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Using air quality modeling to study source–receptor relationships between nitrogen oxides emissions and ozone exposures over the United States

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ABSTRACT

Human exposure to ambient ozone (O_3) has been linked to a variety of adverse health effects. The ozone level at a location is contributed by local production, regional transport, and background ozone. This study combines detailed emission inventory, air quality modeling, and census data to investigate the source–receptor relationships between nitrogen oxides (NO_x) emissions and population exposure to ambient O_3 in 48 states over the continental United States. By removing NO_x emissions from each state one at a time, we calculate the change in O_3 exposures by examining the difference between the base and the sensitivity simulations. Based on the 49 simulations, we construct state-level and census region-level source–receptor matrices describing the relationships among these states/regions. We find that, for 43 receptor states, cumulative NO_x emissions from upwind states contribute more to O_3 exposures than the state's own emissions. In-state emissions are responsible for less than 15% of O_3 exposures in 90% of U.S. states. A state's NO_x emissions can influence 2 to 40 downwind states by at least a 0.1 ppbv change in population-averaged O_3 exposure. The results suggest that the U.S. generally needs a regional strategy to effectively reduce O_3 exposures. But the current regional emission control program in the U.S. is a cap-and-trade program that assumes the marginal damage of every ton of NO_x is equal. In this study, the average O_3 exposures caused by one ton of NO_x emissions ranges from −2.0 to 2.3 ppm-people-hours depending on the state. The actual damage caused by one ton of NO_x emissions varies considerably over space.

1. Introduction

Human exposure to ambient ozone (O_3) has been linked to a variety of adverse health effects including exacerbation of acute and chronic respiratory symptoms, increased hospital admissions, and premature mortality (Dockery et al., 1993; Schwartz, 1996; Bell et al., 2005; Ito et al., 2005; Levy et al., 2005; Jerret et al., 2009). According to the United States Environmental Protection Agency (USEPA), over 140 million Americans are currently living in areas exceeding the health-based National Ambient Air Quality Standard (NAAQS) for O_3 (US EPA 2008a). In order to effectively alleviate O_3 damages on human health, it is important to identify the major sources that contribute to O_3 exposures among residents in a receptor area.

There are two major sources that contribute to ambient O_3 above background levels: local O_3 production and long-range transport of O_3 and its precursors (atmospheric constituents that produce O_3 under proper conditions). In the troposphere, O_3 is formed from reactions of nitrogen oxides ($NO_x = NO + NO_2$) and volatile organic compounds (VOCs) in the presence of sunlight. O_3 in the planet boundary layer (PBL) over the United States in summer has a lifetime of 2–3 days in the east and 3–5 days in the west (Fiore et al., 2002). This allows the O_3 produced at one location to be transported hundreds of kilometers downwind, easily crossing borders of states. In many U.S. states, compliance with the O_3 standards is complicated by transboundary transport of O_3 and its precursors from upwind states (Federal Register, 2005).

The importance of regional transport of O_3 and its precursors has been increasingly recognized by scientific and regulatory communities (Farrell and Keating, 2002; Federal Register, 2005; Tong and Mauzerall, 2008). Historically, O_3 pollution was considered a local problem, and the Clean Air Act (CAA) required state environmental agencies to be responsible for attaining the O_3 standards through controlling precursor emissions within the state boundaries. Since then, new research has revealed that large quantities of O_3 are transported across state boundaries, and that smog is a regional problem (Cleveland et al., 1976; Wolff et al., 1977; National Research Council (NRC), 1991, 2004; Southern Oxidant Study (SOS) Report

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(1995)). The air quality analysis conducted for the Ozone Transport Assessment Group (OTAG) and Clean Air Interstate Rule (CAIR) confirmed that emissions from upwind sources had an important impact on O_3 levels downwind (Farrell, 2001; Farrell and Keating, 2002; Federal Register, 2005). Bergin et al. (2007) used a chemical transport model to examine the impact of statewide emissions on local and downwind O_3 concentrations in the eastern United States. They estimated that on average, 77% of each state's O_3 concentrations are caused by out-of-state emissions. Recently, Tong and Mauzerall (2008) established the source–receptor relationships between state-level NO_x emissions and ambient O_3 concentrations among all 48 contiguous U.S. states, and found that in 38 states, interstate transport contributes more than local emissions to summertime peak O_3 concentrations. None of these studies, however, has investigated the effect of interstate transport on population exposures to O_3.

This study extends the literature in several ways. This study is the first to examine the source–receptor relationship between NO_x emissions and O_3 exposures among states and among census regions over the entire continental United States. Different from the area-weighted source–receptor matrices presented in Tong and Mauzerall (2008), the exposure matrices consider the collocation of O_3 concentration and population distribution. Second, we derive a similar source–receptor relationship among census regions. Such source–receptor matrices (SRM) are valuable in determining the geographical range needed for coordinated regional control of emissions to protect public health. Third, we compare the relative contributions of in-state NO_x emissions to the overall O_3 exposures for each state, to explore how much control each state may have on its O_3 exposures by reducing just its own NO_x emissions. With the recent remanding of the Clean Air Interstate Rule, this research provides timely information to develop future multi-state pollution regulations.

