## Supplementary Information

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Table S1. Per capita $\mathrm{CO}_{2}$ emissions by sector for cities and metropolitan regions

| City | Electricit <br> y <br> ( $\mathrm{tCO} \mathrm{Cl}_{2}$ /c <br> ap) | Heating \& Industrial Fuels ( $\mathrm{tCO}_{2} \mathrm{e} / \mathrm{cap}$ ) | Industrial Processes ( $\mathrm{tCO} \mathrm{CO}_{2} \mathrm{e}$ cap ) | Ground <br> Transpor tation $\left(\mathrm{tCO}_{2} \mathrm{e} / \mathrm{c}\right.$ ap) | Energy $\begin{gathered} \left(\mathrm{tCO}_{2} \mathrm{e} /\right. \\ \text { cap }) \end{gathered}$ | Electricity/ <br> Energy <br> (\%) | Heating <br>  <br> Industrial <br> Fuels/ <br> Energy <br> (\%) | Ground Transporta tion/Energ y <br> (\%) | Large <br> Point <br> Source $\left(\mathrm{tCO}_{2} / \mathrm{ca}\right.$ <br> p) |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Bangkok | 2.77 | 2.49 | 0 | 2.27 | 7.53 | 36.8 | 33.1 | 30.1 | 0.10 |
| Beijing | 3.72 | 4.50 | 0.79 | 0.91 | 9.13 | 40.7 | 49.3 | 10.0 | 1.09 |
| Shanghai | 4.71 | 4.82 | 1.25 | 1.11 | 10.64 | 44.3 | 45.3 | 10.4 | 2.73 |
| Delhi | 0.59 | 0.30 | 0 | 0.42 | 1.3 | 45.0 | 23.0 | 32.0 | 0.25 |
| Cape <br> Town | 3.74 | 0.29 | 0 | 1.41 | 5.44 | 68.8 | 5.3 | 25.9 | 0.05 |
| Sao Paulo | 0.12 | 0.11 | 0 | 0.74 | 0.97 | 12.4 | 11.3 | 76.3 | 0.09 |
| Tokyo | 1.58 | 1.65 | 0.1 | 1.17 | 4.4 | 36.0 | 37.4 | 26.6 | 0.23 |
| Greater <br> Paris | 0.44 | 2.31 | 0.32 | 1.41 | 4.16 | 10.6 | 55.5 | 33.9 | 0.32 |
| Greater <br> London | 2.5 | 2.58 | 0 | 1.22 | 6.3 | 39.7 | 41.0 | 19.4 | 0.59 |
| Los <br> Angeles | 2.46 | 1.37 | 0.22 | 4.92 | 8.75 | 28.1 | 15.7 | 56.2 | 1.65 |
| Manhattan | 3.01 | 3.13 | 0 | 1.53 | 7.67 | 39.2 | 40.8 | 19.9 | 0.08 |
| New York City | 3.01 | 3.13 | 0 | 1.53 | 7.67 | 39.2 | 40.8 | 19.9 | 0.62 |
| Washingto <br> n D.C. | 6.3 | 4.84 | 0 | 6.86 | 18 | 35.0 | 26.9 | 38.1 | 0.21 |

Greater

| 2.47 | 3.30 | 0.57 | 4.0 |
| :--- | :--- | :--- | :--- |

9.82
25.2
33.6
41.2
0.06

Toronto

For Washington D.C., Manhattan, Delhi and Tokyo, the $\mathrm{CO}_{2}$ emissions by sector can be used from this table. Due to data limitation, we needed to divide the heating $\&$ industrial fuels and ground transportation from energy after the allocation of electricity proportion for these four regions. Here emissions from energy in Table S3 include emissions from stationary, mobile combustion and fugitive sources (specifically from industrial fuels, electricity \& heat production, and ground transportation excluding aviation and marine). Referring to the literature [1], we found that more than $30 \%$ emissions can be attributed to ground transportation in some North American cities. And based on the similar fuel economy and habits of vehicle usage in America, we assumed that the average ground transportation proportion of New York City and Los Angeles can be used as the proportion of ground transportation for Washington D.C.. For Manhattan we also assumed the same ground transportation proportion as New York City. For Delhi we adopted the proportion of ground transportation from the literature [2] directly. According to social economic development, population density and quality of public transit, we assumed the average ground transportation proportion for Tokyo can be empirically established on European cities. So the average ground transportation proportion of Greater London and Greater Paris was employed for Tokyo. After the proportion division of ground transportation and electricity from energy, the remainder of energy would be the proportion of heating \& industrial fuels.

