Environment International 35 (2009) 1109–1117

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

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

Daniel Q. Tong [a,b,*], Nicholas Z. Muller [c], Haidong Kan [d], Robert O. Mendelsohn [e]

[a] Science and Technology Corporation, Research Triangle Park, NC 27711, USA
[b] Department of Marine, Earth & Atmospheric Sciences, North Carolina State University, Raleigh, NC 27695, USA
[c] Economics Department, Middlebury College, Middlebury, VT, 05753, USA
[d] School of Public Health, Fudan University, Shanghai, 200032, China
[e] School of Forestry & Environmental Studies, Yale University, New Haven, CT, 06511, USA

ARTICLE INFO

Article history:
Received 7 April 2009
Accepted 10 June 2009
Available online 4 August 2009

Keywords:
Ozone exposure
Exposure modeling
Source–receptor
Transboundary transport
CMAQ
Cap and trade

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

* Corresponding author. US EPA MD E243-03, RTP, NC 27711, USA. Tel.: +1 919 541 5150; fax: +1 919 541 1379.
E-mail address: tong.daniel@epa.gov (D.Q. Tong).

0160-4120/$ – see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.envint.2009.06.008

(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

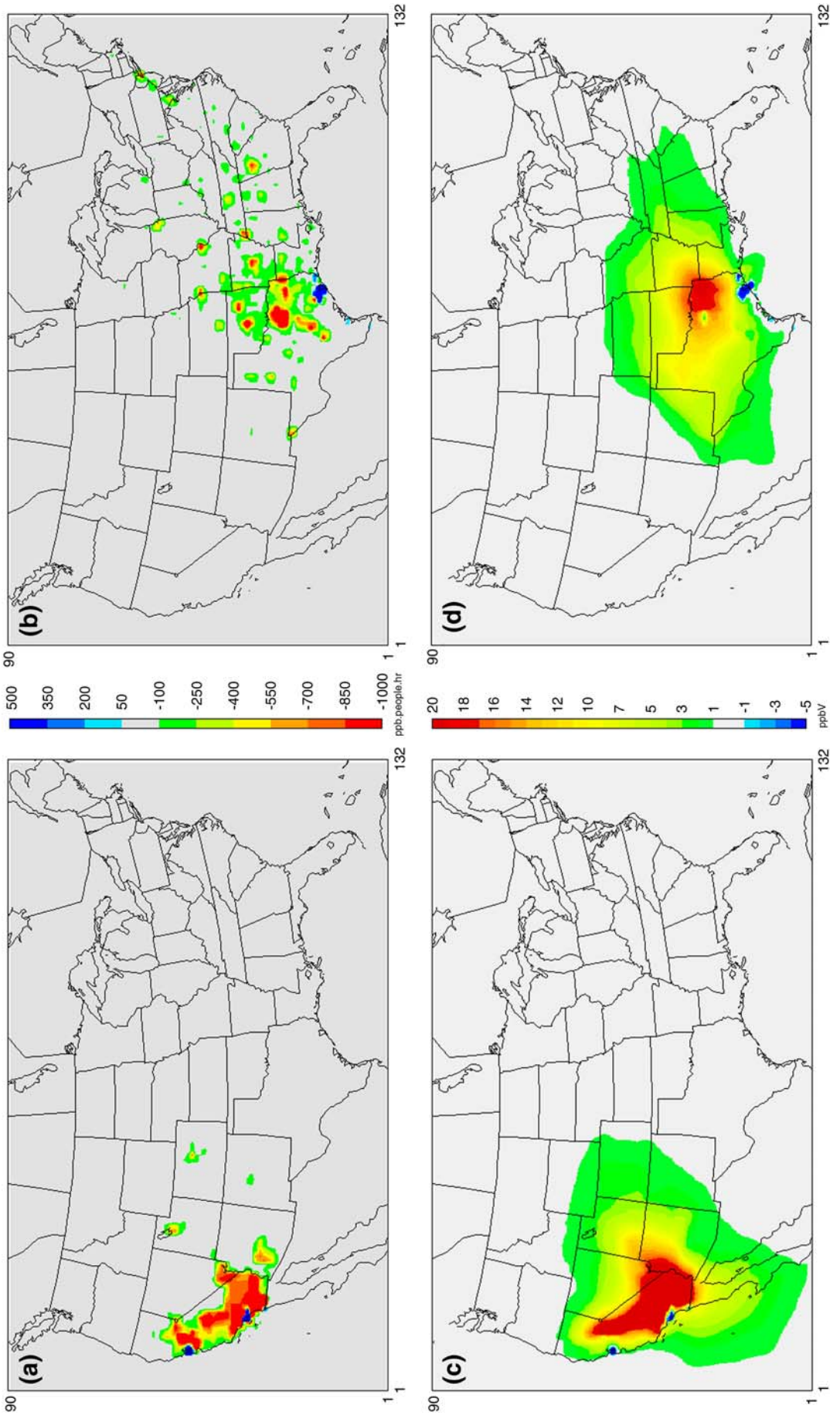


Fig. 1. Effects of NO_x emissions from California (a & c) and Texas (b & d) on daily average O_3 concentrations (c & d) and population exposure to O_3 (a & b) in July 1996.

Table 1
State-level source–receptor relationships between NO_x emissions from source states (SRC) and changes in population-weighted O_3 concentrations (ppbv) in receptor states (RCP) over the continental United States in July 1996. Each row represents O_3 changes in all receptor states resulting from NO_x emissions from the source state; each column represents O_3 concentration changes in a receptor state due to NO_x emissions from all source states.

