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Quantifying the impact of model uncertainties on chemical budgets in trans-Atlantic pollution transport

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Chemical transport models rely upon parameterised gas-phase reaction and photolysis rates, based on data from laboratory studies. These data have associated uncertanties that may impact upon the prediction of the concentrations of key species, such as O3, OH and nitrogen oxides. A unique set of linked aircraft observations in single air masses from the ICARTT (International Consortium for Atmospheric Research on Transport and Transformation) pseudo-Lagrangian experiment allow the isolation of chemical change from transport. We exploit this to investigate the impact of chemical model uncertainties on tropospheric photochemistry in plumes undergoing long-range transport. The aim is to characterise key process which contribute to large uncertainty in oxidant budgets, and identify rates and processes that are a priority for better quantification in the lab. The impact of uncertainties in bimolecular, termolecular and photolysis rates has been investigated using a tropospheric trajectory chemical transport model (CiTTyCAT) and the linked aircraft observations in both anthropogenic polluted and biomass burning air masses. A Monte-Carlo methodology is used to sample the parameter space and ensembles of several thousand model runs generated to provide a firm statistical basis for analysis. Initial results show that uncertainty in ozone concentration is greater than 30% after six days for the case study in question as a result of the bimolecular and termolecular reaction rates. We will present details of key photochemcial uncertainties that result in large uncertainty in ozone photochemistry.