Table S2. Detailed calculations for the inventory-based urban emissions and data sources by
sectors

| City | Source | Energy | Electric ity |  <br> Industrial <br> Fuels | Ground Transportatio n (Local Fuel Sales) | Ground Transportat ion (VKT) | Ground Transportati on (Scaling) | Industrial Processes |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Bangkok | Kennedy et <br> al.(2010),Ph <br> dungsilp <br> (2006) |  | $\checkmark$ | $\checkmark$ | $\sqrt{*}$ | $\checkmark$ |  | Unknown |
| Beijing | Sugar et <br> al.(2012) |  | $\checkmark$ | $\checkmark$ | $\sqrt{*}$ |  |  | $\checkmark$ |
| Shanghai | Sugar et <br> al.(2012) |  | $\checkmark$ | $\checkmark$ | $\sqrt{*}$ |  |  | $\checkmark$ |
| Delhi | Mitra et <br> al.(2003) | $\checkmark$ | ? | ? | ? | ? | ? | ? |
| Cape Town | Kennedy et al.(2011) |  | $\checkmark$ | $\checkmark$ | $\sqrt{*}$ |  |  | Unknown |
| Sao Paulo | Hoornweg et al.(2012) |  | $\checkmark$ | $\checkmark$ | $\sqrt{*}$ |  |  | $\checkmark$ |
| Tokyo | Tokyo Metropolitan Government, 2009 | $\checkmark$ | ? | ? | ? | ? | ? | $\checkmark$ |
| Greater Paris | ARENE/AD <br> EME (2010) |  | $\checkmark$ | $\checkmark$ | ? | ? | ? | $\checkmark$ |
| Greater <br> London | Kennedy et al.(2010) |  | $\checkmark$ | $\checkmark$ |  | $\sqrt{*}$ |  | Unknown |
| Los Angeles | Kennedy et al.(2010) |  | $\checkmark$ | $\checkmark$ |  |  | $\sqrt{*}$ | $\checkmark$ |
| New York City | Kennedy et al.(2010) |  | $\checkmark$ | $\checkmark$ |  | $\sqrt{*}$ | $\checkmark$ | Unknown |
| Washington D.C. | DC Dept. <br> Health <br> (2005) | $\checkmark$ | ? | ? | ? | ? | ? | ? |
| Greater <br> Toronto | Kennedy et <br> al.(2010) |  | $\checkmark$ | $\sqrt{ }$ | $\sqrt{*}$ |  | $\checkmark$ | $\checkmark$ |
| $\checkmark$ data is available $*$ final method choice of emission from ground transportation ?data and method are unsure or unknown (to the authors) |  |  |  |  |  |  |  |  |

Table S3. Comparison of area for cities and metropolitan regions

| City | total land Area <br> (km²_Kennedy_2009) | total land Area <br> (km²_Kennedy_2016) | Area $\begin{gathered} \text { (GADM) } \\ \mathrm{km}^{2} \end{gathered}$ | Area differences (\%) |
| :---: | :---: | :---: | :---: | :---: |
| Bangkok | 1,569 | N/A | 1,574 | 0.31 |
| Beijing | N/A | 16,411 | 16,424 | 0.08 |
| Delhi | N/A | 1,483 | 1,508 | 1.64 |
| Greater London | 1,579 | N/A | 1,604 | 1.58 |
| Los Angeles | 10,518 | N/A | 10,612 | 0.89 |
| Manhattan | N/A | N/A | 69 | N/A |
| New York City | 789 | N/A | 807 | 2.22 |
| Greater Paris | N/A | 12,011 | 12,058 | 0.39 |
| Shanghai | N/A | 6,341 | 6,905 | 8.17 |
| Tokyo | N/A | N/A | 1,805 | N/A |
| Greater Toronto | 7,195 | N/A | 7,636 | 5.77 |
| Washington D.C. | 177* | N/A | 166 | 6.04 |
| Sao Paulo City | N/A | 1,532* | 1,531 | 0.05 |
| Cape Town | 2454 | N/A | 2,451 | 0.12 |

*The area of Washington D.C. and Sao Paulo city were from the local governmental data online separately not from Kennedy’s research data due to the different definition of the regions.