SRC/RCP	AL	AR	AZ	CA	CO	CT	DE	FL	GA	IA	ID	IL	IN	KS	KY	LA	MA	MD	ME	MI	MN	MO	MS	MT
AL	6.7	0.0	0.0	0.0	0.0	0.1	0.2	0.3	4.8	0.0	0.0	0.0	0.1	0.0	0.6	0.2	0.1	0.3	0.0	0.0	0.0	0.0	1.6	0.0
AR	0.8	2.7	0.0	0.0	0.0	0.1	0.1	0.1	0.6	0.1	0.0	0.4	0.5	0.4	1.0	0.6	0.1	0.2	0.0	0.1	0.0	1.4	1.5	0.0
AZ	0.0	0.1	7.9	0.2	0.5	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
CA	0.1	0.1	5.1	10.9	1.6	0.0	0.0	0.0	0.1	0.1	0.6	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.2
CO	0.1	0.2	0.1	0.0	5.5	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.4	0.1	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.2	0.0
CT	0.0	0.0	0.0	0.0	0.0	-2.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.9	0.0	0.4	0.0	0.0	0.0	0.0	0.0
DE	0.0	0.0	0.0	0.0	0.0	0.3	-1.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.2	0.1	0.0	0.0	0.0	0.0	0.0
FL	1.9	0.1	0.0	0.0	0.0	0.2	0.1	4.7	2.2	0.0	0.0	0.0	0.0	0.0	0.1	0.6	0.2	0.1	0.1	0.0	0.0	0.0	0.8	0.0
GA	1.6	0.0	0.0	0.0	0.0	0.2	0.3	0.4	4.2	0.0	0.0	0.0	0.0	0.0	0.2	0.0	0.2	0.4	0.1	0.0	0.0	0.0	0.1	0.0
IA	0.2	0.6	0.0	0.0	0.0	0.1	0.1	0.0	0.1	2.7	0.0	1.4	1.0	0.9	0.5	0.1	0.1	0.2	0.1	0.5	1.2	1.7	0.3	0.0
ID	0.0	0.1	0.0	0.0	0.4	0.0	0.0	0.0	0.0	0.0	2.8	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.6
IL	0.9	0.9	0.0	0.0	0.0	0.4	0.3	0.0	0.5	1.8	0.0	-1.9	4.4	0.3	3.4	0.2	0.3	0.6	0.2	2.0	0.7	1.0	1.0	0.0
IN	0.8	0.3	0.0	0.0	0.0	0.4	0.4	0.0	0.7	0.4	0.0	-2.0	2.1	0.0	2.2	0.0	0.3	0.8	0.2	1.8	0.1	0.6	0.5	0.0
KS	0.2	0.7	0.0	0.0	0.3	0.1	0.1	0.0	0.1	0.7	0.0	0.3	0.3	1.6	0.3	0.1	0.0	0.1	0.0	0.1	0.4	0.8	0.2	0.0
KY	0.9	0.4	0.0	0.0	0.0	0.4	0.5	0.0	0.9	0.0	0.0	0.4	1.7	0.0	3.8	0.1	0.2	1.0	0.2	0.3	0.0	0.4	0.7	0.0
LA	3.6	2.8	0.0	0.0	0.0	0.2	0.2	0.3	1.9	0.0	0.0	0.2	0.4	0.1	0.5	5.2	0.1	0.3	0.1	0.0	0.0	0.6	7.4	0.0
MA	0.0	0.0	0.0	0.0	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	-4.0	0.0	1.4	0.0	0.0	0.0	0.0	0.0
MD	0.1	0.0	0.0	0.0	0.0	1.2	1.8	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.8	-1.3	0.4	0.0	0.0	0.0	0.0	0.0
ME	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.5	0.0	0.0	0.0	0.0	0.0
MI	0.2	0.0	0.0	0.0	0.0	0.7	0.6	0.0	0.2	0.4	0.0	0.7	1.2	0.0	0.8	0.0	0.7	1.1	0.5	1.1	0.4	0.2	0.1	0.0
MN	0.1	0.3	0.0	0.0	0.0	0.2	0.1	0.0	0.1	2.7	0.0	1.2	0.8	0.5	0.3	0.0	0.2	0.2	0.2	0.9	1.1	0.8	0.1	0.0
MO	0.6	2.3	0.0	0.0	0.0	0.1	0.1	0.1	0.3	1.5	0.0	1.3	1.3	0.5	1.7	0.3	0.1	0.3	0.1	0.3	0.4	2.0	1.0	0.0
MS	2.7	0.5	0.0	0.0	0.0	0.1	0.1	0.2	1.4	0.0	0.0	0.1	0.2	0.0	0.7	1.5	0.1	0.2	0.0	0.0	0.0	0.1	4.6	0.0
MT	0.1	0.1	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.1	0.3	0.1	0.1	0.1	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	2.3
NC	0.3	0.0	0.0	0.0	0.0	0.5	0.8	0.0	0.6	0.0	0.0	0.0	0.0	0.0	0.3	0.0	0.4	1.0	0.2	0.0	0.0	0.0	0.0	0.0
ND	0.0	0.1	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.7	0.0	0.3	0.2	0.3	0.1	0.0	0.0	0.1	0.0	0.2	0.6	0.3	0.0	0.1
NE	0.1	0.3	0.0	0.0	0.4	0.0	0.0	0.0	0.1	0.7	0.0	0.2	0.2	1.2	0.2	0.0	0.0	0.1	0.0	0.1	0.3	0.7	0.1	0.0
NH	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	-0.6	0.0	0.8	0.0	0.0	0.0	0.0	0.0
NJ	0.0	0.0	0.0	0.0	0.0	1.4	-0.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.7	0.5	0.9	0.0	0.0	0.0	0.0	0.0
NM	0.1	0.1	0.5	0.0	0.3	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0
NV	0.0	0.0	0.2	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
NY	0.0	0.0	0.0	0.0	0.0	2.5	0.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.7	0.4	1.6	0.0	0.0	0.0	0.0	0.0
OH	0.2	0.0	0.0	0.0	0.0	1.1	1.0	0.0	0.4	0.0	0.0	0.1	0.7	0.0	1.4	0.0	0.7	2.3	0.4	0.9	0.0	0.0	0.0	0.0
OK	0.5	2.0	0.0	0.0	0.1	0.1	0.1	0.0	0.3	0.4	0.0	0.2	0.2	2.9	0.5	0.1	0.0	0.1	0.0	0.1	0.2	1.5	0.6	0.0
OR	0.0	0.1	0.0	0.6	0.4	0.0	0.0	0.0	0.0	0.0	1.2	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.7
PA	0.0	0.0	0.0	0.0	0.0	2.8	1.7	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.8	3.5	0.9	0.0	0.0	0.0	0.0	0.0
RI	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	-0.4	0.0	0.3	0.0	0.0	0.0	0.0	0.0
SC	0.2	0.0	0.0	0.0	0.0	0.2	0.2	0.1	0.6	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.2	0.1	0.1	0.0	0.0	0.0	0.0	0.0
SD	0.0	0.1	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.7	0.0	0.2	0.2	0.3	0.1	0.0	0.0	0.0	0.0	0.1	0.3	0.3	0.0	0.0
TN	1.7	-0.4	0.0	0.0	0.0	0.2	0.3	0.1	1.4	0.0	0.0	0.2	0.7	0.0	3.3	0.4	0.2	0.7	0.1	0.0	0.0	0.4	1.6	0.0
TX	1.9	8.6	0.4	0.0	0.2	0.2	0.2	0.2	1.3	0.5	0.0	0.6	0.5	3.2	1.1	3.2	0.1	0.4	0.1	0.2	0.2	2.7	3.6	0.0
UT	0.0	0.1	0.3	0.0	0.8	0.0	0.0	0.0	0.0	0.0	0.2	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
VA	0.1	0.0	0.0	0.0	0.0	1.1	2.7	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.3	0.0	0.7	2.3	0.4	0.1	0.0	0.0	0.0	0.0
VT	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.0	0.3	0.0	0.0	0.0	0.0	0.0
WA	0.0	0.1	0.0	0.7	0.3	0.0	0.0	0.0	0.0	0.1	2.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	1.7
WI	0.1	0.2	0.0	0.0	0.0	0.3	0.1	0.0	0.1	1.1	0.0	1.7	1.3	0.2	0.3	0.0	0.3	0.2	0.2	2.3	1.3	0.5	0.1	0.0
WV	0.0	0.0	0.0	0.0	0.0	0.3	0.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.2	1.0	0.1	0.0	0.0	0.0	0.0	0.0
WY	0.1	0.2	0.0	0.0	1.4	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.2	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1

rows with a horizontal grid resolution of 36×36 km^2. One grid cell is comparable in size to a northeastern U.S. county, but is smaller than a county in other parts of the country (for instance, Los Angeles County in California encompasses 21 grid cells).

