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Airborne Flux Measurements of Volatile Organic Compounds and NO_x over a European megacity

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Ground level ozone (O_3) and nitrogen dioxide (NO_2) are priority pollutants whose concentrations are closely regulated by European Union Air Quality Directive 2008/50/EC. O_3 is a secondary pollutant, produced from a complex chemical interplay between oxides of nitrogen (NO_x = NO + NO_2) and volatile organic compounds (VOCs). Whilst the basic atmospheric chemistry leading to O_3 formation is generally well understood, there are substantial uncertainties associated with the magnitude of emissions of both VOCs and NO_x. At present our knowledge of O_3 precursor emissions in the UK is primarily derived from National Atmospheric Emission inventories (NAEI) that provide spatially disaggregated estimates at 1x1km resolution, and these are not routinely tested at city or regional scales. Uncertainties in emissions propagate through into uncertainties in predictions of air quality in the future, and hence the likely effectiveness of control policies on both background and peak O_3 and NO_2 concentrations in the UK.

The Ozone Precursor Fluxes in the Urban Environment (OPFUE) project aims to quantify emission rates for NO_x and selected VOCs in and around the megacity of London using airborne eddy covariance (AEC). The mathematical foundation for AEC has been extensively reviewed and AEC measurements of ozone, dimethyl sulphide, CO_2 and VOCs have been previously reported.

During the summer of 2013, approximately 30 hours of airborne flux measurements of toluene, benzene, NO and NO_2 were obtained from the NERC Airborne Research and Survey Facility's (ARSF) Dornier-228 aircraft. Over SE England, flights involved repeated south west to north east transects of $\sim$50 km each over Greater London and it's surrounding suburbs and rural areas, flying at the aircraft's minimum operating flight altitude and airspeed ($\sim$300m, 80m/s). Mixing ratios of benzene and toluene were acquired at 2Hz using a proton transfer reaction mass spectrometer (PTR-MS) and compared to twice hourly whole air canister samples (WAS) which were quantitatively determined post-flight via TD-GCMS. Mixing ratios between the PTR-MS and WAS-TD-GCMS were in good agreement with R^2 values of 0.8 and 1.0 for Toluene and Benzene respectively. Mixing ratios of NO and NO_2 were acquired at 10Hz using a 2 channel NO_x chemiluminescence detector with photolytic converter. These measurements were used with 20 Hz 3-D wind vector data from an AIMMS-20 turbulence probe on the aircraft to calculate highly spatially resolved (1 km) surface-to-atmosphere emission flux rates of these compounds using the eddy covariance method of Karl et al. (2013).

Measured surface to atmosphere emission fluxes of NO_x, benzene and toluene from London were between 40 – 91 mg m2 h-1, 0.1 – 0.4 mg m2 h-1 and 0.2 – 2 mg m2 h-1 respectively, showing the spatial flux heterogeneity over the city.

This demonstrates for the first time the feasibility of airborne eddy covariance flux measurements of reactive NO_x species. We also show the applicability of wavelet analysis using virtually disjunct eddy covariance measurements of anthropogenic compounds in estimating regional fluxes over a European megacity. We compare our measured emission rates with those estimated from "bottom-up" emissions inventories and highlight the agreement between the two.