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Impact of chemical boundary conditions on modelled regional-scale ozone and particulate matter concentrations in the United Kingdom

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Regional air quality modelling relies on accurate representation of transport of chemical species through the boundaries of the modelling domain. Methods to generate chemical boundary conditions for regional air quality simulations varies from the use of constant background concentrations of selected tracers to dynamically varying boundary conditions from global chemical transport models. In this study we evaluate the sensitivity and performance of regional model predictions to four different sets of boundary conditions. We have employed the regional chemical transport model, WRF-CMAQ with boundary conditions from four global chemical transport models: STOCHEM, GEMS, GEOS-Chem, and MACC. The model simulations have been performed for a European domain at 50 km x 50 km horizontal grid resolution and a UK domain at 10 km x 10 km horizontal grid resolution. To study the seasonal variations in the prediction of regional air quality we have performed model simulations for one winter month (January) and one summer month (July) in year 2006. The variations in the simulated spatial distribution of ground level ozone (O_3) and particulate matter ($PM_{2.5}$ and PM_{10}) concentrations show that model simulations are sensitive to the specification of the boundary conditions especially during the winter month. The comparison of modelled and observed vertical distribution of ozone showed that all the model simulations reproduced the observed vertical structure of ozone well, although with some underestimation. The vertical structure simulated in CMAQ with MACC boundary conditions performed better compared to CMAQ simulations using STOCHEM, GEMS, and GEOS-Chem. The comparison of hourly time series of modelled and observed (UK AURN urban and rural stations) O_3 and PM_{10} concentrations revealed that the CMAQ simulation with MACC boundary conditions overestimates ground surface O_3 concentrations (mean bias of 1.443 μg m^{-3} for winter and 8.9 μg m^{-3} for summer) and underestimates ground surface PM_{10} concentrations (mean bias of -4.7 μg m^{-3} for winter and -9.1 μg m^{-3} for summer). The benefit of using dynamically varying boundary conditions will be highlighted for air pollution episodes driven by long-range transport.