2.3. Quantifying source–receptor relationships for O_3 exposures

To quantify the effect one state's NO_x emissions have on O_3 exposure in all states, we conduct 49 model simulations that include one base case and 48 emission perturbation runs. In the base case all natural and anthropogenic emissions are turned on. In each perturbation run we remove the NO_x emissions from one state (the source state) and then examine resultant changes in surface O_3 concentrations in all states (receptor states). The difference in O_3 concentrations between the perturbation and the baseline run is considered as the contribution by the source state from which NO_x emissions are removed. The change in surface O_3 concentrations in the source state is considered as the in-state effect, while the change in other states as out-of-state effects. Next, we combine O_3 changes with population data to estimate the change in O_3 exposures caused by each state's NO_x emissions as given above. Finally, we construct state level and census region level source–receptor matrices describing the relationships among these state/regions. Based on the derived source–receptor matrices, we estimate 1) the relative importance of in-state and out-of-state emissions to ambient O_3 concentrations in each state, and 2) average O_3 exposures per ton of NO_x emitted from each state.

3. Results

3.1. Spatial distribution of O_3 exposures caused by one state's NO_x emissions

We first demonstrate how one state's emissions can affect O_3 exposures inside and outside the source state. Fig. 1 shows the spatial distribution of changes in cumulative O_3 exposures in July 1996 caused by NO_x emissions from Texas and California, the top two NO_x emitters in the United States. The exposure changes display large spatial variability in the distribution of exposure changes caused by a source state's NO_x emissions. The changes caused by California NO_x emissions are mostly confined within the state boundaries. A comparable level of O_3 exposure changes is predicted only in five urban areas outside California. In contrast, O_3 exposures resulting from Texas emissions are distributed across several downwind states. Texas NO_x emissions were found to cause an increase in O_3 exposures in many cities in the eastern U.S., reaching as far as Chicago and

SRC/RCP	NC	ND	NE	NH	NJ	NM	NV	NY	OH	OK	OR	PA	RI	SC	SD	TN	TX	UT	VA	VT	WA	WI	WV	WY
AL	1.6	0.0	0.0	0.1	0.2	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.1	2.5	0.0	2.2	0.0	0.0	0.6	0.1	0.0	0.0	0.4	0.0
AR	0.5	0.0	0.2	0.1	0.1	0.0	0.0	0.1	0.3	0.7	0.0	0.2	0.0	0.5	0.0	1.3	0.2	0.0	0.3	0.0	0.0	0.1	0.4	0.0
AZ	0.0	0.0	0.1	0.0	0.0	3.3	1.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.6	0.0	0.0	0.0	0.0	0.0	0.2
CA	0.1	0.1	0.2	0.0	0.0	1.5	11.1	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.2	0.1	0.1	3.5	0.1	0.0	0.0	0.0	0.1	1.3
CO	0.1	0.1	0.4	0.0	0.0	0.6	0.0	0.0	0.0	0.3	0.0	0.0	0.0	0.1	0.2	0.1	0.1	0.0	0.1	0.0	0.0	0.0	0.1	1.0
CT	0.0	0.0	0.0	0.9	-0.1	0.0	0.0	-0.7	0.0	0.0	0.0	0.0	1.4	0.0	0.0	0.0	0.0	0.0	0.0	0.4	0.0	0.0	0.0	0.0
DE	0.0	0.0	0.0	0.2	0.4	0.0	0.0	0.2	0.0	0.0	0.0	-0.6	0.2	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
FL	0.7	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.2	1.8	0.0	0.2	0.1	0.0	0.2	0.0	0.0	0.0	0.1	0.0
GA	3.1	0.0	0.0	0.2	0.3	0.0	0.0	0.1	0.0	0.0	0.0	0.2	0.1	5.9	0.0	1.1	0.0	0.0	0.7	0.1	0.0	0.0	0.2	0.0
IA	0.1	0.4	1.1	0.1	0.1	0.0	0.0	0.1	0.6	0.3	0.0	0.2	0.1	0.1	0.8	0.3	0.0	0.0	0.1	0.1	0.0	1.1	0.3	0.0
ID	0.0	0.1	0.1	0.0	0.0	0.2	0.2	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.1	0.0	0.0	2.1	0.0	0.0	0.1	0.0	0.0	1.1
IL	0.5	0.1	0.4	0.4	0.4	0.0	0.0	0.4	2.2	0.1	0.0	0.9	0.2	0.4	0.2	1.7	0.0	0.0	0.7	0.4	0.0	1.5	1.7	0.0
IN	0.9	0.0	0.0	0.4	0.5	0.0	0.0	0.5	3.2	0.0	0.0	1.1	0.2	0.5	0.0	1.5	0.0	0.0	1.0	0.5	0.0	0.3	2.2	0.0
KS	0.1	0.3	1.7	0.0	0.1	0.1	0.0	0.1	0.2	1.2	0.0	0.1	0.0	0.1	0.7	0.3	0.1	0.0	0.1	0.0	0.0	0.2	0.2	0.0
KY	1.6	0.0	0.0	0.3	0.5	0.0	0.0	0.4	2.7	0.0	0.0	1.3	0.2	0.8	0.0	2.5	0.0	0.0	1.6	0.3	0.0	0.0	4.3	0.0
LA	0.7	0.0	0.0	0.1	0.2	0.0	0.0	0.1	0.3	0.2	0.0	0.2	0.1	1.1	0.0	1.4	0.6	0.0	0.4	0.1	0.0	0.0	0.3	0.0
MA	0.0	0.0	0.0	-0.7	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	-1.2	0.0	0.0	0.0	0.0	0.0	0.0	0.8	0.0	0.0	0.0	0.0
MD	0.4	0.0	0.0	0.7	2.4	0.0	0.0	0.7	0.1	0.0	0.0	1.6	0.8	0.1	0.0	0.0	0.0	0.0	-0.2	0.3	0.0	0.0	0.4	0.0
ME	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0
MI	0.5	0.1	0.1	0.9	0.6	0.0	0.0	0.9	2.2	0.0	0.0	1.3	0.4	0.3	0.1	0.4	0.0	0.0	0.8	1.0	0.0	1.0	1.4	0.0
MN	0.1	1.4	0.9	0.3	0.2	0.0	0.0	0.2	0.5	0.1	0.0	0.3	0.1	0.1	1.2	0.2	0.0	0.0	0.1	0.3	0.0	2.0	0.2	0.0
MO	0.3	0.1	1.1	0.1	0.2	0.0	0.0	0.1	0.8	0.9	0.0	0.4	0.1	0.3	0.4	1.3	0.1	0.0	0.3	0.1	0.0	0.5	0.8	0.0
MS	0.6	0.0	0.0	0.0	0.1	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.0	0.8	0.0	1.4	0.1	0.0	0.3	0.0	0.0	0.0	0.4	0.0
MT	0.1	0.3	0.3	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.5	0.1	0.0	0.1	0.0	0.0	0.0	0.1	0.1	1.4
NC	5.5	0.0	0.0	0.2	0.6	0.0	0.0	0.2	0.1	0.0	0.0	0.2	0.5	1.4	0.0	0.4	0.0	0.0	2.7	0.1	0.0	0.0	0.3	0.0
ND	0.0	1.7	0.8	0.0	0.1	0.0	0.0	0.1	0.1	0.1	0.0	0.1	0.0	0.0	1.4	0.1	0.0	0.0	0.0	0.1	0.0	0.3	0.1	0.1
NE	0.1	0.6	1.6	0.0	0.0	0.0	0.0	0.0	0.2	0.3	0.0	0.1	0.0	0.1	1.2	0.1	0.0	0.0	0.1	0.0	0.0	0.1	0.1	0.3
NH	0.0	0.0	0.0	-1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.3	0.0	0.0	0.0	0.0
NJ	0.1	0.0	0.0	1.6	-8.2	0.0	0.0	-3.4	0.0	0.0	0.0	-1.6	1.8	0.1	0.0	0.0	0.0	0.0	0.3	0.9	0.0	0.0	0.1	0.0
NM	0.0	0.0	0.2	0.0	0.0	4.1	0.1	0.0	0.0	0.2	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0
NV	0.0	0.0	0.0	0.0	0.0	0.3	1.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.7	0.0	0.0	0.0	0.0	0.0	0.2
NY	0.0	0.0	0.0	3.0	-7.0	0.0	0.0	-3.5	0.0	0.0	0.0	0.6	2.7	0.0	0.0	0.0	0.0	0.0	0.1	3.5	0.0	0.0	0.1	0.0
OH	1.3	0.0	0.0	0.9	1.5	0.0	0.0	1.2	1.6	0.0	0.0	3.7	0.5	0.7	0.0	0.5	0.0	0.0	2.2	0.8	0.0	0.0	5.9	0.0
OK	0.2	0.1	0.7	0.0	0.1	0.1	0.0	0.0	0.1	5.5	0.0	0.1	0.0	0.3	0.3	0.5	0.4	0.0	0.2	0.0	0.0	0.2	0.2	0.0
OR	0.0	0.1	0.1	0.0	0.0	0.2	0.4	0.0	0.0	0.1	3.0	0.0	0.0	0.0	0.1	0.0	0.0	0.5	0.0	0.0	0.1	0.0	0.0	0.6
PA	0.4	0.0	0.0	1.7	2.3	0.0	0.0	2.1	0.2	0.0	0.0	0.7	1.6	0.1	0.0	0.0	0.0	0.0	1.7	1.5	0.0	0.0	0.6	0.0
RI	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	-1.6	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0
SC	2.8	0.0	0.0	0.1	0.2	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.2	3.4	0.0	0.3	0.0	0.0	0.5	0.0	0.0	0.0	0.0	0.0
SD	0.0	0.7	1.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.1	0.0	0.0	1.5	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.2
TN	2.3	0.0	0.0	0.1	0.4	0.0	0.0	0.2	0.5	0.1	0.0	0.4	0.1	1.6	0.0	4.4	0.0	0.0	1.5	0.1	0.0	0.0	1.6	0.0
TX	0.7	0.2	0.8	0.1	0.2	2.5	0.1	0.1	0.3	8.5	0.0	0.3	0.1	1.1	0.2	2.2	4.4	0.0	0.5	0.1	0.0	0.2	0.7	0.0
UT	0.0	0.1	0.1	0.0	0.0	0.7	0.1	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.1	0.0	0.0	4.4	0.0	0.0	0.0	0.0	0.0	0.9
VA	1.9	0.0	0.0	0.5	1.9	0.0	0.0	0.6	0.3	0.0	0.0	1.3	0.9	0.4	0.0	0.2	0.0	0.0	2.8	0.3	0.0	0.0	1.5	0.0
VT	0.0	0.0	0.0	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.4	0.0	0.0	0.0	0.0
WA	0.0	0.1	0.1	0.0	0.0	0.1	0.2	0.0	0.0	0.1	4.9	0.0	0.0	0.0	0.2	0.1	0.0	0.5	0.0	0.0	5.2	0.0	0.1	0.8
WI	0.1	0.4	0.3	0.4	0.2	0.0	0.0	0.3	0.7	0.0	0.0	0.4	0.2	0.0	0.4	0.1	0.0	0.0	0.2	0.4	0.0	2.6	0.3	0.0
WV	0.4	0.0	0.0	0.1	0.4	0.0	0.0	0.2	0.1	0.0	0.0	0.6	0.1	0.1	0.0	0.1	0.0	0.0	1.0	0.1	0.0	0.0	1.2	0.0
WY	0.1	0.2	0.3	0.0	0.0	0.1	0.0	0.0	0.0	0.2	0.0	0.0	0.0	0.1	0.4	0.1	0.0	0.1	0.1	0.0	0.0	0.0	0.1	2.7

