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Sensitivity of ozone to summertime climate in the eastern USA: A modeling case study

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Abstract

The goal of this modeling study is to determine how concentrations of ozone respond to changes in climate over the eastern USA. The sensitivities of average ozone concentrations to temperature, wind speed, absolute humidity, mixing height, cloud liquid water content and optical depth, cloudy area, precipitation rate, and precipitating area extent are investigated individually. The simulation period consists of July 12–21, 2001, during which an ozone episode occurred over the Southeast. The ozone metrics used include daily maximum 8 h average O_3 concentration and number of grid cells exceeding the US EPA ambient air-quality standard. The meteorological factor that had the largest impact on both ozone metrics was temperature, which increased daily maximum 8 h average O_3 by 0.34 ppb K^{-1} on average over the simulation domain. Absolute humidity had a smaller but appreciable effect on daily maximum 8 h average O_3 (−0.025 ppb for each percent increase in absolute humidity). While domain-average responses to changes in wind speed, mixing height, cloud liquid water content, and optical depth were rather small, these factors did have appreciable local effects in many areas. Temperature also had the largest effect on air-quality standard exceedances; a 2.5 K temperature increase led to a 30% increase in the area exceeding the EPA standard. Wind speed and mixing height also had appreciable effects on ozone air-quality standard exceedances.

Keywords: Ozone; Climate change; Meteorology

1. Introduction

High concentrations of ozone (O_3), a major constituent of air pollution, have detrimental effects on human health (Godish, 2004). Reactions of ozone with tissue in the airways are believed to cause weakened immune response, decreased lung function, and increased morbidity from asthma (Bernard et al., 2001; Levy et al., 2001). Also, ozone has been shown to cause significant damage to crops (Heck et al., 1982).

The conditions necessary for high ozone concentrations in the lower troposphere generally include warm weather (Sillman and Samson, 1995), sunlight, and stagnating high pressure systems, making episodes of high tropospheric ozone concentrations generally a summer phenomenon. Because of the

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size and duration of these warm high-pressure systems, ozone episodes tend to occur at a regional scale and last several days. Ozone is formed through complex interactions among nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight (Seinfeld and Pandis, 1998). Both NO_x and VOCs have natural and anthropogenic sources.

Ozone concentrations are influenced by meteorology in many ways. Ozone production is expected to be influenced by temperature because of the temperature dependences of the hundreds of reactions involved. A large contributor to this behavior is the temperature-dependent decomposition rate of peroxyacetylnitrate (PAN) and its homologs, which act as reservoir species for NO_2 (Sillman and Samson, 1995). Water vapor has competing effects on ozone levels. It begins with the photolysis of ozone, which can produce excited oxygen atom, $O(^1D)$, and an oxygen molecule. The $O(^1D)$ can then react with water vapor to produce a hydroxyl radical. The hydroxyl radical undergoes further reactions, some of which eventually lead to ozone production but many of which do not. The amount of ozone that subsequently forms depends on the NO_x/VOC mixture in a location. These reactions can collectively constitute a sink for ozone, due to the consumption of an O_3 molecule and an $O(^1D)$ atom, or they can produce more ozone molecules due to the subsequent chemistry of the hydroxyl radical. High wind speed is generally correlated with low pollutant concentrations due to enhanced advection and deposition; the processes involved, however, are complex, and in some places wind speed is positively correlated with ozone concentration (Tecer et al., 2003). Changes in cloud cover can affect the photochemistry of ozone production and loss, though the extent to which cloud cover affects ozone concentrations is thought to be small (Korsog and Wolff, 1991). Precipitation changes are expected to affect the rates of wet deposition of ozone, aerosols, and precursors. Additionally, changes in mixing height could affect reaction rates and the dilution of pollutants.

Emissions control policy is currently made assuming that climate will remain constant. However, climate changes over the next decades are expected to be significant and may impact O_3 concentrations; for example, global average temperatures are expected to rise 1.5–4.5 K over the next century (IPCC, 2001). Predictions of how wind speed will change in the USA vary depending on the area in question and on the model used. Predictions from one study differ in different areas in the same state (Bogardi and Matyasovszky, 1996). Breslow and Sailor (2002) predict decreases in wind speeds over the USA in the next 50 years. Water vapor concentration (absolute humidity) is generally expected to increase due to the higher saturation vapor pressure of water at higher temperatures (IPCC, 2001); to first order, it is expected that relative humidity remain constant with climate change (Held and Soden, 2000). Norris (2005) has observed decreases in cloud cover in recent decades over most of the planet. Simulations using general circulation models (GCMs) indicate that cloud cover will decrease if temperature is increased (Cess et al., 1990). GCM studies also predict small changes in summer and annual mean precipitation over the eastern USA (Räisänen, 2005); Leung and Gustafson (2005), however, predict changes in the number of summer days with precipitation in the eastern USA. Mickley et al. (2004) and Hogrefe et al. (2004) report increases in mixing heights for simulated future climates, though Murazaki and Hess (2006) see no significant changes in mixing heights in a future climate.

Determining how air quality changes as climate changes is an important step toward estimating future air quality. This may allow policy planners to relax the assumption of constant climate and meteorology, or it may indicate that the assumption of constant climate will have little effect on predicted air quality. It will also help show if climate changes can be accounted for with simple corrections to models run with constant climate or if a sophisticated modeling framework is necessary. In any case, the effects of climate changes on air quality must first be characterized.

The response of ozone to changes in temperature has been examined in the past with both process modeling and statistical studies. Higher temperatures have generally been associated with higher ozone concentrations (Tecer et al., 2003; Bloomfield et al., 1996; Guicherit and van Dop, 1977; Menut, 2003; Neftel et al., 2002), with some exceptions (McMillan et al., 2005). In an observational study of ozone in Chicago, ozone concentrations increase with temperature on days with high temperatures over approximately 50°F (Bloomfield et al., 1996). A chemical transport modeling study simulating an ozone episode over Milan in May 1998 finds a linear positive correlation between peak ozone concentration and temperature (Baertsch-Ritter et al., 2004).

In a modeling study of an ozone episode over southern California in September 1996, Aw and Kleeman (2003) calculate an increase in peak ozone with temperature. High water vapor concentrations have been shown to have inconsistent effects on ozone concentrations from location to location or even at a fixed location due to these canceling influences on chemistry (Jacob et al., 1993), depending on whether an area is VOC- or NO_x-limited (Baertsch-Ritter et al., 2004). Baertsch-Ritter et al. (2004) also notice a weak connection between water vapor and ozone concentrations. Korsog and Wolff (1991) see a negative correlation between cloud cover and ozone, though the correlation between temperature and ozone was stronger. These studies have focused on small areas (e.g., one city) during ozone episodes; the response of ozone over both episode and non-episode areas or over large regions has been the focus of little research. Additionally few studies (Baertsch-Ritter et al., 2004) have calculated sensitivities of ozone concentrations to a comprehensive suite of specific meteorological parameters.

