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Potential effects of climate and emissions changes on surface ozone in the Chicago area

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ABSTRACT

Future changes in climate and precursor emissions will likely have important consequences on ground-level ozone concentrations for the City of Chicago and its surrounding suburban/rural areas. Here we use a regional climate-air quality modeling system to evaluate the combined and individual effects of climate warming (and resulting biogenic emissions increases) and anthropogenic emissions perturbations from 1996-2000 to 2048-2052 and 2095–2099 in this region. Two scenarios are considered, including A1FI (higher warming with increasing anthropogenic emissions) and B1 (less warming with reduced anthropogenic emissions). Relative to 1996-2000, projected changes in climate and anthropogenic emissions together lead to little ozone change for the City of Chicago under A1FI but 5.0-7.8 ppb increases under B1 by 2048-2052 and 2095-2099. For A1FI, the decreasing ratio of volatile organic compounds (VOCs) to nitrogen oxides (NOx) reduces ozone concentrations over Chicago, despite the increasing emissions for both NOx and VOCs. Averaged over the Chicago urban and surrounding suburban area, however, surface ozone increase 2.3-7.1 ppb under A1FI by 2095–2099. Additionally, the seasonal ozone variation is projected to increase 84–127% under A1FI but decrease 23-30% under B1 over the Chicago area. By comparison, projected climate warming alone increases the surface ozone by 2.1-8.7 ppb and its seasonal variation by 22-89% over the Chicago area from 1996-2000 to 2095–2099 under both scenarios. Therefore, effective emission regulation and climate considerations are both important to pollution mitigation in the Chicago area.

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Introduction

Potential climate warming due to anthropogenic emissions of greenhouse gases will likely have important consequences on human health over the City of Chicago and its surrounding suburban/rural areas. Vavrus and Van Dorn (2010) find that heat waves are likely to occur more frequently and intensively and last longer by the end of the century under different emissions scenarios from the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES, Nakicenovic et al., 2000), which likely lead to increased mortality (Hayhoe et al., 2010). Furthermore, climate warming, together with potential changes in emissions of ozone precursors, will likely affect surface ozone concentrations over the Chicago area, adding another dimension of adverse health impacts (e.g., see papers on health effects by Bell et al., 2007; Jacobson, 2008). This is of particular concern because of the dense population and existing high pollution level in this area. Holloway et al. (2008) used a statistical downscaling approach to project future ozone change over Chicago and found that surface ozone concentrations may increase by 6-17 ppb by the end of the century due to climate warming alone (see section on the Comparison with an alternative approach for more details). In this study, we use a regional climate-air quality modeling system to project future ozone changes over the City of Chicago and its surrounding areas, analyzing the individual effects of projected changes in climate and changes in anthropogenic emissions. A similar analysis has been done by Kunkel et al. (2008) for the Northeast US, while other studies (e.g., Hogrefe et al., 2004; Mickley et al., 2004; Leung and Gustafson, 2005; Steiner et al., 2006; Meleux et al., 2007; Tao et al., 2007; Lin et al., 2008; Nolte et al., 2008) have studied the potential effects of climate change on air quality in various parts of the US and in Europe.

Past chemistry modeling studies have suggested that surface ozone levels over the United States are likely to change in future years because of changes in climate and precursor emissions (Hogrefe et al.,

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2004; Murazaki and Hess, 2006; Tao et al., 2007; Kunkel et al., 2008; Lin et al., 2008). Using the Community Multiscale Air Quality (CMAQ) modeling system, Hogrefe et al. (2004) found that summer mean maximum daily 8-hour average (MD8A) surface ozone concentrations over the northern part of the eastern US increased 1.5-10.5 ppb by 2053-2057 and 1.5-16.5 ppb by 2083-2087 relative to 1993-1997 in response to projected climate change (including resulting biogenic emissions perturbations) under the A2 scenario following the IPCC SRES. Using a regional climate-air quality (RCAQ) modeling system, Tao et al. (2007) found that the summertime US ozone concentrations are overall expected to increase from 1998 to 2050 due to projected changes in climate and anthropogenic emissions under the A1FI scenario but to decrease under the B1 scenario. In addition, simulations using global chemical transport models (CTMs) also suggest an increase in surface ozone over the Midwest US due to climate warming under the A1b, A1FI, and B1 scenarios (e.g., Murazaki and Hess, 2006; Lin et al., 2008).