New York City. These cities are also frequently impacted by emissions from other southeastern states (not shown here). The summertime circulation over the eastern United States is typically dominated by the Bermuda high pressure system (Eder et al., 1993; Fiore et al., 2002; Tong et al. 2006). Such meteorological conditions allow persistent transport of O_3 produced in the southeast to northeastern states, a phenomenon sometimes referred to as "an ozone river" (Wolff et al., 1980).

3.2. State-level source–receptor relationships for O_3 exposures

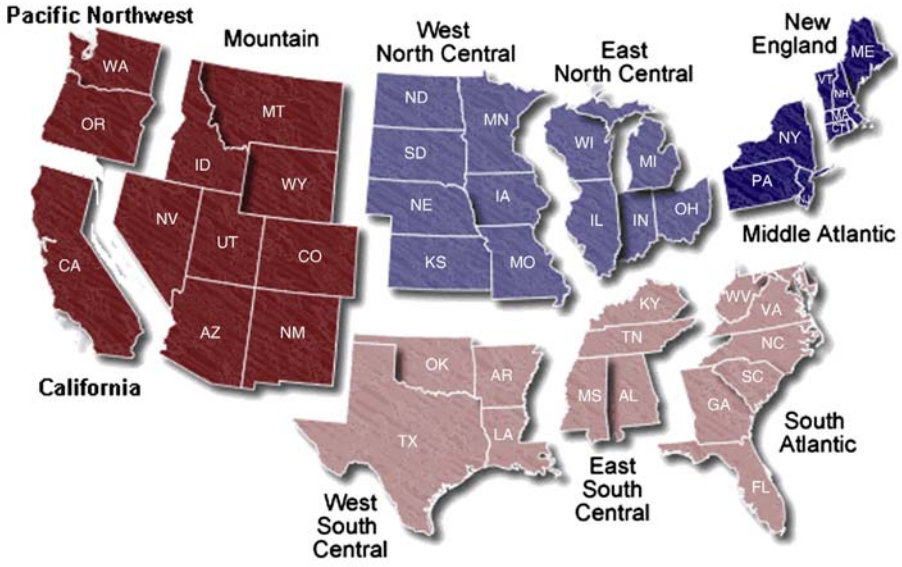
Next we derive the state-level source–receptor relationships between NO_x emissions and O_3 exposures based on the 48 sensitivity simulations. Table 1 shows a source–receptor matrix (SRM) that summarizes the effect that one state's NO_x emissions have on O_3 exposures in all states. As the size of populations varies widely among states, we present here the population-averaged O_3 concentrations so that the level of exposure changes among individual cohorts is comparable between different states. Each row of the source–receptor matrix represents how much a *source* state can affect population-weighted O_3 changes in the 48 states. The number of states with at least a 0.1 ppbv change in O_3 concentration can range from 2 to 40. Texas, the largest NO_x emitter, affects the largest number (40) of receptor states. California, the second largest emitter, affects only 14 states. The majority (75%) of the continental states affect 10–30 downwind states. None of the New England states, which are located downstream of the major O_3 pathway in the east, affect more than eight states.

