Supplementary Materials for

A Call for an Aloft Air Quality Monitoring Network: Need and Feasibility

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Community Multiscale Air Quality (CMAQ) Model

CMAQ is a comprehensive air pollution modeling system that simulates air quality on scales ranging from urban to hemispheric. The model includes detailed representation of processes (emissions, atmospheric chemistry, transport, removal) regulating the fate of airborne pollutants. Information on the dynamical state of the atmosphere is typically derived from simulations of the Weather Research and Forecast (WRF) meteorological model. The model code, documentation, and extensive peer-reviewed literature on model processes, algorithms, applications, and evaluation are publicly available at: https://www.epa.gov/cmaq. The modeling system is widely used for air pollution research and regulatory applications across the world.

CMAQ-Adjoint

A comparative analysis of the influence of ozone concentrations at the surface and aloft on $DM8O_3$ is conducted using an adjoint version of the CMAQ model (*23*). An adjoint model solves an auxiliary set of sensitivity equations that mimic and are linked to the model's governing equations. Unlike the base CMAQ model, the adjoint equations are integrated backward in time, and in doing so they trace influences on a scalar concentration function (often referred to as the adjoint cost or objective function) back to concentrations and sources at preceding model time steps. Backward integration of adjoint equations follows a forward simulation of the CMAQ model during which the required concentrations and model variables at all time steps are saved for use in the adjoint simulation. The advantage of the adjoint sensitivities calculated in this

manner is in their ability to provide time- and location-specific measures of influence on the objective function.

To quantify how surface or aloft concentrations of ozone affect surface $DM8O_3$, the adjoint objective function is defined as the cumulative DM8O3 over a region – depicted by the inner box within the computational domain for these calculations (depicted in Figure S1), for days on which the $DM8O_3$ value exceeds a 70 ppb threshold. A 20-cell margin from the boundary (the inner box in Figure S1) is considered as the target area for this analysis to avoid influences from the lateral boundaries and to better characterize in-domain influences. The calculation of adjoint sensitivities are initially forced at the times and locations where the objective function materializes, in this case hours and cells that comprise MD8O3 exceedances over the target area. The adjoint "forcing" at each location and time then corresponds to the instantaneous sensitivity of the objective function to ozone concentration at that location and time. This "forcing" can be considered analogous to emissions for the forward model, since it acts as the "source" of influences that drive the adjoint simulations. These influences are then integrated backward in time through the adjoint system of equations to preceding times and for various modeled processes (transport, chemistry, deposition, etc) at each time step.

With this definition of the adjoint objective function, the values at each location in Figure 4 should be interpreted as the total DM8O3 contributions in ppb (for MD8O3 > 70 ppb) in the target area for any ppb ozone at that location. As such, these values can be taken as a measure of information that ozone observations at various altitudes and locations may carry with regards to characterization of surface-level ozone. Note that calculated adjoint contributions to surface MD8O3 concentrations extend beyond the inner box in Figure S1, as concentrations in these upwind locations influence surface concentrations over the target area. Similar adjoint analysis performed to estimate the added value of potential aloft NO_x measurements (Figure S2) also suggest significantly larger contributions from aloft NO_x levels to regulating $DM8O_3$, than surface values. These results suggest that aloft NOx, either transported from elevated sources, or recycled from reservoir species is also influential in regulating surface $DM8O_3$ especially in regions away from major NOx emissions. The lower $DM8O_3$ sensitivity to surface NO_x

concentrations arises form O_3 destruction from reactions with NO in vicinity of the NO_x emission sources.