Future changes in climate and ozone precursor emissions, however, contain large uncertainties due to the unknown changes in human activities as well as climate fluctuations (Meehl et al., 2007; Lin et al., 2008). The IPCC reviews the potential climate change over the 21st century based on a variety of assumptions about future pathways of economy, population, and human society (e.g., Meehl et al., 2007). The latest IPCC assessment suggests the global mean surface air temperature may increase by up to 4.0 °C (averaged over multimodel calculations) from 1980-1999 to 2090-2099 under the A1FI scenario (Meehl et al., 2007) with intensive fossil fuel use over the 21st century. However, the global mean temperature increase may be much smaller, an estimated 1.8 °C, under the B1 scenario (Meehl et al., 2007) which has a high level of environmental and social consciousness. Therefore the present study evaluates the potential impacts of these two possible trends in climate on surface ozone concentrations over the 21st century for the Chicago area: (1) the A1FI scenario with high warming and (2) the B1 scenario with less warming. Potential changes in biogenic and anthropogenic emissions of ozone precursors are also considered in this study. Changes in biogenic emissions of hydrocarbons, which contribute to ozone formation, are determined by projected climate change. Projected changes in anthropogenic emissions of ozone precursors are based on the same assumptions about human activities in future years as the assumptions made in the scenarios for greenhouse gas emissions. The same two scenarios were used by Tao et al. (2007) and Lin et al. (2008) to evaluate overall potential ozone changes in the US and by Kunkel et al. (2008) to evaluate potential effects on the Northeast US

The effects of changes in climate and precursor emissions on surface ozone concentrations are distinct among urban, suburban, and rural districts of a metropolitan area because of their differences in the relative abundances of nitrogen oxides (NOx) and volatile organic compounds (VOC) and resulting effects on the daytime production and nighttime destruction of ozone (Sillman and Samson, 1995; Sillman, 1999). During the daytime, surface ozone is mainly produced by photochemical reactions of NOx and VOC in the presence of solar radiation. Depending on the relative abundances of NOx and VOC, the ozone production can be limited by NOx concentrations (i.e., in the NOx-limited regime, where increasing NOx emissions leads to increasing ozone production), VOC concentrations (i.e., in the VOClimited regime, where increasing NOx emissions leads to decreasing ozone production), or both (i.e., in the transition regime, where the ozone production is sensitive to changes in both NOx and VOC) (Sillman, 1999). For urban areas such as the City of Chicago, the high NOx levels typically lead to the VOC-limited regime for ozone production. Over the areas with mixed urban, suburban, and rural districts, in contrast, the ozone production is typically controlled by the availability of both NOx and VOC. During the nighttime, surface ozone is continuously destroyed by dry deposition and NOx titration (Sillman, 1999), where the magnitude of NOx titration is determined by the amount of NOx. Therefore, it is interesting to compare the effects of future climate and precursor emissions on surface ozone production/destruction for the City of Chicago, an urban area, with the effects for its surrounding suburban/rural areas.

This study investigates the potential changes in surface ozone concentrations for the City of Chicago and its surrounding suburban and rural areas from 1996-2000 to 2048-2052 and 2095-2099, analyzing the combined and individual effects of projected changes in climate (including the induced changes in biogenic emissions) and anthropogenic emissions under the A1FI and B1 scenarios. The analysis is focused on summer months (June-August), when the ozone pollution is usually most severe. Both daily mean and maximum daily 8-hour average (MD8A) ozone concentrations are analyzed; the MD8A ozone is the primary target of the National Ambient Air Quality Standard for ozone due to its direct relevance to human health (http://www.epa.gov/air/criteria.html). To facilitate the study, we use a regional climate-air quality (RCAQ) modeling system (Huang et al., 2007; Tao et al., 2007; Kunkel et al., 2008), which incorporates a regional climate model (RCM), an emission model and an air quality model (AQM). The ozone changes over two areas are compared: the City of Chicago (an urban area) and the Chicago area (latitude: 41.202-42.496°N; longitude: 87.524-88.707°W; the mix of urban, suburban, and rural areas, including the City of Chicago). The latter area is selected to represent the boundaries of the City of Chicago and its surrounding largely suburban counties. These two areas have very different levels of NOx, VOC, and thus ozone presently; and their ozone responses to projected changes in anthropogenic emissions are also expected to be very different.