Each column of the matrix indicates how much a *receptor* state can be affected by itself and other states. A receptor state can be affected by up to 28 source states. In general, eastern states are more subject to the influence of interstate transport than western states, due to prevailing meteorology and the smaller sizes of eastern states. States located in upwind regions or near national borders are less affected by inter-state transport. For instance, Washington and Oregon are affected only by three nearby states. For 22 receptor states, there is at least one upwind state that contributes more to O_3 exposures in the receptor state than the state itself.

The descending diagonal of the matrix represents changes in O_3 exposures caused by in-state (intrastate) NO_x emissions. In-state NO_x emissions increase O_3 exposures in 39 states, with the largest in-state effect in California. In nine states, however, in-state emissions decrease in-state O_3 exposures. Seven of these states are in the Northeast (CT, DE, MA, NH, NJ, NY, and RI), where large upwind NO_x emissions suppress in-state O_3 production from in-state NO_x emissions. The two other states (IL and MD) are dominated by major cities in which O_3 titration by NO_x dominates effects in the rest of the state. For these nine states, reducing in-state NO_x emissions does not reduce in-state all-hour O_3 exposures but may actually increase it.

3.3. Source–receptor relationship among census regions

Next we assemble a similar matrix describing the source–receptor relationship among census regions. Fig. 2 presents the effect of each region's NO_x emissions on the O_3 exposures in itself and other census regions. Note the Pacific West region is now



Receptor / Source	California	Pacific Northwest	Mountain	West South Central	West North Central	East South Central	East North Central	South Atlantic	Middle Atlantic	New England
California	247	0	45	2	1	1	1	2	1	0
Pacific Northwest	30	39	31	1	2	1	2	1	1	0
Mountain	6	1	94	11	9	6	6	9	3	1
West South Central	1	0	6	168	41	74	29	60	14	3
West North Central	0	0	3	20	83	25	108	22	22	5
East South Central	0	0	0	10	4	115	44	136	32	5
East North Central	0	0	0	4	27	48	178	89	122	22
South Atlantic	0	0	0	3	0	26	6	268	91	24
Middle Atlantic	0	0	0	0	0	1	2	32	-137	59
New England	0	0	0	0	0	0	0	0	-8	-17

Fig. 2. Source–receptor relationships between NO_x emissions and O_3 exposures among census regions. The geographical scope of each census region is at the top, and the table (bottom) represents O_3 exposure changes (unit: $10^6 \times$ ppm–people–hours) in each census region in July 1996.

divided into California and Pacific Northwest. Each region is responsible for less than half of its O_3 exposures, except in California and the South Atlantic where own emissions contribute over a half. With all the other regions, they cause substantial O_3 exposure damages to neighboring regions. This suggests that Census regions are too small to successfully regulate O_3 exposures because too much of the damages either are caused by or fall outside the region. The problem for the two northeast regions is particularly acute. These two regions are at the mercy of up to six upwind regions who contribute noticeable amounts of their O_3 exposures.

The near zero values of the upper right and lower left corners of the SRM indicate that not all regions impact every other region. Regional definitions that are larger than Census regions may work. For example, California could probably be one region. The Pacific Northwest and Mountain regions could be another region. The entire region east of the Rocky Mountains could be another region.

Note the above discussion of O_3 concentrations and exposures focuses only on the amount of O_3 explained by NO_x emissions from U.S. sources. The O_3 concentrations are higher than the sum of NO_x contributions by all 48 states. Other sources contributing to O_3 concentrations include contributions by other O_3 precursors (VOCs, methane, and carbon monoxide), international transport of O_3 from outside of the U.S., and downward transport of O_3 from the stratosphere where O_3 is produced by a different mechanism (Chapman, 1930). O_3 from all of the above sources interacts with each other. Therefore, their actual contributions to the overall O_3 concentrations can be different from the results derived from individual experiments, in which a single contributing source is turned on and off. Our results derived from completely eliminating NO_x emission should be interpreted as the projected effect on O_3 concentrations or exposures from controlling domestic NO_x emissions.

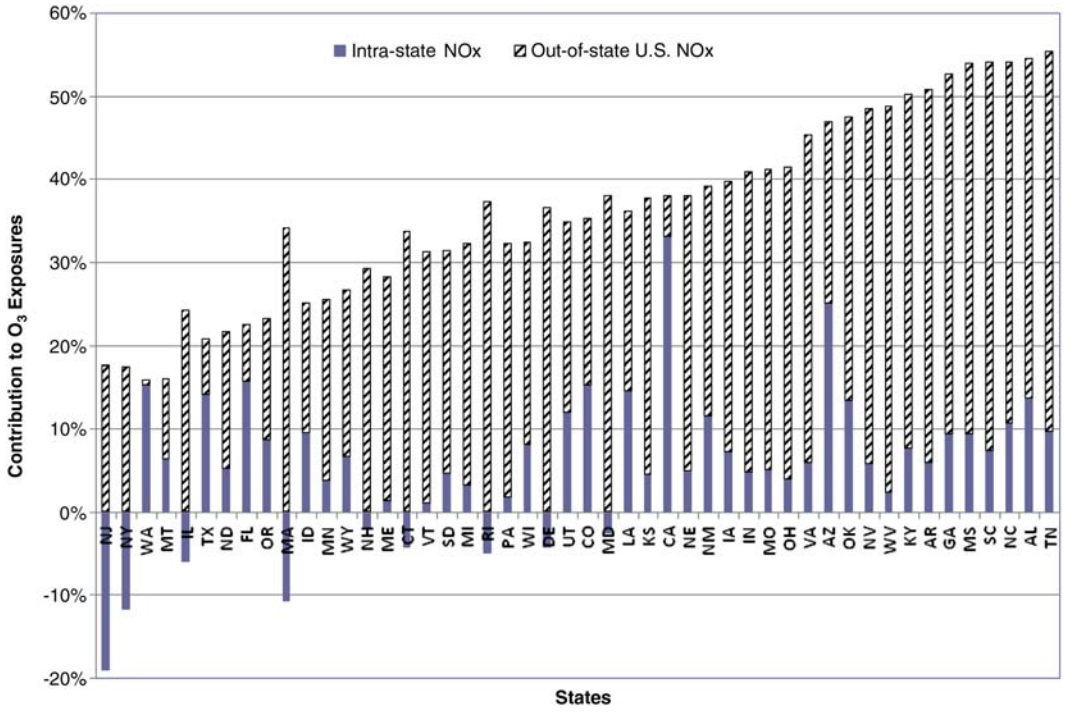


Fig. 3. Percentage of own state and out-of-state contributions to NO_x caused O_3 exposures in each U.S. state.