Firstly, we needed to make sure the city definition were the same with Kennedy's. As assumed that the same definition of one region can reflect the same area information to a certain extent. So we compared GADM area information with Kennedy's data in the same region to reconfirm we had the identical boundaries.

Table S4. Fractional estimation of emission from electricity sector

| City | Industrial <br> Processes $\left(\mathrm{tCO}_{2} / \mathrm{cap}\right)$ | Energy <br> ( $\mathrm{tCO}_{2} / \mathrm{c}$ <br> ap) | Electricity/Ener gy (\%) | Large <br> Point <br> Source $(\mathrm{tCO} 2 / \mathrm{ca}$ <br> p) | In-bounda <br> ry FFE $\left(\mathrm{tCO}_{2} / \mathrm{cap}\right)$ | $\begin{gathered} \text { ODIAC } \\ \text { FFE }\left(\mathrm{tCO}_{2} / \mathrm{ca}\right. \\ \text { p) } \end{gathered}$ | Differenc <br> es (\%) |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Delhi | 0 | 1.3 | 45.0 | 0.25 | 0.97 | 0.90 | -7 |
| Delhi(Max) | 0 | 1.3 | 44.3 | 0.25 | 0.97 | 0.90 | -7 |
| Delhi(Min) | 0 | 1.3 | 36.8 | 0.25 | 1.07 | 0.90 | -16 |
| Tokyo | 0.1 | 4.4 | 36.0 | 0.23 | 3.15 | 3.82 | 21 |
| Tokyo(Max) | 0.1 | 4.4 | 39.7 | 0.23 | 2.98 | 3.82 | 28 |
| Tokyo(Min) | 0.1 | 4.4 | 25.2 | 0.23 | 3.62 | 3.82 | 5 |
| Washington |  |  |  |  |  |  |  |
| D.C. |  |  |  |  |  |  |  |
| Washington |  |  |  |  |  |  |  |
| D.C. (Max) |  |  |  |  |  |  |  |
| Washington |  |  |  |  |  |  |  |
| D.C. (Min) | 0 | 18 | 25.2 | 0.21 | 13.67 | 6.91 | -49 |

In absence of detailed data, Delhi, Tokyo and Washington D.C. only have emissions from the total energy sector (including electricity, heating \& industrial fuels and ground transportation) with the exception of aviation and marine available: for these three cities assumptions were necessary to estimate the fraction of emission from electricity sector. We empirically estimated the fractions by referring to cities with similar geographical location and social economic development level. These fractional estimations contribute to sources of uncertainties in inventory-based in-boundary emissions for these three cities.

We chose the value of $45 \%$ for proportion of electricity in energy for Delhi based on the main coal supply for electricity generation [3] and similar densely populated megacity plagued by poor air quality in the Asian region, such as Beijing (40.7\%) and Shanghai (44.3\%). For Washington D.C., we assumed $35 \%$ as the proportion, a value in between emission percentages for the other two U.S. cities studied, New York City (39.2\%) and Los Angeles (28.1\%). Similarly for Tokyo, 36\% of $\mathrm{CO}_{2}$ emissions from energy contributed to electricity sector emission was assumed based on the average of some megacities in developed countries such as New York City (39.2\%), Los Angeles (28.1\%) and Greater London (39.7\%), (Supplementary Table S1). These values should be treated with caution as actual proportion values may easily differ from our simple assumptions by $10 \%$ or more.