Several studies have used GCM-predicted future climates in models to predict future ozone concentrations. Hogrefe et al. (2004) examine ozone changes over the eastern USA under a changed climate and determine that average daily maximum 8 h ozone concentrations increase by 2.7 ppb by the 2020s and 5 ppb by the 2080s. Murazaki and Hess (2006) predict decreases in background ozone but increases in ozone in areas with high NO_x emissions. These studies do not separate the effects of specific meteorological changes on ozone; instead, they estimate the response to a combined set of changes in climate parameters.

The goal of this study is to determine how concentrations of ozone over the eastern USA respond to changes in climate parameters, specifically temperature, wind speed, absolute humidity, mixing height, cloud cover, and precipitation. This work investigates each of these parameters separately so that the effects of each can be determined. This will help identify the major factors that could have an effect on air quality as climate changes by determining the meteorological factors to which ozone concentrations have the largest sensitivities.

2. Methods and model description

The PMCAMx model (Gaydos et al., 2006) is the modeling tool used in this study. This model uses the framework of CAMx v. 4.02 (Environ, 2004) to simulate horizontal and vertical advection, horizontal and vertical dispersion, wet and dry deposition, and gas-phase chemistry. The Carbon-Bond IV mechanism (Gery et al., 1989), including 34 gas-phase and 12 radical species, was used for gas-phase chemistry calculations. Photolysis rates were calculated using the

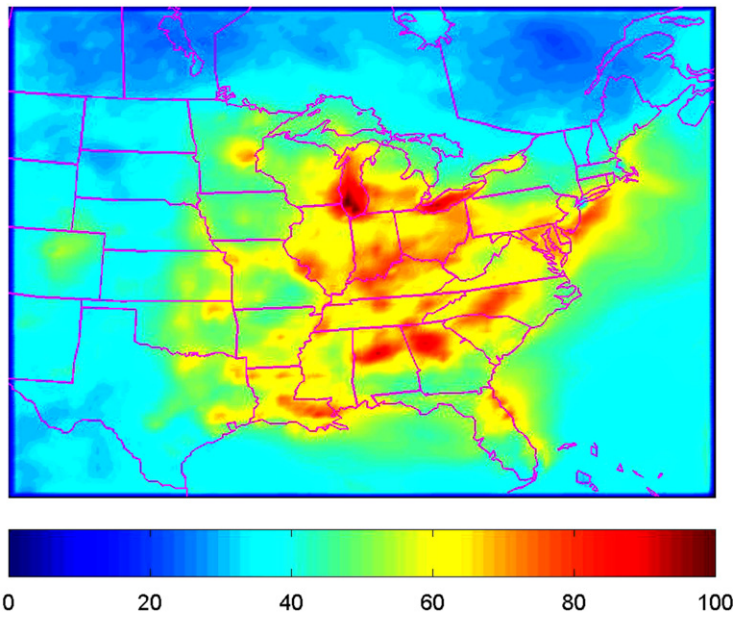

Fig. 1. Average daily maximum 8 h average O_3 concentrations (ppb) for the base case for the period 12–21 July 2001.

RADM method of Chang et al. (1987). The aerosol chemistry and physics modules outlined in Gaydos et al. (2006) were included in the model, though results for the effects of meteorological changes on particulate matter concentrations will be presented in future work. The modeling domain was the eastern half of the US (Fig. 1), and a 36×36 km resolution grid was used with 14 vertical layers, extending from the surface to an altitude of approximately 6 km. Inputs to the model included meteorological conditions, land use data, emissions, and initial and boundary conditions of ozone and aerosol concentrations. The emissions inventory used was the Midwest Regional Planning Organization's Base E inventory (LADCO, 2003), including BIOME3 biogenics (Wilkinson and Janssen, 2001). The period modeled was 12–21 July 2001; the first 3 days were used as model initialization days and were excluded from the analysis. The meteorological input into the model was generated by MM5 using assimilated meteorological data. Over this time period, hot conditions over the Southeast were largely responsible for an ozone episode extending from Atlanta to New Orleans. Temperatures were rather high in the Southeastern, Plains, and Midwestern states and low in the Northeast. Model results are expected to be dependent on this choice of time periods.

Measured and modeled base case ozone concentrations for 15–21 July 2001 in Atlanta, Kansas City, Missouri, and Pittsburgh are shown in Fig. 2. Ozone measurements are from the EPA AIRS database. Concentrations of other gas-phase and aerosol-phase species in Pittsburgh were shown to have reasonable agreement by Gaydos et al. (2006). Some of the discrepancies between the model and measurements are due to some discrepancies

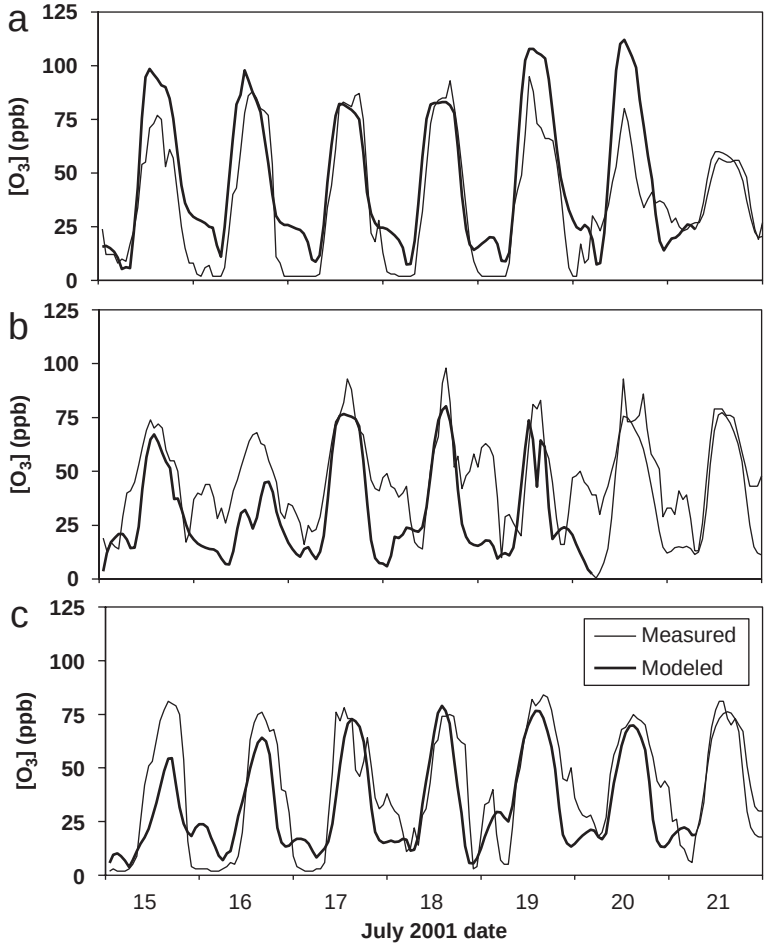

Fig. 2. Measured and modeled hourly ozone concentrations in (a) Atlanta, (b) Kansas City, Missouri, and (c) Pittsburgh for 15–21 July 2001.

between the meteorology predicted by MM5 and the actual meteorology (Gaydos et al., 2006). Overall, PMCAMx shows reasonable skill in capturing the major characteristics of air quality over the eastern USA during this period.