Model description and methodology

The RCAQ system has been described and evaluated in detail by Huang et al. (2007) and Tao et al. (2007). Here we briefly describe the key components incorporated in the system. The RCM is based on the fifth-generation Penn State University/National Center for Atmospheric Research (NCAR) mesoscale modeling system (MM5) version 3 (Dudhia et al., 2000), with the revisions in the buffer zone treatment, ocean interface, and cloud-radiation interactions for regional climate applications (Liang et al., 2001; Liang et al., 2004). In this study, lateral boundary conditions of the RCM are derived from the Parallel Climate Model (PCM). Effects of changes in global climate are included in this analysis by incorporating lateral boundary conditions for future years from the PCM simulations driven by corresponding changes in emissions and concentrations of greenhouse gas and particles. The emission model for the air quality related emissions is the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system (Houyoux et al., 2000) that processes the regional emission inventories for North America. It calculates the gridded and time-dependent emissions of ozone precursors from point, area, mobile, and biogenic sources under the time-varying meteorological conditions derived from the RCM. The AQM is an improved version of the SARMAP Air Quality Model (SAQM; Chang et al., 1997). It improves the scheme for gas-phase chemistry (Huang and Chang, 2001), includes an aerosol module, and modifies the calculation of photolysis rates (Chang et al., 1987). In this study, the AQM is driven by the meteorological data from the RCM and emission data from SMOKE at 1-hour intervals. As in many other regional air quality models (e.g., Hogrefe et al., 2004), the climatological lateral boundary conditions of chemical mixing ratios are used in the AQM (see details in Huang et al., 2007). The RCAQ system has been used in a variety of studies on US ozone air quality and its future changes (e.g., Tao et al., 2003; Huang et al., 2007; Tao et al., 2007; Kunkel et al., 2008). Huang et al. (2007) and Tao et al. (2007) found that RCAQ reproduced the observed summertime US ozone concentrations from the Environmental Protection Agency (EPA) Air Quality System (AQS), including the daily mean (DM) ozone, the maximum daily 8-hour average (MD8A) ozone, and ozone diurnal variations.

Seven 5-year experiments were conducted with the RCAO system over North America at the 30×30 km resolution (see the model domain in Huang et al., 2007); each experiment was conducted for the summer months and the modeled ozone data were output at 1-hour intervals. The lateral boundary conditions of chemical mixing ratios were kept at the present-day levels in all experiments, thus potential changes in long-range chemical transport from outside the computational domain is not considered in the future projection. The first experiment is for the control case, where current emissions and climate during 1996-2000 are used. The subsequent four experiments are for 2048-2052 under A1FI and B1, and 2095-2099 under A1FI and B1, respectively, where the climate, biogenic emissions and anthropogenic emissions all represent the corresponding future years. Changes in biogenic emissions are calculated assuming the land use and the emission factors for each plant type remain unchanged in the future. A one-way interaction is assumed in which future climate affects the biogenic emissions. In other words, changes in isoprene and other biogenic emissions were calculated before the air quality simulation but both emissions and air quality models were driven by the same meteorology from the RCM. The last two experiments are for 2095-2099 under the A1FI and B1 scenarios, respectively, where the climate and climate-induced biogenic emissions represent future years while anthropogenic emissions are kept at the present levels. These 5-year experiments reduce interannual variation in the projected ozone changes. Note that changes in biogenic emissions are always incorporated as part of climate change in the six experiments for future years and in the analyses hereafter if not noted otherwise.

In this study, the present-day anthropogenic emissions were processed by SMOKE based on the inventories from the US EPA 1999 National Emission Inventory (NEI99) for the US, the Big Bend Regional Aerosol and Visibility Observational Study Emissions Inventory for Mexico, and the National Pollutant Release Inventory for Canada. The calculation of future changes in anthropogenic emissions was based on the scaling factors from 2000 to 2050 and 2100 adopted by the IPCC SRES for the US and Canada (available at http://sres.ciesin.org/ final_data.html). For each future period, unlike Woo et al. (2008) who developed emissions growth factors based on region and sector/fuel combinations, each anthropogenic sector uses the same scaling factor over the region (a similar scaling approach is used by Tao et al. (2007)).