To evaluate the uncertainties caused by fraction estimation of emission from electricity sector, we calculated the in-boundary $\mathrm{CO}_{2}$ emissions based on the maximum and minimum proportion of electricity in energy for Delhi, Tokyo and Washington D.C. separately. Note this uncertainty range is only meant to assess the extent to which our assumption on fractional emission from electricity sector affects total in-boundary emission for the three cities, not to provide an absolute uncertainty range as the three cities could potentially deviate outside the range based on similar cities. for Delhi, we used the electricity fraction of $44.3 \%$ in Shanghai as the maximum proportion (note this is actually slightly smaller than the value we use for Delhi) of electricity and $36.8 \%$ in Bangkok as the minimum in the same Asian developing regions (including cities: Bangkok, Beijing, and Shanghai) with dense population and relatively poor air quality; for Washington D.C., we chose the maximum proportion of 39.2\% for electricity in energy in New York City and the minimum proportion of $25.2 \%$ in Greater Toronto in terms of the similar economic development and energy consumption culture in North America (including cities: Los Angeles, New York City, and Greater Toronto); for Tokyo, according to the similar energy structure, role of city and economic development in developed countries (including cities: Greater London, New York City, Los Angeles, and Greater Toronto), we adopted the maximum proportion of $39.7 \%$ for electricity in energy in Greater London and the minimum proportion of $25.2 \%$ in Greater Toronto. Then maximum and minimum proportions for electricity in energy were multiplied total energy emissions and were subtracted from energy emissions. The remaining parts of energy emissions refer to the emissions from heating \& industrial fuels and ground transportation, which were added to the large point sources and industrial processes emissions to establish the relevant
in-boundary $\mathrm{CO}_{2}$ emissions. The corresponding in-boundary $\mathrm{CO}_{2}$ emissions with maximum and minimum electricity proportion in energy and the differences between the two methods of city $\mathrm{CO}_{2}$ emission for these three megacities are showed in Table S4 above.

Out of the three cities, we found without the uncertainty estimation that the difference between inventory-based $\mathrm{CO}_{2}$ emission estimate and ODIAC estimate is within $10 \%$ for Delhi, within $30 \%$ for Tokyo, and over $30 \%$ for Washington D.C.. By applying the maximum/minimum electricity portion assumptions, the difference between the two methods of city in-boundary $\mathrm{CO}_{2}$ emission estimates are 7-16\% for Delhi, 5-28\% for Tokyo, and 38-49\% for Washington D.C..

Table S5. New York City population by Manhattan

| Year | Bore share of NYC total (\%) | Population |
| :---: | :---: | :---: |
| 2000 | 19.20 | $1,537,195$ |
| 2010 | 19.24 | $1,585,873$ |
| 2005 | 19.22 | $1,570,274$ |

The population of other 13 cities can be adopted the reported population data in literature directly [1, 4] directly except Manhattan. In absence of population data in Manhattan, we used the open New York City population data by borough from NYC Open Data (https://opendata.cityofnewyork.us/). The NYC Open Data reports the population and population proportion of New York City by borough ever decade from 1950 to 2040. So we simply assumed the bore share of New York City population by Manhattan in 2005 is the average of proportions in 2000 and 2010. Then the average population proportion of $19.22 \%$ was multiplied by the population of New York City in 2005 in Kennedy’s research paper [4] to calculate the population of 1,570,274 for Manhattan in 2005.

Table S6. In-boundary fossil fuel emissions for cities and metropolitan regions

| City | In-boundary FFE ( $\mathrm{tCO} \mathrm{CO}_{2} / \mathrm{cap}$ ) | Total <br> in-boundary <br> $\mathrm{FFE}\left(\mathrm{MtCO}_{2}\right)$ |
| :---: | :---: | :---: |
| Bangkok | 4.86 | 27.53 |
| Beijing | 7.29 | 115.19 |
| Shanghai | 9.91 | 179.91 |
| Delhi | 0.96 | 12.72 |
| Cape Town | 1.75 | 6.12 |
| Sao Paulo | 0.94 | 10.58 |
| Tokyo | 3.15 | 39.94 |
| Greater Paris | 4.36 | 50.30 |
| Greater London | 4.39 | 32.36 |
| Los Angeles | 8.16 | 77.64 |
| Manhattan | 4.74 | 7.44 |
| New York City | 5.28 | 43.13 |
| Washington D.C. | 11.91 | 6.81 |
| Greater Toronto | 7.98 | 44.35 |