In addition to a base case scenario, a suite of sensitivity simulations were run in which individual meteorological parameters were perturbed to varying degrees (Table 1). Except for cloud and precipitation changes discussed in more detail below, all perturbations were imposed uniformly in space and time on the modeling domain. For example, all surface and air temperatures were increased by 0.5, 1, 1.5, 2.5, 4 and 5 K, keeping other inputs constant. Simulations were run in which base case horizontal wind speed was decreased or increased by 5% or 10%. (Vertical wind speeds were calculated from these horizontal wind speeds to ensure mass conservation.) Simulations to test sensitivity to humidity were run by increasing or decreasing water vapor concentrations by 5%, 10%, or 20%. Sensitivity to cloud liquid water content (LWC) and optical thickness was tested by increasing or decreasing simultaneously both LWC and optical depth (OD) by 5%, 10%, or 20%; sensitivity to precipitation rate was tested by increasing or decreasing precipitation by 5%, 10%, or 20% (while keeping other cloud parameters constant). These sensitivity simulations to cloud LWC/OD and precipitation rate were performed without changing the cloudy or precipitating area. Separate simulations to test sensitivity to cloud cover area were run in which total cloudy area was adjusted by the following factors: −3.9%, −2.5%, +2.2%, and +4.1%. Sensitivity to the area extent of precipitation was tested in simulations in which the area undergoing precipitation was adjusted by the following factors: −4.9%, −2.3%, +2.4%, +4.7%. Base-case cloud cover and precipitation are shown in Fig. 3. The area of cloud cover and precipitation were changed by growing (or shrinking) existing cloudy or precipitating areas into randomly selected but adjacent non-cloudy or non-precipitating cells, where the random selection process was manipulated to achieve the desired number of changed cells. It is important to note that cloud cover and precipitation were changed independently of one another so that their effects could be separated.

Table 1
Meteorological perturbations in this study and quantities directly affected by perturbations

Meteorological parameter	Changes in values examined	Directly affected in simulation
Temperature	+0.5, 1.0, 1.5, 2.5, 4.0, 5.0 K	Reaction rates, aerosol thermodynamics
Wind speed	±5, 10%	Vertical velocity/dilution/entrainment, advection, diffusion coefficients, dry deposition resistance
Absolute humidity	±5, 10, 20%	Reaction rates with [H_2O], aerosol thermodynamics
Mixing height	±One model layer	Vertical diffusivities in layers near mixing height
Cloud LWC and OD	±5, 10, 20%	Radiation transmittance of clouds, aqueous chemistry
Area of cloud cover	−3.9, −2.5, +2.2, +4.1%	Radiation transmittance of clouds, aqueous chemistry
Precipitation rate	±5, 10, 20%	Wet deposition
Area of precipitation cover	−4.9, −2.3, +2.4, +4.7%	Wet deposition

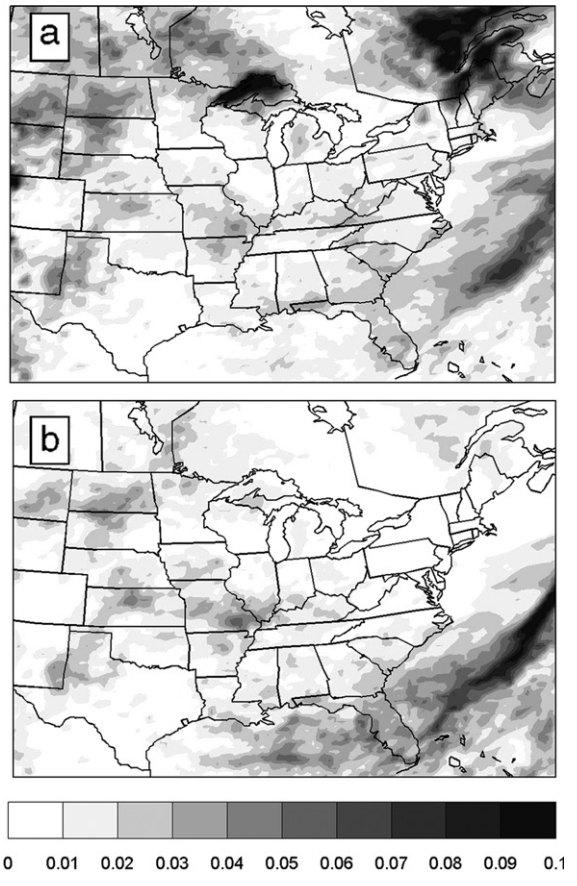

Fig. 3. Column- and simulation-averaged base case cloud LWC (a) and precipitation water ($g\,m^{-3}$).

Additionally, sensitivity to mixing height was tested by simulations in which the mixing height, as determined from vertical diffusivities by the method of O'Brien (1970), was increased or decreased by one model layer by changing the vertical diffusivity in only the layer immediately above (in the case of mixing height increase) or below (in the case of mixing height decrease) the original mixing height. Mixing height changes were implemented only when vertical diffusivities had the polynomial shape of O'Brien (1970) from which a mixing height could be determined (approximately 2/3 of grid cell time steps). This corresponded to average changes in mixing height of approximately 150 m. All simulations used the same anthropogenic and biogenic emissions as in the base case.

The main metrics used for comparison are the number of surface grid cells exceeding the US EPA's 8 h average O_3 concentration standard of 0.08 ppm at any point during the simulation as well as the simulation-long average daily maximum 8 h O_3 concentration (both in specific locations and averaged over all land grid cells in the domain), following Hogrefe et al. (2004). Only concentrations in surface-level grid cells are considered. While both metrics emphasize peak daytime ozone concentrations, the exceedance metric by definition focuses on heavily polluted or "episode" areas while the daily maximum ozone metric includes the response of ozone under background conditions as well. The exceedance metric also takes into account some of the spatial variability in responses, which frequently vary around the domain. Because ozone concentrations can be more or less sensitive under polluted versus background conditions, we will see that these metrics often yield different, but complementary, information. The base case values for the average daily maximum 8 h ozone concentrations are shown in Fig. 1.

3. Results and discussion

3.1. Temperature

Both air-quality standard exceedances ($R^2 = 0.999$) and the average daily maximum 8 h ozone concentration ($R^2 = 0.998$) increased linearly with temperature (Fig. 4(a)), which is a somewhat surprising result given the non-linear nature of the chemistry of ozone formation. The fraction of land area over the air-quality standard increased linearly from 8.6% (or 509 grid cells) in the base case to 14.0% (831 grid cells) in the $T+5$ K case. The simulation- and domain-averaged daily maximum 8 h average ozone concentration increased linearly by 0.34 ppb K^{-1}. The response was non-uniform throughout the domain, and the variability of responses is considered in Section 4 (along with the variability in responses to the other meteorological perturbations). Additionally, the peak hourly concentration at any point in the domain for the entire simulation increased by an average of 4.7 ppb K^{-1}. This is somewhat larger than the 3.2–3.5 ppb K^{-1} calculated for the Los Angeles area (Aw and Kleeman, 2003) and the 2.8 ppb K^{-1} for Milan, Italy (Baertsch-Ritter et al., 2004), though less than the 6 ppb K^{-1} calculated using daily maximum temperature downwind of Milan (Neftel et al., 2002). The largest increases came in areas already experiencing high ozone concentrations, but temperature increases had little effect in areas with low ozone concentrations.

