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Modelling the global tropospheric ozone budget: exploring the variability in current models

O. Wild
Frontier Research Center for Global Change, JAMSTEC, Yokohama, Japan
now at: Centre for Atmospheric Science, University of Cambridge, UK

Abstract. What are the largest uncertainties in modelling ozone in the troposphere, and how do they affect the calculated ozone budget? Published chemistry-transport model studies of tropospheric ozone differ significantly in their conclusions regarding the importance of the key processes controlling the ozone budget: influx from the stratosphere, chemical processing and surface deposition. This study surveys ozone budgets from previous studies and demonstrates that about two thirds of the increase in ozone production seen between early assessments and more recent model intercomparisons can be accounted for by increased precursor emissions. Model studies using recent estimates of emissions compare better with ozonesonde measurements than studies using older data, and the tropospheric burden of ozone is closer to that derived here from measurement climatologies, 335±10 Tg. However, differences between individual model studies remain large and cannot be explained by surface precursor emissions alone; cross-tropopause transport, wet and dry deposition, humidity, and lightning also make large contributions. The importance of these processes is examined here using a chemistry-transport model to investigate the sensitivity of the calculated ozone budget to different assumptions about emissions, physical processes, meteorology and model resolution. The budget is particularly sensitive to the magnitude and location of lightning NO_x emissions, which remain poorly constrained; the 3–8 TgN/yr range in recent model studies may account for a 10% difference in tropospheric ozone burden and a 1.4 year difference in CH_4 lifetime. Differences in humidity and dry deposition account for some of the variability in ozone abundance and loss seen in previous studies, with smaller contributions from wet deposition and stratospheric influx. At coarse model resolutions stratospheric influx is systematically overestimated and dry deposition is underestimated; these differences are 5–8% at the 300–600 km grid-scales investigated here, similar in magnitude to the changes induced by interannual variability in meteorology. However, a large proportion of the variability between models remains unexplained, suggesting that differences in chemical mechanisms and dynamical schemes have a large impact on the calculated ozone budget, and these should be the target of future model intercomparisons.

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