CMAQ-Adjoint was used to simulate air quality and the backward sensitivities described above, over a domain covering the southeastern U.S. (depicted in Figures 4 and S1) discretized with a horizontal grid of 12km resolution, for the July 1-14, 2011 period. The adjoint version of CMAQ used here includes all gas-phase chemistry, transport, removal processes relevant for representing tropospheric O_3. The adjoint is based on CMAQv4.5 (https://www.epa.gov/cmaq/cmaq-models-0). Gas-phase chemistry is represented using the Carbon Bond 5 (CB05) chemical mechanism which includes 187 chemical reactions amongst 72 gaseous species (5). The vertical extent from the surface to 50hPa was discretized with 35 layers of variable thickness. Meteorological fields were derived from simulations with the Weather Research and Forecast (WRF) model, while anthropogenic emissions of various species were based on the 2011 National Emissions Inventory (NEI). 3-D chemical initial fields and boundary conditions were derived from previously conducted CMAQ simulations over a larger continental domain which have been extensively evaluated against a variety of surface and aloft measurements (5). This CMAQ-adjoint model configuration, domain size and discretization, and simulation duration enable an appropriate attribution of the backward sensitivities of surface-level $DM8O_3$ to O_3 and precursor levels at various altitudes

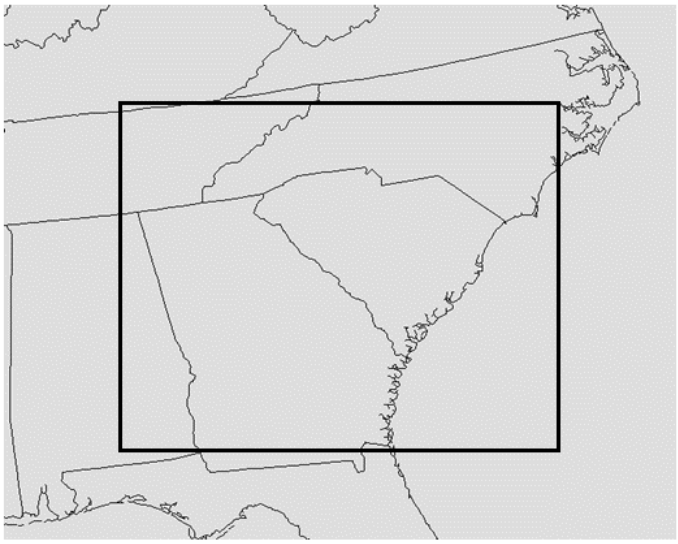

Figure S1: The computational domain for the CMAQ-adjoint simulations. The inner box is the target area over which the adjoint objective function is defined.

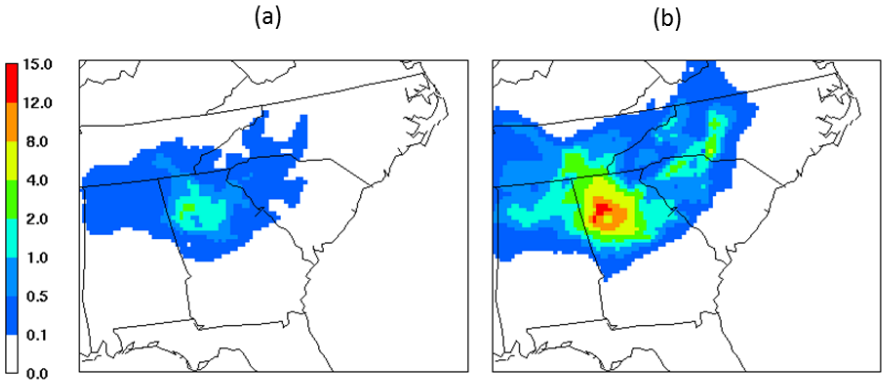

Figure S2: Adjoint sensitivity of surface-level daily maximum 8-hour average O_3 >70ppb ($DM8O_3$) to NO mixing ratios at different altitudes across the southeastern U.S. during 1-14 July, 2011. Estimated change in simulated $DM8O_3$ (in ppb) due to (a) a 1 ppb change in representation of NO at the surface (0-20m) and (b) a 1 ppb change in representation of NO at 400-500m altitude. Improvements in characterization of NO_x mixing ratios aloft would also yield improved predictive capabilities for $DM8O_3$. NO_x aloft, either transported from elevated sources or recycled from reservoir species (such as organic nitrates) is also influential in regulating surface $DM8O_3$ in regions away from major surface-level NO_x sources. Thus as measurement accuracy of compact, low-cost NO_x sensor is improved, the proposed aloft network could be enhanced to also provide aloft NO_x measurements to improve surface O_3 predictions.