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Simulating atmospheric composition over a South-East Asian tropical rainforest: performance of a chemistry box model

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Abstract. Atmospheric composition and chemistry above tropical rainforests is currently not well established, particularly for south-east Asia. In order to examine our understanding of chemical processes in this region, the performance of a box model of atmospheric boundary layer chemistry is tested against measurements made at the top of the rainforest canopy near Danum Valley, Malaysian Borneo. Multi-variate optimisation against ambient concentration measurements was used to estimate average canopy-scale emissions for isoprene, total monoterpenes and nitric oxide. The excellent agreement between estimated values and measured fluxes of isoprene and total monoterpenes provides confidence in the overall modelling strategy, and suggests that this method may be applied where measured fluxes are not available, assuming that the local chemistry and mixing are adequately understood. The largest contributors to the optimisation cost function at the point of best-fit are OH (29%), NO (22%) and total peroxy radicals (27%). Several factors affect the modelled VOC chemistry. In particular concentrations of methacrolein (MACR) and methyl-vinyl ketone (MVK) are substantially overestimated, and the hydroxyl radical (OH) concentration is substantially underestimated; as has been seen before in tropical rainforest studies. It is shown that inclusion of dry deposition of MACR and MVK and wet deposition of species with high Henry's Law values substantially improves the fit of these oxidised species, whilst also substantially decreasing the OH sink. Increasing OH production arbitrarily, through a simple OH recycling mechanism , adversely affects the model fit for volatile organic compounds (VOCs). Given the constraints on isoprene flux provided by measurements, a substantial decrease in the rate of reaction of VOCs with OH is the only remaining option to explain the measurement/model discrepancy for OH. A reduction in the isoprene+OH rate constant of 50%, in conjunction with increased deposition of intermediates and some modest OH recycling, is able to produce both

isoprene and OH concentrations within error of those measured. Whilst we cannot rule out an important role for missing chemistry, particularly in areas of higher isoprene flux, this study demonstrates that the inadequacies apparent in box and global model studies of tropical VOC chemistry may be more strongly influenced by representation of detailed physical and micrometeorological effects than errors in the chemical scheme.

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