Biogenic emissions were calculated using the Biogenic Emissions Inventory System (BEIS) embedded in SMOKE for both current and future periods. The calculation was based on the dependence of biogenic emissions on meteorology, i.e., air temperature and solar radiation; the results varied every hour and from one year to another (Tao et al., 2007; Kunkel et al., 2008). The land use types were kept at the present-day levels in all experiments. Projected changes in total emissions of ozone precursors are summarized in Table 1 and analyzed in the Results section. Impacts of wildfires on precursor emissions are not considered in this study due to the large uncertainties related to occurrence and magnitude of future fires.

Results

Current summertime surface ozone concentrations

Concentrations of surface ozone over polluted areas are largely determined by the daytime production through photochemistry and the nighttime transformation through NOx titration and dry deposition under the influence of mixing processes within the planetary boundary layer (PBL). Table 2 shows the modeled summertime surface ozone concentrations over Chicago and its surrounding areas during 1996–2000. For the City of Chicago, modeled MD8A ozone

Table 1

Present-day June–August emission budgets of ozone precursors and their fractional changes in future years^a.

| Period | NOx | VOC | Isoprene | СО |
|---------------------|-------|-------|----------|-------|
| The Chicago area | | | | |
| 1996-2000 | 5.7 | 6.0 | 0.3 | 33.1 |
| A1FI 2048-2052 | 33% | 4% | 19% | 56% |
| A1FI 2095-2099 | 98% | 65% | 61% | 99% |
| B1 2048-2052 | - 56% | - 36% | 16% | -47% |
| B1 2095-2099 | -81% | - 56% | 18% | -65% |
| The City of Chicago | | | | |
| 1996-2000 | 12.6 | 16.9 | 0.5 | 103.6 |
| A1FI 2048-2052 | 34% | 4% | 19% | 56% |
| A1FI 2095-2099 | 101% | 66% | 61% | 99% |
| B1 2048-2052 | - 58% | - 38% | 17% | -47% |
| B1 2095-2099 | -83% | - 59% | 20% | -65% |

^a The numbers during the period of 1996–2000 denote the present-day budgets of ozone precursor emissions (units: metric tons per summer per km²); and the numbers during 2048–2052 and 2095–2099 denote the fractional changes of emission budgets from 1996–2000 to the corresponding future periods, respectively. The emissions incorporate both anthropogenic and biogenic sources.

mixing ratios are only 39.2 ppb and the DM ozone are only 19.1 ppb during 1996–2000. These relatively low seasonal mean ozone concentrations are because of the excessive NOx levels, which suppress the daytime ozone production (i.e., in the VOC-limited regime) and destroy ozone effectively in the nighttime. Over the surrounding suburban and rural areas, by comparison, NOx concentrations are lower, therefore the ozone production is more efficient, the NOx titration is weaker and the resulting ozone concentrations are higher. Averaged over the Chicago area, the MD8A ozone concentrations are 27.9 ppb during 1996–2000.

Projected changes in climate and precursor emissions

Changes in surface ozone concentrations in future years depend on changes in both climate and the human-related emissions. Averaged over the Chicago area, the surface (2 m above ground) air temperature modeled by the RCM increases by about 1.3 °C under A1FI and 0.8 °C under B1 from 1990–2000 to 2045–2055; the increases are about 3.5 °C under A1FI and 1.0 °C under B1 by 2090–2099. These are in good agreement but somewhat smaller than the temperature changes found in Hayhoe et al. (2010).