The differences in daily maximum 8 h ozone concentration between the $T+2.5$ K case and the base case are shown in Fig. 5. Generally, ozone concentrations in remote or oceanic areas changed little or decreased slightly, while concentrations in more populated and polluted areas increased appreciably with temperature. Areas with low base case ozone concentrations tended to have smaller sensitivities to temperature than did areas with high base case ozone concentrations. In Atlanta, for example, the average daily maximum 8 h average O_3 concentration increased by approximately 4 ppb for a 2.5 K temperature increase. Many areas that, under base case conditions, were slightly under the 80 ppb limit were pushed over the limit as temperatures increased. The effect of temperature on ozone concentrations appears to be a large one; increasing temperatures by 5 K increased by 63% the area exceeding the air-quality standard during the simulated week. This suggests that the effect of rising temperatures on ozone concentrations is important.

PAN chemistry is largely responsible for the dependence of ozone formation on temperature. In the episode area, hourly PAN concentrations tended to be lower for the high-temperature cases than for the base case, indicating that PAN was decomposing to form peroxyacetyl radical and NO_2, which could then form ozone. These differences in PAN concentrations occurred primarily during daylight hours, when photolysis of NO_2 leads to ozone production. Outside the episode area,

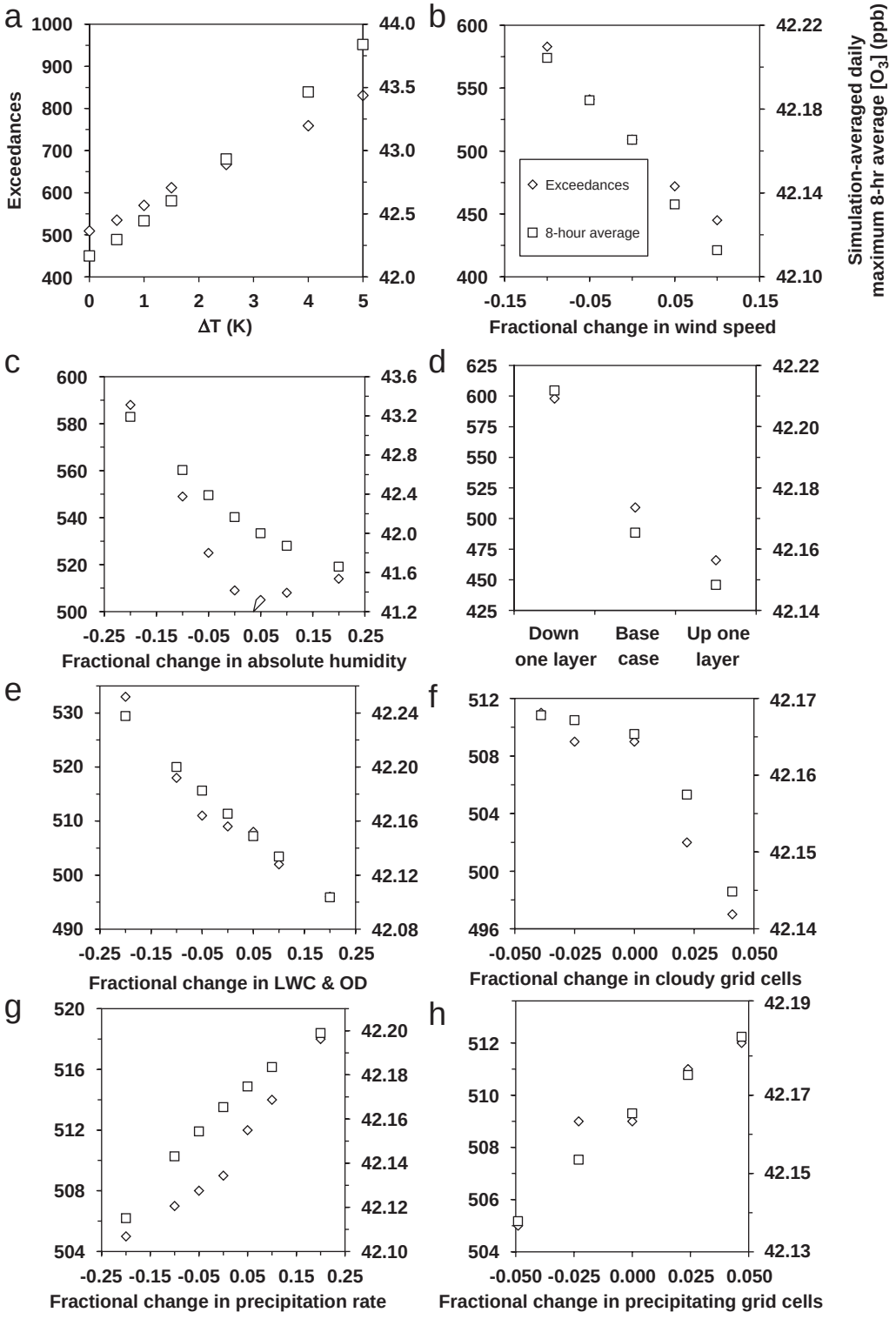

Fig. 4. Exceedances and simulation-averaged daily maximum 8 h average O_3 concentrations versus perturbation in (a) temperature, (b) wind speed, (c) absolute humidity, (d) mixing height), (e) cloud LWC and optical depth, (f) cloudy area, (g) precipitation rate, and (h) precipitating area.

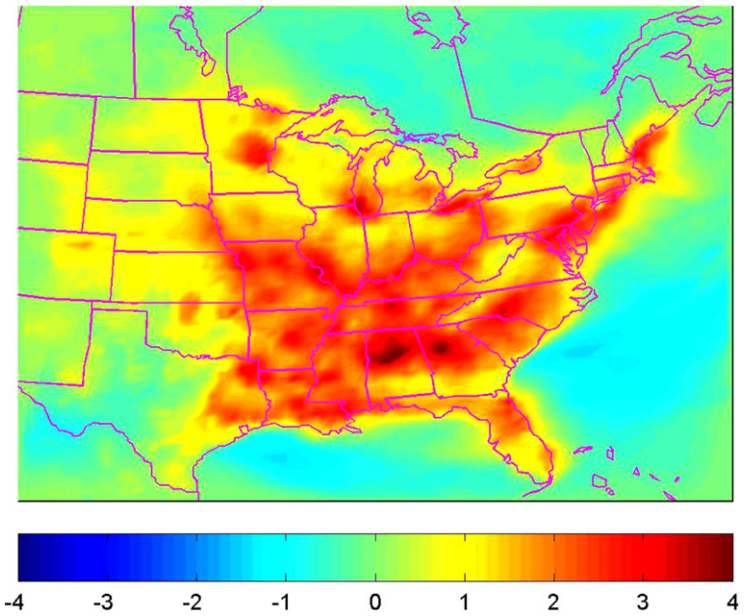

Fig. 5. Differences in average daily maximum 8 h average O_3 concentration (ppb) between $T+2.5$ K case and base case.

both PAN and NO_x concentrations remained nearly constant among simulations. An additional simulation, in which a 2.5 K temperature increase affected only PAN formation and decomposition rates, was compared to the full $T+2.5$ K simulation. Allowing the temperature change to affect only the PAN chemistry accounted for 97% of the change (between the full $T+2.5$ K case and the base case) in grid cells exceeding the air-quality standard and overpredicted the change in average daily maximum 8 h average O_3 concentration by only 16%.

