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Extractive Electrospray Ionisation Mass Spectrometry (EESI-MS) for organic aerosol analysis

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Organic compounds are ubiquitous in the troposphere and constitute a large fraction of aerosol mass. Characterising the composition of organic aerosols (OA) is key to understanding their formation, transformation and impacts on atmospheric chemistry and air quality. Here, we present a novel online technique for characterising OA, Extractive Electrospray Ionisation Mass Spectrometry (EESI-MS) [1]. EESI-MS employs gentle solvent-based capture and ionisation of aerosols. This generates molecular ions and allows individual aerosol species to be identified and monitored over time.

We will show that EESI-MS is an excellent tool for characterising OA generated in laboratory experiments, focusing on the quantitative abilities of the technique. Unlike related offline techniques, changes in EESI ion abundance provide a reliable measure of aerosol-phase concentration changes during aerosol evolution [2]. This "relative quantification" has been exploited in recent work to monitor composition changes during heterogeneous oxidation of oleic acid aerosols [3]. We extract previously unconstrained kinetic parameters related to condensed-phase reactions of Criegee intermediates using EESI-MS and process-based aerosol modelling.

Furthermore, we establish correlations between organic aerosol toxicity and chemical composition for OA formed from the prominent indoor pollutant and biogenic VOC, limonene [4]. We compare EESI spectra to complementary measurements of health-relevant reactive oxygen species (ROS). We find persistent levels of ROS are present in limonene OA, and hypothesise that the aerosol contains a reservoir of reactive species, including unsaturated compounds, which generate additional ROS over time.

[1] Gallimore and Kalberer, ES&T, 47, 7324-31, 2013.
[2] Gallimore et al., ACP, 17, 14485–500, 2017.
[3] Gallimore et al., JGRA, 122, 4364–77, 2017.
[4] Gallimore et al., ACP, 17, 9853–68, 2017.