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Sources of greenhouse gases and carbon monoxide in central London (UK)

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Biosphere-atmosphere exchange of carbon dioxide (CO_2) has been on the scientific agenda for several decades and new technology now also allows for high-precision, continuous monitoring of fluxes of methane (CH_4) and nitrous oxide (N_2O). Compared to the natural environment, flux measurements in the urban environment, which is home to over 50% of the population globally, are still rare despite high densities of anthropogenic sources of pollutants. We report on over three years of measurements atop a 192 m tower in central London (UK), Europe's largest city, which started in October 2011.

Fluxes of methane, carbon monoxide (CO) and carbon dioxide are measured by eddy-covariance (EC) at the British Telecom tower (51˚ 31' 17.4" N 0˚8' 20.04" W). In addition to the long-term measurements, EC fluxes of nitrous oxide (N_2O) were measured in February 2014.

All four trace gases exhibit diurnal trends consistent with anthropogenic activities with minimum emissions at night and early afternoon maxima. Segregating emissions by wind direction reveals heterogeneous source distributions with temporal patterns and source strengths that differ between compounds. The lowest emissions for CO, CO_2 and CH_4 were recorded for NW winds. The highest emissions of methane were in the SE sector, in the NE for CO_2 and in the W for CO. Fluxes of all 3 gases exhibited marked seasonal trends characterised by a decrease in emissions in summer (63% reduction for CO, 36% for CO_2 and 22% for CH_4). Monthly fluxes of CO and CO_2 were linearly correlated to air temperature (R^2 = 0.7 and 0.59 respectively); a weaker dependence upon temperature was also observed for CH_4 (R^2 = 0.31).

Diurnal and seasonal emissions of CO and CO_2 are mainly controlled by local fossil fuel combustion and vehicle cold starts are thought to account for 20-30% of additional emissions of CO during the winter. Fugitive emissions of CH_4 from the natural gas distribution network are thought to be substantial, which is consistent with the weaker seasonality of CH_4 fluxes compared with CO and CO_2. Annual estimates of CO_2 emissions (41 kt km^{-2}) obtained by EC were consistent with data upscaled from the London Atmospheric Emissions Inventory (LAEI; 46 kt km^{-2}). Good agreement between measurements and inventory data was also found for CO (measured 156 t km^{-2}; LAEI 145 t km^{-2}) and for N_2O (measured 0.36 t km^{-2}; LAEI 0.42 t km^{-2}), although based on a much shorter measurement period. By contrast, a two-fold difference was found between inventory and measured CH_4 fluxes (measured 75 t km^{-2}; LAEI 34 t km^{-2}), which could indicate an underestimation by the inventory of CH_4 emissions from anthropogenic sources or the existence of unaccounted biogenic sources. Measurements of isotopic CH_4 taken 2 km SE of the tower near the banks of the river Thames reveal multiple episodes of ^{13}C-depleted morning peaks consistent with biogenic sources. We speculate that the Thames can act as an additional significant source of biogenic methane especially at low tide and after heavy rainfall, which could explain the large emissions observed in the S-SE sector.