Table S7. Differences between calculations of emissions from ground transportation

| City | Fuel sales <br> $(\mathrm{ML})$ | VKT-Fuel <br> consumption(ML) | Scaled fuel <br> consumption <br> $(\mathrm{ML})$ | Difference (\%) |
| :---: | :---: | :---: | :---: | :---: |
| Bangkok | 2741 | 2662 | $\mathrm{~N} / \mathrm{A}$ | -2.9 |
| Greater Toronto | 6691 | 6988 | $\mathrm{~N} / \mathrm{A}$ | 4.4 |
| New York City | $\mathrm{N} / \mathrm{A}$ | 4179 | 4107 | -1.7 |

Emissions from ground transportation in Kennedy et al [5] were calculated by three methods based on local fuel sales within the city, vehicle kilometers travelled (VKT) in the boundary of the city, and scaling from wider regional data. The first method is to use local fuel sales multiplied by the IPCC emissions factors with default. For $\mathrm{CO}_{2}$ emissions, this use of local fuel sales data is preferred by the IPCC and appropriate where the number of daily commuter trips across the city boundary is smaller than the number of trips within the city. The second approach based on gasoline consumptions estimated from VKT is appropriate for central cities where the number of cross-boundary commuter trips is large and the fuel sales may occur out of the central city boundary. This VKT for cities used computer models or by vehicle counting surveys differ between cities due to the different counting techniques and the unique model for each city. The third method is to scale gasoline consumptions from state, provincial, or wider regional data based on the consumption that vehicle in the city travel the same average annual kilometers as in the wider region. And the scaling factor may be determined by population corresponding to the total travel commute times. Although the third method is not desirable as other two approaches, this method is necessary when there is no reliable data in the city. In order to estimate the greatest source of uncertainty caused by ground transportation calculation, the first method was applied to Cape Town, Greater Toronto, and Bangkok; the second method was established for Bangkok, Greater London, and New York City; moreover, the third approach was used for Los Angeles, New York City, and Greater Toronto. Three of the cities, Greater Toronto, New York City, and Bangkok were applied by two different methods of these three separately. The comparisons between the calculations of these three approaches show that the differences can be less than $5 \%$ and these three methods can establish reasonably close estimates.

Table S8. Differences between calculations of emissions from DARTE and inventory -based data

| City | Year <br> (Kennedy) | Area (km2) | Ground <br> Transportation(t <br> CO2) | DARTE(MgCO2) | Difference <br> $(\%)$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| Los Angeles | 2000 | 10,612 | $46,835,143.0$ | $42,842,342.9$ | 9.3 |
| New York <br> City | 2005 | 807 | $12,500,100.0$ | $11,588,570.2$ | 7.9 |
| Washington <br> D.C. | 2000 | 178 | $3,922,019.8$ | $1,667,169.2$ | 135.3 |
| Manhattan | 2005 | 69 | $2,402,519.2$ | $1,155,456.9$ | 107.9 |

Table S9. Differences between CO2 emissions by electricity sector from eGRID and inventory -based data

| City | Year(Kennedy) | Area <br> $(\mathrm{km} 2)$ | eGrid plant annual <br> CO2 <br> emissions (t CO2) | CARMA <br> (t CO2) | Difference (\%) |
| :---: | :---: | :---: | :---: | :---: | :---: |
| Los Angeles | 2000 | 10,612 | $16,895,543.2$ | $15,672,507.2$ | -7.2 |
| New York <br> City | 2005 | 807 | $15,153,227.8$ | $5,061,039.6$ | -66.6 |
| Washington <br> D.C. | 2000 | 178 | $191,797.2$ | $120,541.9$ | -37.2 |
| Manhattan | 2005 | 69 | $1,888,797.3$ | $122,144.3$ | -93.5 |

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