Two additional simulations were run to examine the robustness of these results. A base case spanning 19–28 July 2001 (disregarding three spin-up days) and a $T+2.5$ K simulation for the same period were run. In this case, the area exceeding the air-quality standard increased by 37% for a 2.5 K temperature increase, and average daily maximum 8 h average [O_3] increased by 1.7% or 0.70 ppb, while in the original simulations exceedances increased by 31%, and average daily maximum 8 h average [O_3] increased by 1.8% or 0.77 ppb. These indicate that there is at least some robustness with respect to meteorological conditions for these results.

3.2. Wind speed

Both the exceedances and the average daily maximum 8 h ozone concentration tended to decrease as wind speed was increased (Fig. 4(b)). The relation for each metric with wind speed was linear (exceedances $R^2 = 0.995$; 8 h O_3 $R^2 = 0.993$). Decreasing wind speed by 10% increased the area over the air-quality standard from 509 cells in the base case to 583 cells in the reduced wind case, an increase of 14.5%. A 10% reduction in wind speed also led to a small increase in average daily maximum 8 h ozone concentration over all land grid cells—from 42.17 ppb in the base case to 42.20 ppb in the reduced wind case. The small change in average daily maximum 8 h ozone concentration is due largely to the nonuniformity of responses throughout the domain. The changes in daily maximum 8 h average ozone concentration for a 5% reduction in wind speed are shown in Fig. 6. Generally, a decrease in wind speed caused daily maximum 8 h ozone concentrations to increase in polluted areas and decrease in remote or less-polluted areas. The temporal responses of ozone concentrations in Atlanta and Pittsburgh for the 5% reduction in wind speed case with respect to the base case are shown in Fig. 7. Ozone concentrations in Atlanta, which already had high base case ozone concentrations were consistently greater than base case conditions, while concentrations in Pittsburgh, which had lower ozone concentrations during the period modeled, had both positive and negative changes, resulting in a smaller average change in Pittsburgh than in Atlanta.

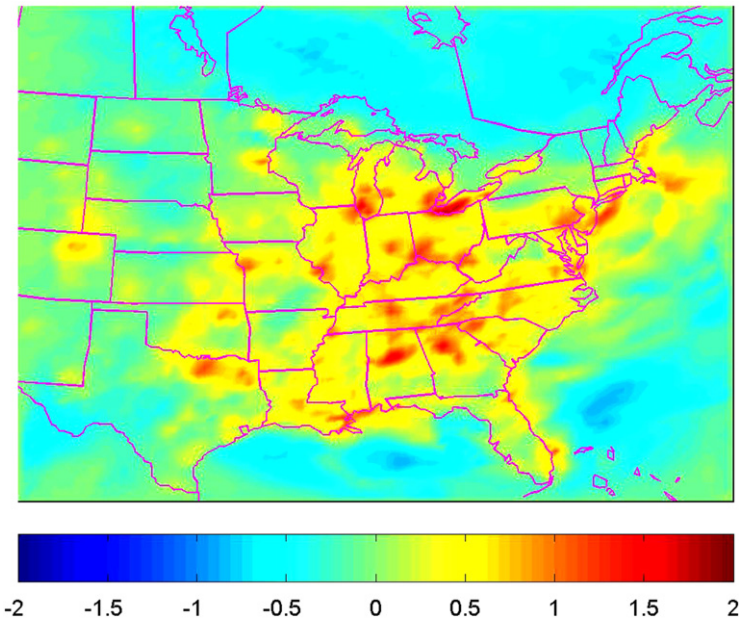

Fig. 6. Differences in average daily maximum 8 h average O_3 concentration (ppb) between Wind-5% case and base case.

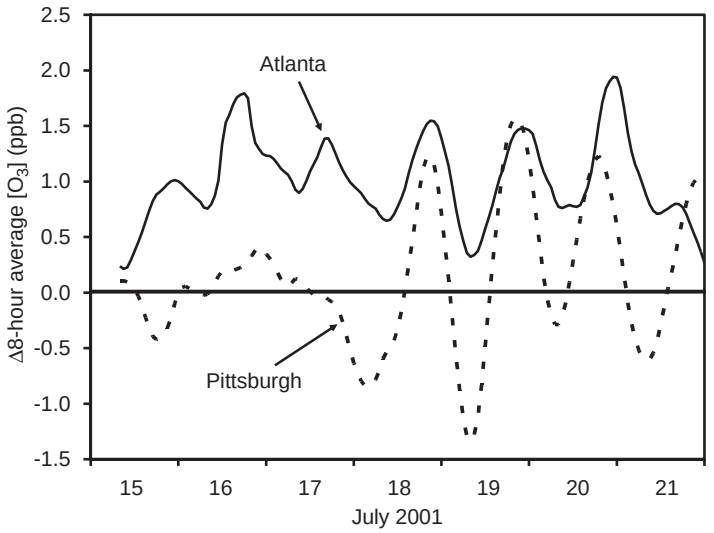

Fig. 7. Differences in moving 8 h average ozone concentration in Atlanta and Pittsburgh between Wind speed-5% simulation and base case. A positive $\Delta[O_3]$ corresponds to an increase of O_3 over the base case when the wind speed is decreased.

Changes in dry deposition, mixing, and dilution contributed to the response of ozone concentrations to changes in wind speed. A 10% reduction in wind speed resulted in an approximately 1.5–2% reduction in dry deposited ozone mass, a 4–6% reduction in chemical consumption of ozone, and a 4–6% decrease in the net ozone mass flux advected into the domain, resulting in a $<1\%$ increase in average daily maximum 8 h average ozone concentrations. A reduction in wind speed of 10% decreased horizontal advection of ozone into the domain by 9.7% and horizontal advection out of the domain by 10.8%. Thus it appears that changes in dry deposition, mixing, and dilution of ozone and dilution of precursors all contribute to the link between ozone concentrations and wind speed. The

resulting changes in ozone concentrations due to changes in wind speed are appreciable given the changes in cells exceeding the air-quality standard, but still considerably smaller than the changes in ozone due to changes in temperature; the sensitivity of ozone concentrations on wind speed, therefore, appears to be of secondary importance.

