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Net Ozone Production during the Chemical Evolution of Biomass Burning Plumes: Evaluation of a Hierarchy of Chemical Mechanisms

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As part of the "Quantifying the impact of BOReal forest fires on Tropospheric oxidants over the Atlantic using Aircraft and Satellites" (BORTAS) experiment, a range of atmospheric models incorporating a hierarchy of chemical mechanisms (from explicit to those suitable for integration by global CTMs) were used, in concert with a comprehensive suite of aircraft observations, to investigate (multiday) O_3 production and loss within biomass burning outflow over the western boundary of the North Atlantic and the resultant perturbation to tropospheric chemistry.

The Master Chemical Mechanism (MCM; http://mcm.leeds.ac.uk/MCM) is a near-explicit chemical mechanism used in a wide variety of science and policy applications where chemical detail is required. It is used either directly, or as a benchmark representation against which to develop and optimise reduced chemical schemes. The "common representative intermediates" (CRI) mechanism developed in parallel with the MCM, provides a more economical alternative whilst maintaining an adequate treatment of the formation of O_3. The CRI mechanism contains a series of generic intermediate radicals and products (based on the ozone creation potential for a given VOC) each of which is able to represent a larger set of species in the MCM. These generic intermediates mediate the breakdown of larger VOCs into smaller fragments, the chemistry of which is treated explicitly.

Detailed photochemical models incorporating both the MCMv3.2 and CRIv2 mechanisms have been constrained with BORTAS aircraft observations in order to investigate the chemistry controlling ozone formation and loss and NO_y speciation along each flight track as well as forward and back trajectory simulations incorporating both aircraft observations and bottom up biomass burning emission inventories. Such simulations have been used to evaluate the CRI mechanism and subsequent reduced versions, as well as GEOS-Chem chemistry, against the benchmark MCM and to evaluate the initialisation and speciation methodologies applied during the bottom up model studies. For the majority of the simulations, the CRI 5-10 day ozone time profiles match reasonably well with the MCM simulations, with the majority of the photochemical ozone being formed over the first 12-15 hours of daylight after emission. Where forward trajectory simulations intersect with the aircraft flight tracks, comparisons of simulated and measured reactive species are in reasonably good agreement.