Projected future changes in emissions of ozone precursors from anthropogenic and biogenic sources over the Chicago area are presented in Table 1. Overall, ozone precursor emissions significantly increase under A1FI and decrease under B1, especially by 2095–2099. It is noted, however, that biogenic emissions always increase with the warming climate under both scenarios. Under B1, it is the decreasing

| Table | 2 |
|-------|---|
|-------|---|

Present-day June-August surface ozone concentrations and their future changes^a.

| Period | The Chicago area | | The City of Chicago | |
|----------------|------------------|-------------------|---------------------|------------|
| | Daily mean | MD8A ^b | Daily mean | MD8A |
| 1996-2000 | 27.9 (2.0) | 46.6 (3.2) | 19.1 (1.9) | 39.2 (4.1) |
| A1FI 2048-2052 | -0.3 (32%) | 1.5 (37%) | -0.5 (9%) | -0.2 (6%) |
| A1FI 2095-2099 | 2.3 (84%) | 7.1 (127%) | -0.5 (103%) | 0.8 (101%) |
| B1 2048-2052 | 2.9 (1%) | 1.1 (4%) | 6.1 (18%) | 7.5 (8%) |
| B1 2095-2099 | 1.5 (-30%) | -4.5 (-24%) | 7.8 (-24%) | 5.0 (-23%) |

^a The modeled ozone changes are the result of all changes in climate, biogenic emissions, and anthropogenic emissions together. The numbers during the period of 1996–2000 denote the present-day 15-month (three summer months per year for 5 years) means and standard deviations (SDs, in parentheses) of surface ozone concentrations (units: ppb); and the numbers during 2048–2052 and 2095–2099 denote the absolute changes of the 15-month mean ozone (units: ppb) and the fractional changes of the 15-month ozone SDs (in parentheses) from 1996–2000 to the corresponding future periods, respectively.

^b Maximum daily 8-hour average ozone.

anthropogenic sources that lead to the overall reductions in total VOC emissions.

The magnitudes of projected changes in NOx emissions are larger than those of changes in VOC emissions under both scenarios. From 1996–2000 to 2095–2099, NOx emissions double while VOC emissions increase only about 65% under A1FI; under B1, the decreases in NOx emissions are about 80% as compared to the 60% reductions in VOC emissions. Therefore the relative concentrations of VOC to NOx (i.e., VOC/NOx ratio) decrease under A1FI but increase under B1 in future years. Since the magnitude of the VOC/NOx ratio is a proxy of the three ozone production regimes (NOx-limited, VOC-limited and in transition), its changes lead to substantial and distinct changes in ozone production over Chicago (presently in the VOC-limited regime) and its surrounding areas (presently in the transition of NOx- and VOC-limited regimes) among these regimes, as analyzed in the following sections.

Projected changes in surface ozone

Table 2 and Fig. 1 present the modeled changes in summer average MD8A and DM ozone concentrations over Chicago and its surrounding areas in response to projected changes in climate and anthropogenic emissions together from 1996-2000 to 2048-2052 and to 2095-2099 under the two scenarios. Under the A1FI scenario, the decreasing VOC/NOx ratio in future periods leads to reducing ozone concentrations over Chicago, despite the increasing emissions for both NOx and VOC. The MD8A ozone is decreased 0.2 ppb by 2048-2052 and increased 0.8 ppb by 2095-2099 relative to the 1996-2000 level; and the DM ozone are decreased 0.5 ppb during both future periods. The relatively small ozone reductions (and sometimes slight enhancements) are because the negative effects of decreasing VOC/NOx ratio on ozone are largely canceled out by the positive effects of climate change (increased summer temperatures, etc.) (Table 3, see also Hogrefe et al., 2004; Murazaki and Hess, 2006; Tao et al., 2007; Lin et al., 2008). Table 3 shows that, with warmer climate alone, the MD8A and DM ozone increases 8.7 and 5.1 ppb, respectively, over the City of Chicago from 1996-2000 to 2095-2099 under A1FI. By comparison, averaged over the Chicago area, the decreasing ratio of VOC/NOx has relatively small effects on surface ozone in future periods. For example, the MD8A ozone is increased by 7.1 ppb and 8.7 ppb from 1996-2000 to 2095-2099 due to climate change with and without changes in human-related emissions, respectively.

