Geophysical Research Abstracts
Vol. 16, EGU2014-11828, 2014
EGU General Assembly 2014

Airborne observed and receptor-oriented modelled urban increments of anthropogenic CO_2, CO and NOX concentrations in the megacity of London in summer 2012

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A better characterization of the emissions and the dynamics of anthropogenic CO_2 in large-urban centres are needed to implement more effective mitigation measures to combat climate change. This study aims to establish a representative emissions ratio of anthropogenic CO_2 (CO_2ff) in the megacity of London using CO and NOX as tracers. Observations of CO_2, CO and NOX mixing ratios obtained onboard the NERC-ARSF aircraft undertaken on 12 July 2012 over the city of London were used. Airborne observations were taken at ~380 m along four transects crossing London, two in the morning (10:30 to 12:30 GMT) and two in the afternoon (15:30-16:30 GMT). The ratio of the amounts of CO and CO_2 in excess of natural abundances (denoted as ΔCO and ΔCO_2, respectively) from the airborne observations was used to determine the fraction of CO_2 derived from burning fossil fuels (CO_2ff). Total observations of CO and CO_2 were compared to NOX observations and background concentrations were determined as the intercept when NOX mixing ratios equalled zero derived from standardised major axis linear regression. Excess concentrations were calculated by subtracting total amounts minus the background. ΔCO showed good correlation with ΔCO_2 in the morning transects (R=0.95) but not in the afternoon (R=-0.50). The mean ($\pm 1\sigma$) CO/CO_2ff was derived from linear regression using the morning measurements and valued 5.0±0.4 ppb ppm-1.

Lagrangian Particle Dispersion (LPD) simulations in backward mode were undertaken to model urban increments of anthropogenic CO_2 and CO and to calculate the emissions ratio from the emissions inventory EDGAR v4.2. The LPD model FLEXPART was run with the meteorological data from the European Centre for Medium-Range Weather Forecasts (spatial resolution of 0.2 x 0.2 degrees; 91 vertical levels) and multiplied with the EDGAR emissions inventory (spatial resolution 0.1 x 0.1 degrees) to obtain an increment at each receptor point along the transects. Annual and temporal scaled emissions were used. Modelled urban increments of CO correlated well with observations of excess CO (R>0.81) with a slope close to 1:1 when using scaled emissions (0.69-1.10, 2σ). Modelled CO_2ff correlated well with observed excess CO_2 when using temporal scaled emissions (R=0.83) with a slope ranging from 0.58:1 to 0.99:1 (2σ). The CO/CO_2ff ratio from modelled increments was 5.8±0.6 ppb ppm-1.

Results show the CO/CO_2ff ratio from observed excess abundances and from the EDGAR emissions inventory were in accordance for the city of London for the survey campaign. Future studies will involve using other emissions inventories such as the National Atmospheric Emissions Inventory (NAEI) for the UK and the London Atmospheric Emissions Inventory (LAEI); and model urban increments of other airborne campaigns.