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Seasonal differences of urban organic aerosol composition - an ultra-high resolution mass spectrometry study

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The understanding of the chemical composition of atmospheric aerosols, their properties and reactivity are important for assessing aerosol effects upon both global climate change and human health. The composition of organic aerosols is poorly understood mainly due to their highly complex chemical composition with several thousand compounds. In the present study the water-soluble organic fraction of ambient particles collected at an urban site in Cambridge, UK, during different seasons were analysed with ultra-high resolution mass spectrometry. For several thousand peaks in the mass specta (between 3000-6000) an elemental composition could be assigned and summer samples generally contained more components than winter samples. Up to 80% of the peaks in the mass spectra contain nitrogen and/or sulphur functional groups and only about 20% of the compounds contain only C, H and O atoms. In summer the fraction of compounds with oxidized nitrogen and sulphur groups increases compared to winter indicating a photo-chemical formation route of these multifunctional compounds. In addition to oxidized nitrogen compounds a large number of highly unsaturated reduced nitrogen-containing compounds were detected, corresponding likely to cyclic amines. A significant number of oxidized PAHs have been detected in summer samples, which were not present in winter, indicating again photo-chemical aging processes. Both, amines and long-chain aliphatic acids (also frequently observed in these urban samples) are likely signatures of biomass burning and primary biological sources. Potential biomass burning markers are discussed. Particle-phase oligomerisation reactions have only been observed to a very limited degree. Compounds larger than m/z 350 almost exclusively contained N and/or S functional groups indicating that the high molecular weight compounds in these organic aerosol extracts might be mainly due to particle-phase heterogeneous reactions of organic compounds with inorganic particle components (such as sulphate and nitrate) and are not formed predominantly in organic oligomerisation reactions as it is observed frequently in laboratory and smog chamber studies.