Projected ozone changes due to climate and anthropogenic emissions perturbations together under the B1 scenario are roughly opposite to those under A1FI over Chicago and its surrounding areas, especially from 1996–2000 to 2095–2099. Due to the larger decreases in NOx emissions than in VOC emissions, the VOC/NOx ratio is increased, thus the ozone concentration is enhanced over the City of Chicago and reduced over the surrounding areas. It is seen that the MD8A ozone increases 5.0 ppb and the DM ozone increases 7.8 ppb

Table 3

Present-day June–August surface ozone concentrations and their future changes in response to the climate change alone^a.

| Period | The Chicago area | | The City of Chicago | |
|--------------------------------|------------------------|------------------------|------------------------|------------------------|
| | Daily mean | MD8A ^b | Daily mean | MD8A |
| 1996-2000 | 27.9 (2.0) | 46.6 (3.2) | 19.1 (1.9) | 39.2 (4.1) |
| A1FI 2095-2099 B1 2095-2099 | 5.7 (64%) 2.1 (34%) | 8.4 (87%) 3.5 (31%) | 5.1 (89%) 2.8 (25%) | 8.7 (76%) 5.1 (22%) |

^a The modeled ozone changes are the result of all changes in climate and biogenic emissions only, i.e., the anthropogenic emissions are kept at the present-day levels. The numbers during the period of 1996–2000 denote the present-day 15-month (three summer months per year for 5 years) means and standard deviations (SDs, in parentheses) of surface ozone concentrations (units: ppb); and the numbers during 2095–2099 denote the absolute changes of the 15-month mean ozone (units: ppb) and the fractional changes of the 15-month ozone SDs (in parentheses) from 1996–2000 to 2095–2099.

^b Maximum daily 8-hour average ozone.

over the City of Chicago by 2095–2099. Meanwhile, the MD8A ozone decreases 4.5 ppb averaged over the Chicago area over the same period (Table 2 and Fig. 1), although the DM ozone increases slightly by 1.5 ppb primarily because of the decreasing NOx titration in the nighttime. By comparison, the warming climate alone results in 3.5–5.1 ppb increase in the MD8A ozone over both Chicago and its surrounding areas (Table 3).

Overall, incorporating changes in climate and anthropogenic emissions together, the modeled contrast in summertime surface ozone concentrations between the City of Chicago and the surrounding areas is enhanced from current to future periods under the A1FI scenario with increasing precursor emissions but reduced under the B1 scenario with decreasing precursor emissions (Table 2). The effects of climate warming itself, however, are to enhance the pollution levels under both scenarios, although more so under the A1FI than B1 scenario (Table 3).

For the City of Chicago, depending on the magnitudes of projected reductions in precursor emissions under the B1 scenario, projected ozone changes by 2048-2052 relative to 1996-2000 differ from changes by 2095-2099 (Table 2). In this case, NOx emissions decrease around 60% and VOC emissions decrease 40% from 1996-2000 to 2048-2052; the corresponding decreases by 2095-2099 are 80% and 60%, respectively. In response, the MD8A ozone increases 7.5 ppb by 2048-2052 but only 5.0 ppb by 2095-2099 relative to the 1996-2000 level. This difference is expected to be larger if the positive effects of climate warming on ozone are not included, since the climate warming is more significant by 2095–2099. It appears that the relative abundances of VOC and NOx during 2048-2052 lead to more efficient ozone production than two other periods being examined. This feature, however, is not clear from the projected changes in the DM ozone, which are also influenced by changes in nighttime ozone transformation through NOx titration; more NOx emissions increase the titration effects on ozone. Overall, changes in daytime ozone

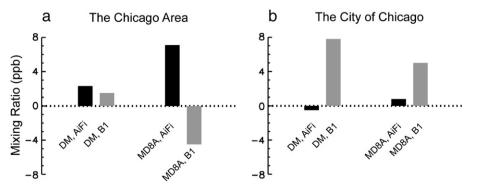


Fig. 1. Simulated changes in June-August surface ozone concentrations from 1996–2000 to 2095–2099 resulting from potential changes in climate, biogenic emissions, and anthropogenic emissions together. 'DM' represents the daily mean ozone and MD8A represents the maximum daily 8-hour average ozone.

production and nighttime ozone transformation together under B1 result in an increase of 6.1 ppb in the DM ozone by 2048–2052 as compared to the increase of 7.8 ppb by 2095–2099. Therefore changes in the relative abundances of NOx and VOC have larger impacts on the MD8A ozone than on the DM ozone.

