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Atmos. Chem. Phys., 6, 403-418, 2006
www.atmos-chem-phys.net/6/403/2006/
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Simulating regional scale secondary organic aerosol formation during the TORCH 2003 campaign in the southern UK

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Abstract. A photochemical trajectory model has been used to simulate the chemical evolution of air masses arriving at the TORCH field campaign site in the southern UK during late July and August 2003, a period which included a widespread and prolonged photochemical pollution episode. The model incorporates speciated emissions of 124 non-methane anthropogenic VOC and three representative biogenic VOC, coupled with a comprehensive description of the chemistry of their degradation. A representation of the gas/aerosol absorptive partitioning of ca. 2000 oxygenated organic species generated in the Master Chemical Mechanism (MCM v3.1) has been implemented, allowing simulation of the contribution to organic aerosol (OA) made by semi- and non-volatile products of VOC oxidation; emissions of primary organic aerosol (POA) and elemental carbon (EC) are also represented. Simulations of total OA mass concentrations in nine case study events (optimised by comparison with observed hourly-mean mass loadings derived from aerosol mass spectrometry measurements) imply that the OA can be ascribed to three general sources: (i) POA emissions; (ii) a "ubiquitous" background concentration of $0.7 \mu\text{g m}^{-3}$; and (iii) gas-to-aerosol transfer of lower volatility products of VOC oxidation generated by the regional scale processing of emitted VOC, but with all partitioning coefficients increased by a species-independent factor of 500. The requirement to scale the partitioning coefficients, and the implied background concentration, are both indicative of the occurrence of chemical processes within the aerosol which allow the oxidised organic species to react by association and/or accretion reactions which generate even lower volatility products, leading to a persistent, non-volatile secondary organic aerosol (SOA). The contribution of secondary organic material to the simulated OA results in significant elevations in the simulated ratio of organic carbon (OC) to EC, compared with the ratio of 1.1 assigned to the emitted components. For the selected case study events, $[\text{OC}]/[\text{EC}]$ is calculated to lie in the range 2.7-9.8, values which are comparable with the high end of the range reported in the literature.

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Citation: Johnson, D., Utembe, S. R., Jenkin, M. E., Derwent, R. G., Hayman, G. D., Alfarra, M. R., Coe, H., and McFiggans, G.: Simulating regional scale secondary organic aerosol formation during the TORCH 2003 campaign in the southern UK, *Atmos. Chem. Phys.*, 6, 403-418, 2006. [Bibtex](#) [EndNote](#) [Reference Manager](#)