3.3. Absolute humidity

The response of ozone concentrations to changes in absolute humidity was a complicated one, varying in space for a given change in humidity and varying even in the sign of the response to increases in humidity. The average daily maximum 8 h average ozone concentration decreased steadily as absolute humidity was increased (Fig. 4(c)); this response was nearly linear ($R^2 = 0.957$). When absolute humidity was decreased 20%, the average daily maximum 8 h average ozone concentration decreased by 0.5 ppb, from 42.2 to 41.7 ppb. The same trend does not hold for ozone air-quality standard exceedances. The response of the exceedances resembled a parabola with a minimum number of exceedances occurring when humidity was increased by 5%. For increases in absolute humidity, air-quality standard exceedances changed very little, though for decreased humidity, exceedances increased more dramatically. Though ozone air-quality standard exceedances responded more strongly to changes in wind speed than changes in absolute humidity, humidity had the stronger effect on average daily maximum 8 h average ozone concentration. The large impact of changes in humidity on average daily maximum 8 h average ozone concentration appears to make this an important sensitivity; however, the very small response in air-quality standard exceedances that correspond to increases in absolute humidity underscores the complexity of the hydroxyl radical chemistry that follows ozone photolysis and reaction of $O(^1D)$ with water vapor and its self-canceling effects on ozone concentrations. The sensitivity of ozone concentrations on absolute humidity appears to be of secondary importance in this case.

The differences in average daily maximum 8 h average ozone concentration are shown in Fig. 8. In general, increases in water vapor led to decreases in ozone concentrations in the hotter South (including over the Gulf of Mexico and the Atlantic Ocean), Great Plains, and Great Lakes regions and mixed responses in the cooler North. In Fig. 8, the warmer areas showed decreases in ozone concentrations while cooler areas mostly showed little response or small increases in daily maximum 8 h average ozone concentration. For the 20% increase in absolute humidity, differences in Atlanta in 8 h average ozone concentration remained below base case values for the entire simulation, but in Pittsburgh

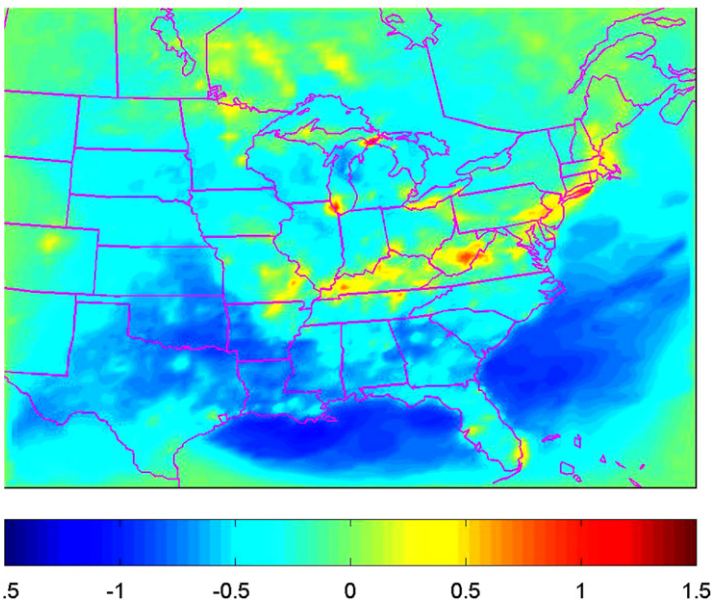

Fig. 8. Differences in average daily maximum 8 h average O_3 concentration (ppb) between Absolute humidity + 10% case and base case.

the sign of the difference changed between positive and negative (Fig. 9).

3.4. Mixing height

Increasing the mixing height by one model layer (approximately 150 m on average) decreased both the exceedances and the average daily maximum 8 h average ozone concentration (Fig. 4d). Decreasing the mixing height had the opposite effect. A one-level decrease in mixing height changed the number of cells over 8 h ozone air quality standard from 509 in the base case to 598, while a one-level increase in mixing height lowered the number of exceedances to 466. Decreasing mixing height increased the average daily maximum 8 h average ozone concentration from 42.17 ppb in the base case to 42.21 ppb, while increasing mixing height decreased this average to 42.15 ppb. The changes in the exceedances indicate that the link between mixing height and ozone concentrations is an important one, even if the effect on average daily maximum 8 h average ozone concentrations was rather small.

The differences in average daily maximum 8 h average ozone concentrations between the reduced mixing height case and the base case are shown in Fig. 10. Generally, decreasing mixing height increased average daily maximum 8 h average ozone in the more populated or polluted areas, and decreased ozone in some less polluted or remote areas. Atlanta and New Orleans fell into the first group, with decreased mixing height mostly leading to increased ozone and 8 h average concentrations differing by as much as 3.4 ppb from base case conditions. Remote Canadian and oceanic areas fell into the latter group, with decreased mixing height leading to decreased ozone and increased mixing height leading to increased ozone. The New York/Philadelphia area also fell into this group. Mixing height, in other words, had a larger impact when O_3 was elevated (~80 ppb) than during average (~40 ppb) periods. This may be due to changes in the NO_x/VOC chemistry in the area due to changes in dilution, and there may also be changes in the overnight ozone sink via NO_x. Though the hourly response of ozone concentrations fluctuated in sign, the changes in daily maximum 8 h average concentration tended to have the same sign in a given location; in Atlanta and New Orleans, an increase in mixing height resulted in an average decrease in daily maximum 8 h average ozone of 0.57 and 0.38 ppb respectively, and in Pittsburgh, a decrease in mixing height resulted in an average increase in daily maximum 8 h average ozone of 0.29 ppb.

3.5. Cloud LWC and OD

Both the exceedances and the simulation-average daily maximum 8 h average ozone concentration decreased as LWC and OD were increased

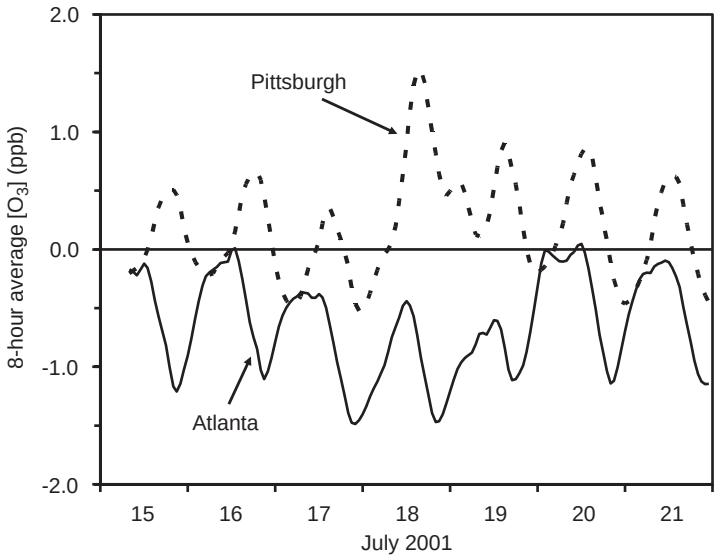

Fig. 9. Differences in moving 8 h average ozone concentration in Atlanta and Pittsburgh between Absolute humidity + 20% simulation and base case. A positive $\Delta[O_3]$ corresponds to an increase of O_3 over the base case when the absolute humidity is increased.

