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Application of Spin Traps to Detect Reactive Intermediates and Reactive Oxygen Species in Secondary Organic Aerosol

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The chemical composition and evolution of secondary organic aerosol (SOA) in the atmosphere represents one of the largest uncertainties in our current understanding of air quality. Studies suggest that reactive oxygen species (ROS), either present in ambient aerosol and exogenously delivered or generated by particle components *in vivo*, are potentially a major contributor to aerosol toxicity. Several methods are currently available for probing ROS concentrations in aerosol, however, the role of radicals and reactive intermediates in both the formation and ageing of aerosol, as well as their contribution to the health-relevant properties of ambient aerosol, at present remains highly uncertain. Recently, a study by Stevanovic *et al.*[1] introduced the profluorescent spin trap BPEAnit (9,10-bis-(phenylethynyl)-anthracene-nitroxide), where they subsequently used this method to quantify particle-bound radicals in aerosol generated from the combustion of biomaterial and fuel.

In this work, BPEAnit is applied to study the concentrations of particle-bound radicals in SOA. Radical concentrations were estimated for SOA generated from the ozonolysis of α-pinene, β-caryophyllene and limonene in a flow tube, to probe the assays response to SOA formed from biogenic volatile organic compounds. SOA is captured in an impinger containing BPEAnit/DMSO, before subsequent analysis using fluorescent spectroscopy. Using this method, a pseudo-1^{st} order rate constant of $k = 7.3 \pm 1.7 \times 10^{-3}$ s^{-1} was derived for the overall decay of organic radicals in α-pinene SOA, implying a radical lifetime on the order of several minutes. Furthermore, building on our recent work in Giorio *et al.*,[2] the spin trap PBN (N-*tert*-butyl-α-phenylnitrone) was used to trap Criegee intermediates (CIs) present in the aerosol phase, with the resulting adducts analysed using high performance liquid chromatography high-resolution mass spectrometry. Experiments were conducted with this method using SOA generated from β-caryophyllene ozonolysis in a flow tube, demonstrating the technique's unique capability to detect multiple CIs present in SOA and representing the first detection of CIs in organic aerosol. Detected CIs were estimated to constitute approximately 0.8% of the total particle mass, demonstrating the potential of this method to provide quantitative data of condensed phase CIs.