Attribution of tropospheric ozone to precursor emissions through improved tagging techniques

Tim Butler

Institute for Advanced Sustainability Studies

Potsdam, Germany

For decades, air quality models have done a reasonable job of simulating high ozone episodes in urban areas, but have been less successful with simulating regional and global background ozone. Increasingly strict air quality regulations along with a rise in background ozone in some regions leads to the situation in which background concentrations can play an increasing role in non-attainment of air quality standards. Uncertainties in STE (Stratosphere-Troposphere Exchange) and emissions of ozone precursors contribute to the poor performance of models with respect to background ozone, but recent work has also shown that model representations of chemical processes may also be contributing to this. In particular, the long-lived oxidation intermediates of slowly reacting VOC (Volatile Organic Compound) species have the potential to produce more ozone than previously thought. This chemistry is currently not well represented in the chemical mechanisms used in state-of-the-art air quality and chemistry-climate models, which generally use lumped representations of VOC species.

Additionally, global simulations of tropospheric ozone indicate a strong link between increasing concentrations of methane in the atmosphere and increasing concentrations of tropospheric background ozone. The mechanisms of this link have not been explored in great detail, especially given that most of the remote troposphere is distant from anthropogenic emission sources, and are likely to be in a NOx (oxides of nitrogen)-sensitive chemical regime. If global background tropospheric ozone is so sensitive to methane, then it is quite possibly also sensitive to the long-lived oxidation intermediates of slowly reacting VOC species, which are sufficiently long-lived to be transported away from anthropogenic emission centres into remote regions of the troposphere. Chemical mechanisms currently employed in global scale chemical transport models are not detailed enough to represent this potential sensitivity.

There is also a need for better diagnostic information from models in order to understand the production of ozone and attribute it to emission sources. One technique which has been commonly used is to perturb the emissions from certain sources and examine the response in ozone in particular receptor regions. Such perturbation approaches can provide valuable information about model sensitivities, but they potentially ignore nonlinearities in atmospheric chemistry, and they provide very little mechanistic detail. Other approaches involve tagging different NOx emission sources or VOC emission sources to directly obtain information about ozone chemical production. The atmosphere contains some regions which are more sensitive to NOx and some which are more sensitive to VOC. Hybrid tagging approaches, in which NOx and VOC emissions are both tagged have not been attempted.

This project will develop such hybrid tagging approaches and employ them in a global model of atmospheric chemistry and transport (CAM-CHEM) as well as a regional model (WRF-CHEM). In both cases the techniques will be employed with a variety of different chemical mechanisms in order to investigate the sensitivity of tropospheric ozone production to the choice of chemical mechanism. At the global scale, the use of these tagging techniques will enable attribution of global and hemispheric tropospheric background ozone to STE, methane, and regional sources of NOx and VOC. At the regional scale the use of these tagging techniques will enable identification of cases in which relatively long-lived oxidation intermediates of VOCs are transported between polluted regions, thus contributing to ozone formation in addition to local emissions.