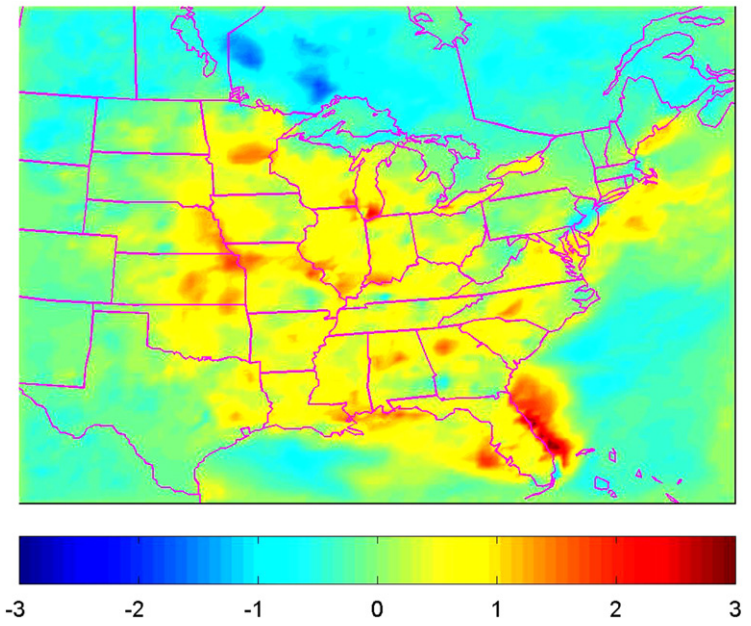

Fig. 10. Differences in average daily maximum 8 h average O_3 concentration (ppb) between reduced mixing height case and base case.

(Fig. 4(e)). The slope of the linear ($R^2 = 0.997$) relation between average daily maximum 8 h average ozone concentration and change in LWC and OD was -0.003 ppb $\%^{-1}$, which is rather small compared to most of the non-cloud meteorological factors investigated. The relation between air-quality standard exceedances and change in LWC and OD was also rather linear ($R^2 = 0.936$). The response in the number of grid cells exceeding the air-quality standard was also small; a 20% decrease in LWC and OD increased the exceedances from 509 to 533, which is a 4.7% increase.

For the entire simulation period, 8 h average O_3 stayed with 1.0 ppb of base case values for a 10% increase or decrease in LWC and OD in Atlanta, New Orleans, and Pittsburgh. Most hourly concentrations differed by less than 0.1 ppb from base case concentrations, though some differed by several ppb (up to 5 ppb). The net result of these changes in hourly concentrations was the small change in 8 h average concentrations. The largest increase in average daily maximum 8 h average ozone concentration for a 10% decrease in LWC and OD was 1.4 ppb near Miami. The small responses in both the number of air-quality standard exceedances and daily maximum 8 h average concentrations indicate that changes in cloud LWC and OD, while keeping cloudy area fixed, have small impacts on ozone concentrations.

3.6. Cloud cover

Both the exceedances and the average daily maximum 8 h average ozone concentration tended to decrease slightly as the number of cloudy grid cells was increased, though the relations were both non-linear (Fig. 4(f)). Generally, decreases in cloud cover had little effect on ozone concentrations, and increases in cloud cover led to small decreases in ozone. Changing cloud cover by approximately 4% kept average daily maximum 8 h average O_3 within 1.5 ppb of base case conditions for each location in the domain.

One of the areas that saw the most dramatic effect of cloud cover on ozone concentrations was Atlanta. A 4.1% increase in cloud cover resulted in a decrease in average daily maximum 8 h ozone concentration of approximately 1 ppb in Atlanta. The changes in 8 h average ozone concentrations for an increase and a decrease in cloud cover are shown in Fig. 11. The decrease in cloud cover had a minute effect on ozone concentrations in Atlanta and that an increase in cloud cover generally led to a small but appreciable decrease in 8 h average ozone concentrations (with a small increase in ozone at the end of the simulation). In Pittsburgh, 8 h average O_3 differed by no more than 0.12 ppb from base case conditions for either the 4.1% increase or 3.9% decrease in cloud cover. Additionally, 8 h

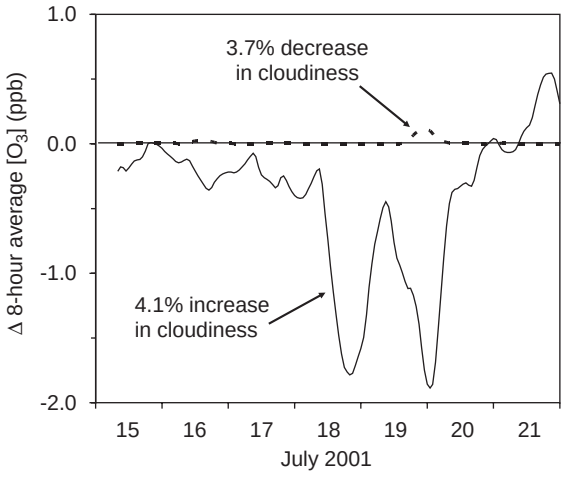

Fig. 11. Differences in moving 8 h average ozone concentration in Atlanta between 4.1% increase and a 3.7% decrease in cloud cover and base case. A positive $\Delta[O_3]$ corresponds to an increase of O_3 over the base case when the cloudiness is changed.

average ozone concentrations for these two simulations in New Orleans differed by no more than 0.2 ppb from base case conditions. Some areas, however, did see large changes in ozone concentrations. For example, in areas near the western and southern borders of Missouri, average daily maximum ozone concentrations decreased by more than 2 ppb when cloudy area was increased by 4.1%. These areas, however, were not near the air-quality standard under base case conditions, so there was little effect on the number of cells over the air-quality standard. These changes were somewhat significant over a rather small area; generally, only areas near the interface between cloudy and non-cloudy areas would likely be affected by this cloud adjustment scheme, and areas with cloud cover tend to have low ozone concentrations, so it is not surprising that this scheme would affect areas with lower base case ozone more than other areas. These relatively small responses indicate that the sensitivity of ozone concentrations to changes in cloud cover is appreciable for the average concentration metric, but small for the exceedance metric.

3.7. Precipitation rate

Holding the area of precipitation constant, increases in the rate of precipitation led to very small increases in both average daily maximum 8 h average O_3 and exceedances of the ozone air-quality standard (Fig. 4(g)). The average daily maximum 8 h average concentration increased linearly ($R^2 = 0.983$) with a slope of 0.002 ppb%$^{-1}$, and the number of exceedances of the air-quality standard also increased rather linearly ($R^2 = 0.959$) with precipitation rate. The exceedances increased from 509 in the base case to 518 when precipitation rate was increased by 20%. The sensitivity of ozone to precipitation rate, using both ozone metrics, therefore, appears to be a secondary one.

