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Quantifying uncertainty in the tropospheric ozone budget as represented in global CTMs

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The ability of current global chemistry-transport models to simulate tropospheric composition and its variability has been tested in a number of recent model intercomparison studies organised under the ACCENT/IPCC and RETRO projects. While the general agreement in tropospheric ozone between these models, and between models and observations, is reassuring, the studies have highlighted significant differences between models in their representation of the processes controlling it. These differences may be due to natural processes (e.g., variability in meteorology), numerical limitations (e.g., model resolution) or to genuine differences in scientific understanding. Identifying the source of these differences is important for reducing the uncertainty in model ozone budgets and for interpreting the results of multi-model 'ensemble' studies. In this study the FRSGC/UCI CTM has been rerun 70-80 times to investigate the sensitivity of the calculated ozone budget to different assumptions about the physical and chemical processes controlling it, to differences in meteorological data and to model resolution. While much of the difference between previous model studies originates in the use of different emissions, differences in recent, better-constrained intercomparison studies are dominated by differences in cross-tropopause transport, deposition, humidity, and lightning. Contrasting influences on the lifetimes of ozone and methane suggest that differences in the magnitude and location of lightning emissions of NOx are currently a greater source on uncertainty for the ozone budget than differences in water vapour or convection. Increases in horizontal and vertical resolution reduce ozone production and cross-tropopause transport and increase surface deposition, but interannual variability in meteorological fields may affect these ozone budget terms by a similar magnitude. More focused studies are required to target the key weaknesses in current models so that the uncertainty in simulation of tropospheric composition and its variability may be reduced.