Compared to other data sources such as survey, ambient and personal monitors, air quality modeling data has been less frequently used to estimate air pollutant exposures in epidemiological studies. It has been shown that the use of air quality modeling to estimate O_3 exposure can alleviate several important limitations faced by existing monitor-based approaches (Bell, 2006; Tong et al., 2006, 2007). The monitor-based approaches that use spatial interpolation techniques to estimate O_3 at locations without monitor data are highly uncertain for areas at large distances from monitors, particularly for air pollutants such as O_3 whose ambient concentrations vary considerably within a short distance (Rao et al., 1997). Other advantages of using air quality modeling data to estimate O_3 exposures include: 1) the ability to cover a large region with or without O_3 monitors; 2) high spatial and temporal resolutions making it possible to better capture peak O_3 and duration; 3) the ability to capture the spatial heterogeneity in O_3 distribution through detailed information of emission sources and topography, and through full implementation of chemical and physical processes (Tong et al., 2007). In addition, air quality models can allow us to attribute O_3 exposures to certain source categories or source regions. This work demonstrates how to use air quality modeling to link source apportionment to exposure assessment.

2. Methods

2.1. Estimating population exposure to outdoor O_3

We estimate O_3 exposures by combining O_3 concentrations predicted by air quality modeling with geographically distributed population. The county-level population data for 1996 come from the 2000 U.S. Census of Population (US Census Bureau; http://www.census.gov). We present in this study the aggregate O_3 exposures for all age groups. Alternatively, we could have calculated exposures to each age/gender/race group in order to integrate this analysis with epidemiological studies that are based on specific sub-groups. However, the state level exposure results are not very different by age group. Rather than presenting a myriad of similar exposure results by sub-group, we present here the single aggregate estimate.

The county-based population data is assigned to model grid boxes, so that both O_3 concentrations and population distribution can be processed in a consistent way. Although the U.S. NO_x emissions also cause O_3 changes outside the U.S., only the population exposures within the U.S. boundaries are considered in this analysis. The effect of emissions from outside the U.S. is included in the boundary conditions. We estimate two outdoor O_3 exposure metrics: the population-averaged O_3 concentrations (in parts per billion by volume, ppbv) and the cumulative O_3 exposures (in ppbv–people–h). The cumulative exposure for a state is calculated using the following formula:

$$\text{Cumulative Exposure} = \sum_{i=1}^{M} \sum_{j=1}^{N} (C_{i,j} \times P_i) \quad (1)$$

where M is the number of grid cells within a state, N is the number of hours considered, C is ozone concentration (ppbv), i and j are the index for grid cell and hour, respectively. P_i is the population within grid cell i. When a grid cell extends into more than one state, only the fraction of the cell located within the receptor state is accounted for using an area-weighting approach. The population-averaged O_3 concentrations, or mean exposure, is thus calculated from normalizing the value of cumulative exposure by the state population and then by the numbers of hours and grid cells.

2.2. Predicting O_3 concentrations

We use an atmospheric chemistry and transport model, the Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006), to simulate tropospheric O_3 and related gases. Ambient O_3 concentrations are simulated by a number of physical and chemical processes that include chemical production from its precursors (NO_x, carbon monoxide, hydrocarbons, etc.), transport by wind and turbulence, and removal by deposition and chemical transformations. These processes are implemented in CMAQ with horizontal and vertical advection based on the Piecewise Parabolic Method (PPM), turbulent diffusion based on K-theory, chemistry in the gas, liquid, and particulate phases using a modified version of the CBM-IV chemical mechanism, dry deposition, and RADM cloud physics and chemistry. The model configuration is the same as was evaluated in Tong and Mauzerall (2006).

The key inputs to the CMAQ model include meteorology, emissions of O_3 precursors, initial concentrations, and boundary conditions. Hourly meteorological parameters, such as temperature, wind speed and direction, humidity, pressure, and solar radiation, are obtained from the 5th Generation Mesoscale Model (MM5) (Grell et al., 1994). Anthropogenic emissions of nitrogen oxides (NO_x), sulfur dioxide (SO_2), carbon monoxide (CO), VOCs, and ammonia (NH_3) are processed from the county level U.S. EPA 1996 National Emissions Inventory by the Sparse Matrix Operator Kernel Emissions (SMOKE) model (Houyoux et al., 2000). Vehicle emissions of NO_x, VOCs, CO and primary particulate matter (PM) are prepared using the MOBILE5 model (EPA, 2003). Biogenic emissions, including NO_x, isoprene and monoterpenes, are obtained from the biogenic emissions inventory system, version 3 (BEIS3) (Pierce et al., 1998). Initial concentrations and boundary conditions are extracted from a multi-year simulation using the global chemistry transport model, MOZART-2 (Horowitz et al., 2003).

The model domain includes all 48 contiguous U.S. states and parts of Southern Canada and Northern Mexico (Fig. 1). The time period for the CMAQ simulations is from July 1st to July 31st 1996. The model results of the first two days are not used to minimize the effect of initial concentrations. The domain is divided into 132 columns by 90