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Title: Observation and modelling of HOx radicals in a boreal forest

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Abstract:

Measurements of OH and HO2 radicals were conducted in a pine-dominated forest in southern Finland during the HUMPPA-COPEC-2010 (Hyytiälä United Measurements of Photochemistry and Particles in Air - Comprehensive Organic Precursor Emission and Concentration study) field campaign in summer 2010. Simultaneous side-by-side measurements of hydroxyl radicals were conducted with two instruments using chemical ionization mass spectrometry (CIMS) and laserinduced fluorescence (LIF), indicating small systematic disagreement, OHLIF / OHCIMS Combining double low line (1.31 ± 0.14). Subsequently, the LIF instrument was moved to the top of a 20 m tower, just above the canopy, to investigate the radical chemistry at the ecosystematmosphere interface. Comprehensive measurements including observations of many volatile organic compounds (VOCs) and the total OH reactivity were conducted and analysed using steady-state calculations as well as an observationally constrained box model.

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> Production rates of OH calculated from measured OH precursors are consistent with those derived from the steady-state assumption and measured total OH loss under conditions of moderate OH reactivity. The primary photolytic sources of OH contribute up to one-third to the total OH production. OH recycling, which occurs mainly by HO2 reacting with NO and O3, dominates the total hydroxyl radical production in this boreal forest. Box model simulations agree with measurements for hydroxyl radicals (OHmod. / OHobs. Combining double low line 1.00 ± 0.16), while HO2 mixing ratios are significantly under-predicted (HO2mod. / HO2obs. Combining double low line 0.3 ± 0.2), and simulated OH reactivity does not match the observed OH reactivity. The simultaneous under-prediction of HO2 and OH reactivity in periods in which OH concentrations were simulated realistically suggests that the missing OH reactivity is an unaccounted-for source of HO 2.

br> Detailed analysis of the HOx production, loss, and recycling pathways suggests that in periods of high total OH reactivity there are additional recycling processes forming OH directly, not via reaction of HO2 with NO or O3, or unaccountedfor primary HO x sources. Under conditions of moderate observed OH reactivity and high actinic flux, an additional RO2 source of approximately 1 × 106 molec cm-3 s-1 would be required to close the radical budget. Nevertheless, a major fraction of the OH recycling occurs via the reaction of HO2 with NO and O3 in this terpene-dominated environment.

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