3.4. Local contribution to O_3 exposure

Fig. 3 shows the percentage of in-state and out-of-state domestic NO_x contributions to O_3 exposures in each contiguous U.S. state. The sum of domestic NO_x emissions account for −1% to 55% of O_3 exposures. The wide ranges of NO_x contributions to O_3 exposures indicate the large variability in O_3 mitigation one state can gain through reducing U.S. NO_x emissions. For eight southeastern states (KY, AR, GA, MS, SC, NC, AL and TN), eliminating domestic NO_x emissions can result in a decrease in O_3 exposures by over 50% among their residents. For the sixteen states listed on the left of Fig. 3, domestic NO_x emissions are responsible for less than 30% of O_3 exposures.

Among the 48 states, in-state emissions contribute −19% to +33% of O_3 exposures. The in-state contributions are negative in the nine states because, as discussed earlier, we chose to use daily 24 h average concentration as the O_3 metric for exposure assessment. For 88% of the states, the in-state contribution accounts for less than 15% of exposures. On average, in-state NO_x emissions, out-of-state NO_x emissions, and other sources contribute 8%, 26%, and 66% of total O_3 exposures in the 48 states, respectively.

3.5. Average O_3 exposure per ton of NO_x emissions

Previous studies revealed that there is large spatial variability in the O_3 damages caused by NO_x emissions (Mauzerall et al., 2005; Tong et al., 2006). Such a finding has important policy implications because the current NO_x emission trading programs assumes the location of NO_x emissions does not matter. These earlier experiments, however, were conducted for either a single source sector (Mauzerall et al., 2005) or sources within and surrounding the Atlanta, GA metropolitan area (Tong et al., 2006). It remains unclear if such spatial variability of NO_x effects exists for emissions from one whole state, where NO_x are emitted from multiple emission sources that are distributed over a large space. Here we calculate the average O_3 exposures per ton of NO_x emitted from each state (Fig. 4). Although our results are limited to O_3 exposure, the results are also indicative of exposure related health effects because health effects are proportional to O_3 exposures.

Average O_3 exposures caused by one ton of NO_x emissions range from −2.0 to 2.3 ppm–people–hours (unit). Although varying widely from state to state, the average exposures are relatively similar for NO_x emissions from nearby states. For the northeastern states, 1 ton of NO_x produces less than 0.5 unit of O_3 exposures. NO_x emitted from these states has the smallest O_3 effects, and thus should not be traded with states outside the region. The same amount of NO_x emitted from mountain states and two southeastern states produces 0.5 to 1.0 unit of O_3 exposures. The NO_x emissions from most eastern and the western coastal states are more damaging, generating over 1.0 unit of O_3 exposures. Therefore, it is possible to group the 48 states into three broad groups, if emission trading programs are to be expanded to cover more states or more source sectors on the basis of their actual damages.

3.6. Discussion of limitations and uncertainties

Several important limitations and uncertainties need to be recognized when using air quality modeling to estimate population exposures to ambient O_3. Unlikely direct measurements, air quality model data has to be evaluated against measurements prior to being used to estimate O_3 exposures. O_3 concentrations predicted by CMAQ were compared with measurements at 987 Air Quality System (AQS) (the former AIRS) and 123 Clean Air Status and Trends Network (CASTNet) (Tong and Mauzerall, 2006). The model was found to be able to reproduce surface O_3 for a wide range of conditions (30–80 ppbv) with a normalized mean bias (the average of differences between model estimates and observations normalized by observations) within ±15% across the U.S. domain. In addition, CMAQ can reproduce 8-hour daily maxima with a cross-domain mean bias (averaged difference between prediction and observations) of 8 ppbv (Tong and Mauzerall, 2006). Comparisons of O_3 vertical profiles using ozonesonde data show the model under-predicts observations in the upper troposphere (Tong and Mauzerall, 2006). The discrepancy is due to the use of a zero-flux upper boundary condition in CMAQ and missing sources of NO_x emissions, such as transported NO_x from the stratosphere, lightning and aircraft discharges. As photochemistry largely controls O_3 concentrations in the polluted boundary layer, the underestimation of free tropospheric O_3 is expected to have only a small effect on our results. As O_3 exposure weighs concentrations in urban area more than in less populated rural areas, it is important for the model to predict O_3 concentration accurately in urban areas. Comparisons of model predictions and measurements at urban AQS sites show that the model underestimates observations by a MB of −5 ppbv and a NMB of −5% (Tong and Mauzerall, 2006). Such biases suggest the model has a tendency to under-predict O_3 concentrations in urban high-population areas in the baseline simulation.

This study adopted a zero-out modeling approach that quantifies the one state's effect by removing 100% of its NO_x emissions. Due to the nonlinear O_3 response to NO_x emissions, our results can not be proportionally interpolated to the effect of a smaller reduction. The source–receptor relationships presented here represent the mean state-level impact of each state's NO_x emissions on monthly mean O_3 concentrations. Actual changes at a particular location within a receptor state or at a specific time vary and will frequently be different from the state-level monthly average (Tong and Mauzerall, 2008). Therefore, our results can not be used to quantify the effect of one state on nonattainment in areas that exceeded the O_3 standards. Another source of uncertainties lies in the model grid size, which has a horizontal grid spacing of 36 km. Although such resolution is considered high for model simulations that cover the entire continental United States, the artificial dilution of power-plant and urban plumes in an Eularian air quality model could result in higher average but lower peak O_3 concentrations (Jang et al., 1995). Recently, Cohan et al. (2006) examined the effect of grid resolution on the O_3 response to NO_x emissions and found that simulations using 36-, 12- and 4-km grid resolution all yielded similar predictions of average O_3 sensitivity to NO_x emissions. Nevertheless, a future study using higher resolution data or other modeling approaches

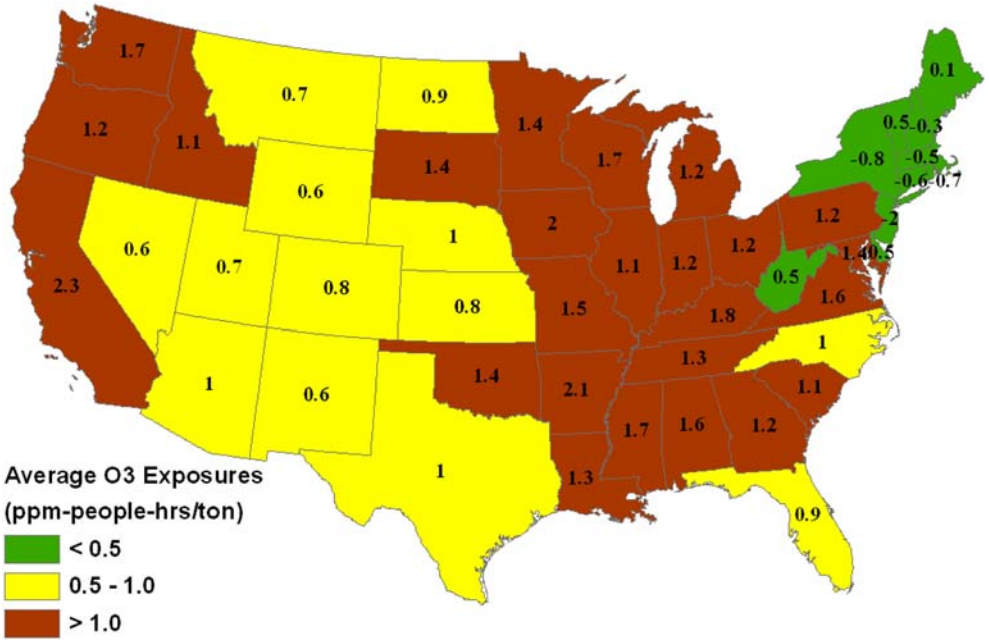


Fig. 4. Average O_3 exposures caused by one ton of NO_x emissions from each state.

to capture the sub-gird variability will be useful to better quantify human exposure to O_3 under different emission schemes.

