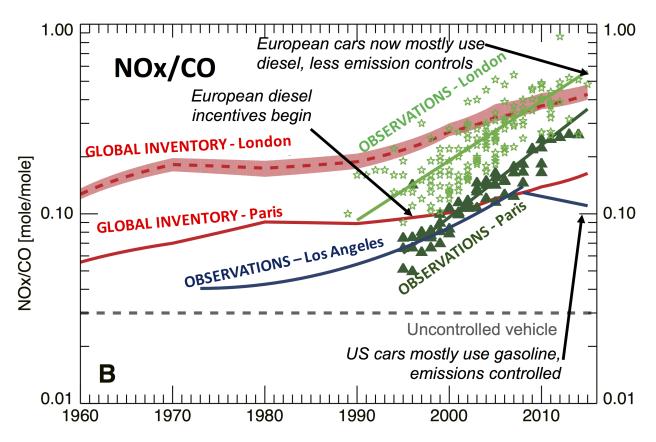
## Analysis of Long-term Observations of $NO_x$ and CO in Megacities and Application to Constraining Emissions Inventories

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Long-term atmospheric NO<sub>x</sub>/Carbon Monoxide (CO) enhancement ratios in megacities provide evaluations of emission inventories. A fuel based emissions inventory approach that diverges from traditional bottom-up inventory methods explains 1970 – 2015 trends in NO<sub>x</sub>/CO enhancement ratios in Los Angeles, CA. Combining this comparison with similar measurements in other U.S. cities demonstrates that motor vehicle emissions controls were largely responsible for U.S. urban NO<sub>x</sub>/CO trends in the past half-century. Differing NO<sub>x</sub>/CO enhancement ratio trends in U.S. and European cities over the past 25 years highlight alternative strategies for mitigating transportation emissions, reflecting Europe's increased use of light-duty diesel vehicles and correspondingly slower decreases in NO<sub>x</sub> emissions compared to the U.S. A global inventory widely used by chemistry-climate models fails to capture long-term trends and regional differences in U.S. and Europe megacity NO<sub>x</sub>/CO enhancement ratios, possibly contributing to these models' inability to accurately reproduce observed long-term changes in tropospheric ozone.



**Figure 1.**  $NO_x/CO$  enhancement ratios from roadside monitor measurements at air quality monitoring stations in Paris (dark green symbols) and London (light green stars), along with log-linear trends for the Paris (dark green line) and London (light green light) monitoring data. The trend for analogous Los Angeles air quality monitoring observations is shown for comparison (blue line). Also shown are the MACCity global inventory's  $NO_x/CO$  emissions ratios for Paris (solid red line) and averaged for all London grid cells (dashed red line) and the range for individual London grid cells (red shading).