Projected changes in seasonal variability of surface ozone

The temporal variation of ozone concentrations is another important aspect from the perspective of pollution control. Given a temporally averaged ozone level during a time period, a higher variation implies a higher probability of the occurrence of excessive ozone concentrations. Surface ozone over a given region is derived from two sources: (1) the production derived from local precursor emissions and (2) the transport from outside the region and the production due to the transported precursors. The first source depends greatly on the local meteorological conditions (i.e., air temperature, solar radiation, etc.) and the availability of local precursor emissions, and thus can vary significantly from time to time. The second source, however, largely contributes to the background ozone and varies relatively weakly. Therefore the temporal variation of total ozone concentrations is mainly determined by the variation in the first source. The enhancing (reducing) local emissions greatly increase (decrease) the temporal variation of the first source and thus the total ozone concentrations.

This feature is apparent from 1996-2000 to 2095-2099 because of the significant changes in local emissions. Incorporating changes in both anthropogenic emissions and climate together, the modeled 15month (three summer months per year for 5 years) variation of ozone concentrations over both Chicago and its surrounding areas is increased by 84-127% under A1FI, whereas there is a decrease of 23–30% under B1 (Table 2). Therefore, emission reduction would have another benefit on ozone mitigation by decreasing its temporal variation. In contrast, when anthropogenic emissions perturbations are excluded, the warming climate alone leads to a 22-89% increase in the ozone temporal variation over both Chicago and its surrounding areas (Table 3), more so under the A1FI than B1 scenario. This suggests that projected climate warming not just increases the overall summer ozone concentrations but also likely provides a meteorological condition in favor of high ozone events. Similarly, the study of Vavrus and Van Dorn (2010) suggests that the frequency, intensity, and duration of heat wave events will likely increase by the end of the century under the same scenarios. Mickley et al. (2004) also suggest that the climate changes projected for future years would induce more stagnant episodes, increasing the possibility of high ozone events.

Comparison with an alternative approach

Holloway et al. (2008) used a statistical downscaling approach, using the Statistical DownScaling Model (SDSM v. 4.1, Wilby et al., 2002) to analyze the effects of climate change projected by three general circulation models (GCMs), including the PCM, the Geophysical Fluid Dynamics Laboratory climate model (CM2.1), and the Hadley Centre Climate Model (HadCM3), on the summertime ozone concentrations over Chicago. First, the statistical relationships between surface ozone concentrations and a number of climate variables are established and validated using existing historical datasets. The climate variables highly correlated with ozone include surface temperature, surface winds, surface relative humidity, incoming solar radiation, and cloud cover. Then, these variables are selected as predictors, with values derived from the GCMs, to project future ozone change; the relationships between ozone and climate variables are assumed constant during current and future periods.

Using the PCM outputs as predictors, the statistical downscaling approach derives a 24% (or 12.3 ppb) increase in the MD8A ozone for the City of Chicago from 2002–2006 to 2070–2099 under the A1FI

scenario, as compared to the increase of 22% (or 8.7 ppb) from 1996-2000 to 2095–2099 by the RCAO dynamic downscaling approach discussed earlier. Averaged over the statistical downscaling calculations associated with the three GCMs, they projected an ozone increase of 17.0 ppb over Chicago from 2002-2006 to 2070-2099 due to climate warming alone. Under B1, it projects a much smaller increase of 4% (or 2.0 ppb) as compared to the increase of 13% (or 5.1 ppb) found by the RCAQ approach. The difference may be partially contributed by the small-scale meteorological processes that are resolved by the RCM but not by the GCMs. Nevertheless, the ensemble results of the three independent statistical downscaling calculations using the PCM, CM2.1, and HadCM3 outputs, respectively, produce a 12% (or 6.2 ppb) ozone increase under B1, which is close to our RCAQ result. Their high-end estimation associated with CM2.1, with ozone increases of 53% under A1FI and 16% under B1, is partly because CM2.1 has higher climate sensitivity than PCM used here.

Conclusions

Using a regional climate–air quality modeling system, this study investigates potential changes in summertime surface ozone concentrations for the City of Chicago and its surrounding suburban and rural areas from 1996–2000 to 2048–2052 and 2095–2099 under the IPCC SRES A1FI and B1 scenarios. Factors analyzed here include projected changes in climate (and the corresponding biogenic emissions) and anthropogenic emissions. The lateral boundary conditions for meteorological parameters are derived from PCM, which has relatively low climate sensitivity compared to other major GCMs (e.g., HadCM3 and CM2.1).