The O_3 concentrations are calculated using the USEPA 1996 national emission inventory, which does not reflect the reduction resulting from many emission regulations implemented since then (Farrell and Keating, 2002; Federal Register, 2005; USEPA, 2008b). The USEPA estimated that anthropogenic NO_x emissions have been reduced by 25% from 1996 to 2004 (USEPA, 2009). To estimate the effect of changed emissions on O_3 exposures, we have run the model with updated emissions taken from the 2002 national emission inventories and updated emissions from electricity generation units based on the 2004 Continuous Emission Measurements (CEMs) data. We focus here on the exposure changes in the eastern United States, where population is dense and the emission reduction substantial. Fig. 5 depicts the changes in population exposures to 24-hour average (left) and daily maximum 8-hour (right) O_3 in July from 1996 to 2004. The population-average exposure levels have decreased by 6%–30% in the Southeast and Northeast, but have either remained unchanged or increased by 6%–18% in the Upper Midwest. The changes are caused by variability in emissions and meteorology, and the O_3 production efficiency of unit NO_x emissions under different chemical and meteorological conditions. Therefore, a similar analysis using the updated 2009 emission inventories would provide a more up to date description of inter-state transport of O_3 pollution over the United States.

4. Summary and discussion

Regardless of numerous environmental regulations that aimed at reducing high O_3 concentrations, more than half of the U.S. population still live in areas exceeding the health-based National Ambient Air Quality Standard (NAAQS) for O_3 (USEPA, 2008a,b). The present study investigates the source–receptor relationships between NO_x emissions and O_3 exposures among the 48 contiguous states and ten census regions in the United States. The results show that a source state's NO_x emissions can influence 2 to 40 downwind states by at least a change in population-averaged O_3 exposure of 0.1 ppbv. A 0.1 ppvb exposure has been associated with a 0.0085% increase in total mortality (derived from Bell et al., 2005). In general, health effects are proportional to human exposures.

Further, each state can be affected by up to 28 source states by at least 0.1 ppbv. The large number of interacting states suggests that the impact of one state's NO_x emissions can be more far reaching than the

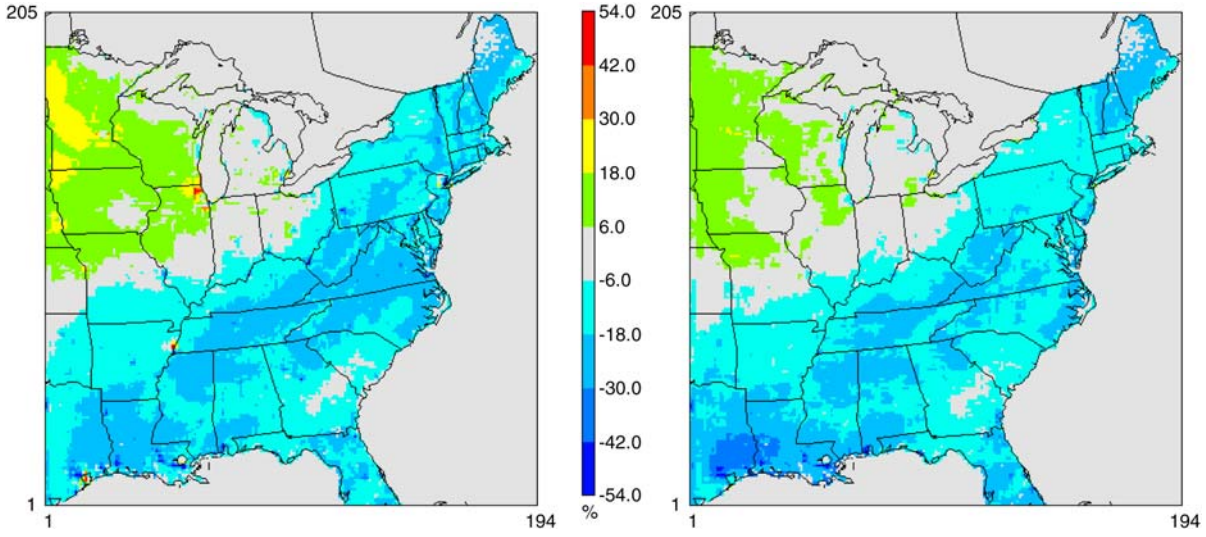


Fig. 5. Changes in population exposures to 24-hour average (left) and daily maximum 8-hour (right) O_3 in July from 1996 to 2004.

source–receptor matrices for O_3 concentrations showed (Tong and Mauzerall, 2008) and that the exposures in each state are controlled by multiple upwind states.

The importance of inter-state transport on O_3 exposure varies from state to state. For California and Florida, their O_3 exposures are only moderately impacted by neighboring upwind states, whose combined effect is less than 15% of that from local NO_x emissions. These states can reap the majority of health benefits by controlling their own emissions. State-specific source benefit analyses provide strong policy incentives for these states to seek a state level solution to reduce O_3 pollution. For most other states, they either affect O_3 exposures in downwind states and will undervalue benefits, or are influenced considerably by sources in upwind states. For 22 states there is at least one single upwind state whose NO_x emissions can contribute more to O_3 exposures in the receptor state than the state itself. For 43 states, the cumulative emissions of NO_x by upwind states have a bigger impact than the states own emissions on O_3 exposures. On average, in-state emissions of NO_x are responsible for only 8% of in-state concentrations of O_3. In general, the US needs a regional strategy to control O_3 exposures.

Meanwhile, our results also show that the geographical range of transboundary transport is limited for O_3 pollution caused by NO_x. The Pacific Coastal and Mountain regions affect O_3 exposures in each other's region. These regions generally do not interact with other regions in the middle and eastern U.S. The middle U.S. regions affect themselves and the eastern regions, but not the western regions. As a receptor they are impacted by themselves, but not by the western or eastern regions. The eastern U.S. regions are affected by the middle and eastern U.S., and they affect exposures only in themselves. Such source–receptor relationships suggest that transboundary transport of O_3 pollution is a process of regional rather than national scale. The NO_x-caused O_3 problem is best resolved through a regional NO_x program.

Many current NO_x emission trading programs are developed based on the premise that the location of NO_x emissions does not matter. We found the average O_3 exposures caused by one ton of NO_x emissions range from −2.0 to 2.3 ppm–people–hours over different states. Our results confirm the finding of previous studies that the actual damages caused by NO_x vary largely over space. These results are troubling for a trading program that assumes the marginal damage of NO_x is the same at each source. Emission trading from places where NO_x reduces exposures to places where NO_x increases exposures are likely to make things worse. The spatial variability exists not only for emissions from a single location or emission sector, but also for emissions from an entire state. To correct this problem, our predicted average exposure per ton of emission at each source can be used as a first approximation to calculate damage indices for emission trading across states (see Muller and Mendelsohn, 2009). This would tend to discourage emissions from moving from low damage to high damage locations.

