Background ozone over the United States in Summer: Origin, trend, and contribution to pollution episodes

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Abstract

Observations indicate that ozone (O_3) concentrations in surface air over the United States (U.S.) in summer contain a 20-45 ppbv background contribution, presumably reflecting transport from outside the North American boundary layer. We use a three-dimensional global model of tropospheric chemistry driven by assimilated meteorological observations to investigate the origin of this background, and to quantify its contribution to total surface O_3 on both average and highly polluted summer days. The model simulation is evaluated with a suite of surface and aircraft observations over the U.S. from the summer of 1995. The model reproduces the principal features in the observed distributions of O_3 and its precursors, including frequency distributions of O_3 concentrations and the development of regional high-O_3 episodes in the eastern U.S. Comparison of simulations with 1995 vs. 1980 global fossil fuel emissions indicates that the model captures the previously observed decrease in the high end of the O_3 probability distribution in surface air over the U.S. (reflecting reduction of domestic hydrocarbon emissions) and the increase in the low end and the median (reflecting, at least in the model, rising Asian emissions). In the model, background O_3 produced outside of the North American boundary layer contributes an average 25-35 ppbv to afternoon O_3 concentrations in surface air in the western U.S. and 15-30 ppbv in the eastern U.S. during the summer of 1995. This background generally decays to below 15 ppbv during the stagnation conditions conducive to exceedances of the 8-hour 0.08 ppmv (80 ppbv) National Ambient Air Quality Standard (NAAQS) for O_3. A high background contribution of 25-40 ppbv is found during 9% of these exceedances, reflecting convective mixing of free tropospheric O_3 from aloft, followed by rapid production within the U.S. boundary layer. Anthropogenic emissions in Asia and Europe are found to increase afternoon O_3 concentrations in surface air over the U.S. by typically 4-7 ppbv, both under average and highly polluted conditions. This enhancement is particularly large (up to 14 ppbv) for O_3 concentrations in the 50-70 ppbv range, and would represent a major concern if the NAAQS were to be tightened.

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