Model-derived ozone responses to simultaneous changes in climate and anthropogenic emissions are very different between the City of Chicago and its surrounding areas due to their differences in the relative abundances of NOx and VOC. Over the City of Chicago (presently in the VOC-limited regime for ozone production), the MD8A and DM ozone concentrations increase 5.0-7.8 ppb under B1 with warmer climate and reduced anthropogenic precursor emissions in the two future periods. They, however, change little under A1FI because the negative effects of increasing precursor emissions on ozone are largely canceled out by the positive effects of climate change (warmer summer temperatures, induced increases in biogenic emissions, etc.). The warming climate itself enhances the ozone concentrations by 2.8-8.7 ppb over the City of Chicago from 1996-2000 to 2095–2099, more so under the A1FI than B1 scenario. Average over the Chicago area (presently in the transition of VOC- and NOxlimited regimes for ozone production), climate warming and precursor emissions increases together lead to a 2.3-7.1 ppb increase in MD8A and DM ozone by 2095-2099 under A1FI. This differs from the B1 scenario, where while the reducing precursor emissions lead to a 4.5 ppb decrease in the MD8A ozone despite the positive effects of climate warming. Meanwhile, the warming climate itself leads to a 2.1-8.4 ppb increase of ozone under both scenarios. Our results on ozone impacts of climate change alone are similar to Holloway et al. (2008) under A1FI but higher than their results under the B1 scenario.

Projected temporal variation of surface ozone is greatly influenced by climate change and precursor emissions perturbations. From 1996–2000 to 2095–2099, the enhanced emissions under A1FI lead to an 84–127% increase in the 15-month variation of both MD8A and DM ozone over both the City of Chicago and its surrounding areas; whereas the reduced emissions under B1 lead to a 23–30% decrease in the 15-month ozone variation. This suggests that local emission regulation would have negative effects on the temporal variation of pollutant concentrations, likely leading to reducing possibility of high pollution events. By comparison, projected climate warming itself leads to a 22–89% increase in the temporal variation of ozone under both scenarios, suggesting more favorable meteorological conditions for high ozone events in future periods. This is consistent with more frequent and intensive heat waves in a warming world found by Vavrus and Van Dorn (2010).

Many difficulties exist in projections of surface ozone concentrations because of uncertainties in projections of future climate and precursor emissions as well as uncertainties in climate or air quality models being used. The A1FI and B1 scenarios, although giving a plausible range of future climate and precursor emissions, are certainly not what future world will follow exactly. The scaling factors for future anthropogenic emissions are adopted from the IPCC SRES estimate for the US and Canada and are not designed specifically for the Chicago area and do not differentiate individual sectors. Additionally, the PCM model used here has relatively low climate sensitivity; therefore, for a given scenario, our results on the ozone impacts of climate change alone are expected to be closer to the low end of current estimation. Other sources of uncertainty in our analyses include the exclusion of potential changes in wildfire emissions, the model spatial resolution, and the treatment of emissions scaling assuming constant land use. Also, the effects of climate change on concentrations of particulates in the Chicago area are also not examined in this study.

Nevertheless, this study, together with previous studies by Hogrefe et al. (2004), Murazaki and Hess (2006), Lin et al. (2008), Tao et al. (2007), and Holloway et al. (2008) using different scenarios and/or climate and air quality models, suggests that future climate changes could enhance the overall summertime ozone concentrations and likely increase the occurrences of high ozone events over the Chicago area. Furthermore, our findings are also consistent with Holloway et al. (2008) using the statistical approach for ozone projections. If anthropogenic emissions increase as projected by the A1FI scenario, summertime ozone concentrations would be reduced over the City of Chicago (see also Tao et al., 2007) due to the overall negative effects of increasing NOx on ozone when its production is limited by VOC (Sillman, 1999). In this case, however, we find not only the pollution levels over the surrounding suburban and rural areas are substantially enhanced, but the possibility of high ozone instances also likely increases significantly. Therefore, effective climate initiative and emission regulation are both necessary for ozone pollution mitigation over the Chicago area.

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