Acknowledgements

Funding for this work was provided by the Glaser Progress Foundation. We thank the Offices of Information Technology at North Carolina State University for the computational resources to conduct the model simulations. The authors are grateful to professors Denise Mauzerall and S.T. Rao for their comments on earlier versions of this manuscript.

References

Bell ML. The use of ambient air quality modeling to estimate individual and population exposure for human health research: a case study of ozone in the Northern Georgia region of the United States. Environ Int 2006;32(5):586–93.
Bell ML, Dominici F, Samet JM. A meta-analysis of time-series studies of ozone and mortality with comparison to the national morbidity, mortality, and air pollution study. Epidemiology 2005;16(4):436–45.
Byun DW, Schere KL. Review of the governing equations, computational algorithms, and other components of the models-3 community multiscale air quality (CMAQ) Modeling System. Appl Mech Rev 2006;59(2):51–77.
Chapman S. On ozone and atomic oxygen in the upper atmosphere. Philos Mag 1930; S7:369–83.
Cleveland J, Kleiner B, McRae JE, Warner JL. Photochemical air pollution: transport from the New York City area into Connecticut and Massachusetts. Science 1976;191:179–81.
Cohan DS, Hu Y, Russell AG. Dependence of ozone sensitivity analysis on grid resolution. Atmos Environ 2006;40(1):126–35.
Dockery DW, et al. An association between air pollution and mortality in six U.S. cities. N Engl J Med 1993;329(24):1753–9.
Eder BK, Davis JM, Bloomfield P. A characterization of the spatiotemporal variability of non-urban ozone concentrations over the eastern United States. Atmos Environ 1993;27A:2645–68.
Farrell A. Multi-lateral emissions trading: lessons from inter-state NO_x trading in the United States. Energy Policy 2001;29(13):1061–72.
Farrell A, Keating T. Transboundary environmental assessment: ozone regionality in the United States. Environ Sci Technol 2002;36(12):2537–44.
Federal Register, 2005. Rule To Reduce Interstate Transport of Fine Particulate Matter and Ozone (Clean Air Interstate Rule); Revisions to Acid Rain Program; Revisions to the NOX SIP Call. p RIN 2060-AL76, May 2005.
Fiore AM, Jacob DJ, Bey I, Yantosca RM, Field BD, Fusco AC. Background ozone over the United States in summer: origin, trend, and contribution to pollution episodes. J Geophys Res 2002;107(D15) 10.1029.
Grell, G.A., Dudhia, J., Stauffer, D.R., 1994. A description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5). NCAR Technical Note.
Horowitz LW, et al. A global simulation of tropospheric ozone and related tracers: description and evaluation of MOZART, version 2. J Geophys Res 2003;108(D24):4784.
Houyoux MR, Vukovich JM, Coats Jr CJ, Wheeler NM, Kasibhatla PS. Emission inventory development and processing for the Seasonal Model for Regional Air Quality (SMRAQ) project. J Geophys Res 2000;105(D7):9079–90.
Ito K, De Leon SF, Lippman M. Associations between ozone and daily mortality: analysis and meta-analysis. Epidemiology 2005;16:446.
Jang JC, Jeffries HE, Tonnesen GS. Sensitivity of ozone to model grid resolution II: detailed process analysis for ozone chemistry. Atmos. Environ. 1995;29:3101–14.
Jerret M, Burnett RT, Pope III CA, Ito K, Thurston G, Krewski D, Shi Y, Calle E, Thun M. Long-term ozone exposure and mortality. N Engl J Med 2009;360:1085–95.
Levy JI, Chemerynski SM, Sarnat JA. Ozone exposure and mortality: an empiric Bayes metaregression analysis. Epidemiology 2005;16:458.
Mauzerall DL, Sultan B, Kim N, Bradford D. NO_x emissions: variability in ozone production, resulting health damages and economic costs. Atmos Environ 2005;39:2851–66.
Muller, N, and R. Mendelsohn. 2009.: Efficient Pollution Regulation: Getting the Prices Right. Amer. Econ. Rev. (forthcoming).
National Research Council (NRC). Rethinking the ozone problem in urban and regional air pollution. committee on tropospheric ozone formation and measurement. Washington, DC: National Academy Press; 1991.
National Research Council (NRC). Air quality management in the United States. Washington D.C.: National Research Council of the National Academies; 2004.
Pierce T, Geron C, Bender L, Dennis R, Tonnesen G, Guenther A. Influence of increased isoprene emissions on regional ozone modeling. J. Geophys. Res. 1998;103 (D19):25611–29.
Rao ST, Zurbenko IG, Neagu R, Porter PS, Ku JY, Henry RF. Space and time scales in ambient ozone data. Bull. Am. Meteorol. Soc. 1997;78:2153–66.
Schwartz J. Air pollution and hospital admissions for respiratory disease. Epidemiology 1996;7(1):1–9.
Southern Oxidant Study (SOS) Report. State of the Southern Oxidants Study -1 Policy-Relevant Findings In Ozone Pollution Research 1988 – 1994. Raleigh, North Carolina, 27695: North Carolina State University; 1995.
Tong DQ, Mauzerall DL. Spatial variability of summertime tropospheric ozone over the continental United States: implications of an evaluation of the CMAQ model. Atmos Environ 2006;40:3041–56.
Tong DQ, Mauzerall DL. Summertime state-level source–receptor relationships between nitrogen oxides emissions and surface ozone concentrations over the continental United States. Environ Sci Technol 2008;42(21):7976–84.
Tong DQ, Muller NZ, Mauzerall DL, Mendelsohn RO. Integrated assessment of the spatial variability of ozone impacts from emissions of nitrogen oxides. Environ Sci Technol 2006;40(5):1395–400.
Tong DQ, Mathur R, Schere K, Kang D, Yu S. The use of air quality forecasts to assess impacts of air pollution on crops: a case study of assessing soybean yield losses from ozone in the United States. Atmos Environ 2007;41:8772–84.
U.S. Environmental Protection Agency (USEPA), 2008a. Green Book — 8-Hour Ozone Nonattainment Areas. Available online http://www.epa.gov/air/oaqps/greenbk/gntc.html (accessed May 2, 2008).
US. Environmental Protection Agency (USEPA), 2008b. National Emissions Inventory (NEI) Air Pollutant Emissions Trends, Office of Air Quality Planning and Standards (OAQPS), Research Triangle Park, NC. http://www.epa.gov/ttn/chief/trends/trends06/nationaltier1upto2006basedon2002finalv2.1.xls (accessed on July 26, 2008).
Wolff GT, Lioy PJ, Wight GD, Meyers RE, Cederwall RT. An investigation of long-range transport of ozone across the midwestern and eastern United States. Atmos Environ 1977;11:797–802.
Wolff GT, Lioy PJ, Wight GD. Development of an ozone river associated with synoptic scale episodes in the eastern Untie States. Environ Sci Technol 1980;14:1257–60.