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Chemically-speciated aerosol fluxes above three UK cities

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There is considerable uncertainty regarding the processes controlling the formation and transformation of organic aerosol (OA) in the atmosphere. Regional aerosol, advected into the city, mixes with the primary urban emissions of particles and VOCs to create an environment where particle composition is controlled by a number of factors, not least chemistry and complex urban meteorology. Measurements of chemically-speciated particle fluxes above urban areas can give us insight into exchanges between urban centres and the free troposphere, as well as the chemical and physical processing which occurs during transport.

We present chemically-speciated aerosol fluxes and concentrations measured three contrasting cities in the United Kingdom. An aerodyne quadropole-AMS was deployed on a tall building in the three urban centers; the Nelson Monument in Edinburgh, the Portland Tower in Manchester and the BT Tower in London. We use the coupled eddy covariance-AMS system as described in Nemitz *et al*., 2008 to measure the flux of non-refractory (NR) PM1 above the three cities. Supporting measurements of CO and CO_2 fluxes made by eddy covariance are also presented and emission factors relative to these pollution tracers have been calculated.

The organic aerosol component of the NR-PM1 mass has been further analysed using statistical techniques to ascertain the contribution to the OA flux and concentration of so-called hydrocarbon-like organic aerosol (HOA) and oxygenated organic aerosol (OOA). OA dominates the mass of PM1 in all three cities with nitrate the next major component of the mass in Edinburgh and Manchester and sulfate the next major component in London. HOA dominates the OA mass in Edinburgh and for some time periods in Manchester, in contrast to the finding of Zhang *et al*., for urban areas and situation in London where OOA was the larger proportion of the OA mass.

At all locations, HOA has a large relative emission flux relative to OOA. The HOA fluxes are generally are generally upward and show diurnal trend. Average flux densities of hydrocarbon-like organic aerosol (HOA) range from 63 ng m^{-2} s^{-1} in Edinburgh to 440 ng m^{-2} s^{-1} in London. Excellent correlation was found between emission fluxes of CO and CO_2. However, HOA fluxes show a much weaker correlation with these gases. Changes in the relative emission strengths of HOA and CO_2 are likely related to changes in the fuel mix, vehicle fleet and the changing contribution of building heating. However, the relatively constant CO/CO_2 flux ratio suggests that i) these changes were not pronounced and ii) was without a significant biogenic component; implying the CO_2 emission flux was due mainly to combustion. Assuming that both HOA and CO_2 (and CO) originate from similar sources, the measurements suggest that HOA undergoes considerable processing between emission and measurement of the flux high above the urban centre. The fraction of HOA that is evaporated and/or chemically processed is likely to depend on measurement height, temperature, radiation, photochemistry, and transport time-scale. Nevertheless, as the transport time-scale during these measurements is on the order of minutes to tens of minutes, the flux ratio HOA/CO_2 is likely to be more representative than emission ratios derived from urban concentration ratios, which accumulate over tens of minutes to hours. Emission ratios range from 0.24 to 1.37 g $(kg\ C)^{-1}$.

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