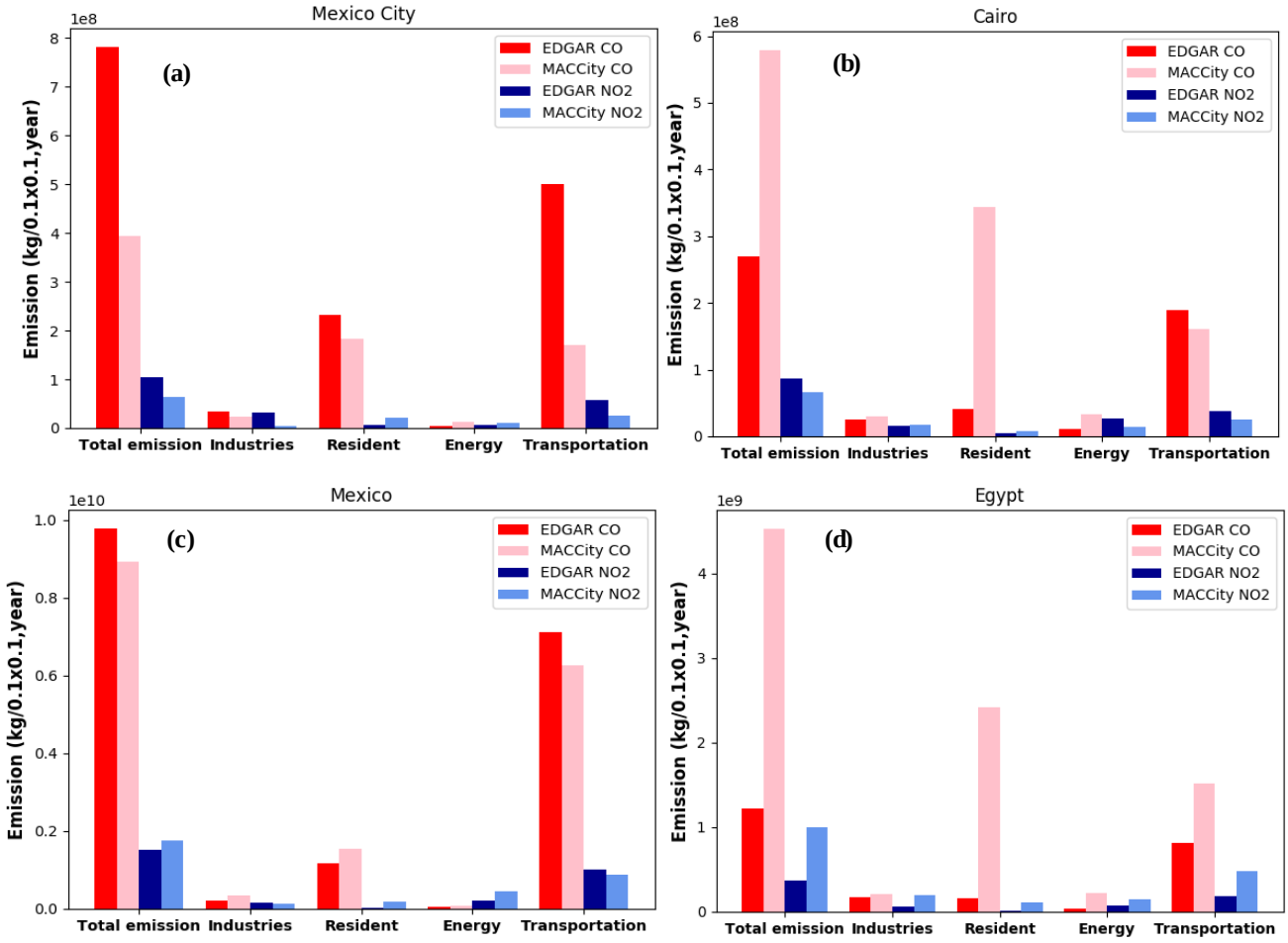

Figure S4. Comparison of EDGAR and MACCity CO and NO2 emissions by sector for a. Mexico City b. Cairo c. Mexico and d. Egypt

Table S3. Selected ground based measurement sites in Mexico City and Los Angeles

Mexico	
Station	Location (Latitude, longitude)
Alcoman (ACO)	19.635501, -98.912003
Atizapan(ATI)	19.576963, -99.254133
Camarones (CAM)	19.4684, -99.1698
Centro de Ciencias de la Atmósfera (CCA)	19.468404, -99.176111
Cuajimalpa (CUA)	19.365313, -99.291705
FES Acatlán (FAC)	19.482473, -99.243524
Iztacalco (IZT)	19.384413, -99.117641
La Presa (LPR)	19.53473, -99.11772
Montecillo (MON)	19.460415, -98.902853
Merced (MER)	19.42461, -99.119594
Mguel Hidalgo (MGH)	19.41162, -99.20266
Pedregal (PED)	19.32515, -99.2041
San Agustín (SAG)	19.532968, -99.030324
Tlalnepantla (TLA)	19.529077, -99.204597
Tlahuac (TAH)	19.246459, -99.010564
Tultitlán (TLI)	19.602542, -99.177173
UAM Xochimilco (UAX)	19.304441, -99.103629
UAM Iztapalapa (UIZ)	19.360794, -99.07388
Villa de las Flores (VIF)	19.658223, -99.09659
Xalostoc (XAL)	19.525995, -99.0824
Los Angeles	
Azusa	34.13648 -117.92392
Lancaster-43301 Division Street	34.66959 -118.13068
Compton-700 North Bullis Road	33.901445 -118.204989
Glendora-Laurel	34.14437 -117.85038
Los Angeles-North Main Street	34.06653 -118.22676
Los Angeles-Westchester Parkway	33.95507 -118.43049

Pasadena-S Wilson Avenue	34.13265	-118.12714
Pico Rivera-4144 San Gabriel	34.01029	-118.0685
Pomona	34.06698	-117.75138
Reseda	34.1992	-118.53275
Santa Clarita	34.38337	-118.52839
West Los Angeles-VA Hospital	34.05109	-118.4564

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