Changing precipitation rate by 10% changed 8 h average ozone concentrations by less than 0.3 ppb in Atlanta and New Orleans. However, these areas generally saw clear conditions in the base case, so it is not surprising that adjusting the rate of precipitation without adjust the area of precipitation would have little effect in these areas. In Pittsburgh, where conditions were less clear and base case ozone concentrations were lower, a 10% increase in precipitation rate increased the 8 h average ozone concentration by 1.0 ppb on 1 day and by no more than 0.2 ppb on the other days. The small changes in ozone concentrations indicate a secondary sensitivity of ozone concentrations to precipitation rate when precipitating area is held constant.

3.8. Precipitation extent

Both exceedances of the ozone air-quality standard and average daily maximum 8 h average ozone concentrations changed little as the area undergoing precipitation was increased (Fig. 4(h)). The average daily maximum 8 h average ozone concentration increased linearly ($R^2 = 0.992$) with the area undergoing precipitation, while the response of air quality standard exceedances was somewhat nonlinear. For an approximately 5% change in precipitating area, the number of grid cells exceeding the air-quality standard changed by less than 1%, and the average daily maximum 8 h average ozone concentration changed by less than 0.03 ppb. For a 5% decrease in precipitating area, the largest decrease in the average ozone metric was 3.3 ppb in a small area on the Tennessee–Kentucky border. These sensitivities are quite small compared to the sensitivity of ozone concentrations to other meteorological parameters.

The changes in the ozone metrics that do occur are generally due to small changes in wet deposition of ozone precursors. Deposition rates of ozone changed by less than 1% between the changed-precipitation cases and the base case. It appears that

removing small amounts of precursor gases changed the concentrations of species in the NO_x–VOC system sufficiently to generate slightly more ozone. Additionally, it is worth noting that changes in dry deposition that result from changes in soil moisture are not included in the model; it is likely that inclusion of this would reduce the already small increase of ozone concentrations with precipitating area.

3.9. Strength of interactions

An additional simulation was run to test the additivity of and interactions among individual meteorological perturbations, since future changes in climate will affect more than just temperature or any other single meteorological factor. The changes imposed in this simulation are shown in Table 2. The changes in ozone resulting from this combined-change simulation were compared to the sum of the changes that resulted when each of the eight changes were imposed separately. The individual changes are also shown in Table 2. The average daily maximum 8 h average ozone concentration in this combined-change simulation was 0.19 ppb greater than the base case value. The sum of the changes in average daily maximum 8 h ozone from the individual perturbations that comprise this combined scenario was 0.41 ppb, and the change predicted for only a 2.5 K increase in temperature was 0.77 ppb. Using exceedances of the air-quality standard as the relevant ozone metric yielded similar results. The number of grid cells exceeding the air-quality standard in this combined-change case was 23 greater than the number of exceedances in the base case. The sum of changes due to the individual perturbations was 66 grid cells, and the change due to the 2.5 K temperature increase alone was 158 cells. From these results, it appears that changes in other meteorological factors can potentially mitigate the impact of temperature on ozone concentrations and that the sum of the changes due to individual meteorological perturbations generally does not add up to the change that results when the perturbations are imposed simultaneously. However, the sum of the individual changes is a reasonable order-of-magnitude approximation of the combined change.

Table 2
Meteorological perturbations in combined-change simulation

Meteorological parameter	Change in parameter	Change in 8 h metric due to parameter alone (ppb)	Change in exceedance metric due to parameter alone
Temperature	+2.5 K	0.77	158
Wind speed	+5%	−0.03	−37
Absolute humidity	+10%	−0.29	−1
Mixing height	+One model layer	−0.02	−43
Cloud LWC and OD	+10%	−0.03	−7
Precipitation rate	+10%	0.02	5
Area of cloud cover	+4.1%	−0.02	−12
Area of precipitation cover	+4.7%	0.02	3
Sum of individual changes:		0.41	66
Predicted from combined-change simulation:		0.19	23

4. Comparison among meteorological parameters

In order to compare the meteorological sensitivities of the two ozone metrics to one another, the calculated sensitivities were multiplied by potential future changes in the corresponding meteorological parameters to yield an estimate of a range of expected changes in ozone due to each individual meteorological parameter. These changes are summarized in Table 3. The projected meteorological changes in Table 3 have been investigated to varying degrees. A large body of work exists on future temperature changes, for example, but there is little and conflicting work on changes in mixing height. IPCC (2001) projects global average temperatures increases of 1.5 to 4.5 K over the next century. Mickley et al. (2004) report increases in mixing heights of 100–240 m over the Midwest and Northeast using the IPCC A1B scenario, and Hogrefe et al. (2004) report increases in mixing height for the A2 scenario; Murazaki and Hess (2006), however, see no significant changes in mixing height for the A1 scenario. Breslow and Sailor (2002) predict decreased wind speeds of 1.0–3.2% for the USA in the next 50 years, though predicted changes in wind speeds have been very non-uniform spatially (Bogardi and Matyasovszky, 1996). Simulations using GCMs agree that cloud cover will decrease if temperature is increased, with 19 GCMs predicting an average cloud cover decrease of 2.1% for a

Table 3
Summary of expected meteorological changes and their effects on ozone concentrations

Meteorological parameter	Expected change of parameter	Sensitivity of $[O_3]$[a] mean (5%, 95%)	Expected effect on $[O_3]$[a] mean (5%, 95%)	Expected change in exceedances[b]
Temperature	+1.5 to +4.5 K[c]	+0.34 ppb K^{-1} (−0.23, +1.2)	+0.5 to +2 ppb (−1 ppb to +5 ppb)	+20 to +60%
Absolute humidity	+7 to +21%[d]	−0.025 ppb $\%^{-1}$ (−0.073, +0.034)	−0.5 to −0.2 ppb (−2 ppb to 0.7 ppb)	0%
Wind speed	−1.4 to −4.5%[e]	−0.005 ppb $\%^{-1}$ (−0.17, +0.14)	$+7\times10^{-3}$ to +0.02 ppb (−0.6 ppb to +0.8 ppb)	+2 to +6%
Mixing height	−1 layer to +1 layer[f]	−0.032 ppb $layer^{-1}$ (−1.0, +0.72)	−0.03 ppb to +0.03 ppb (−1 ppb to +1 ppb)	−8 to +20%
Cloud LWC and OD	−15 to +15%[f]	−0.003 ppb $\%^{-1}$ (−0.051, +0.035)	−0.05 to +0.05 ppb (−0.8 ppb to +0.8 ppb)	−2 to +2%
Cloudy area	−4.4 to −0.2%[g]	−0.001 ppb $\%^{-1}$ (-2.8×10^{-3}, $+2.8\times10^{-3}$)	$+1\times10^{-4}$ to $+3\times10^{-3}$ ppb (−0.01 ppb to +0.01 ppb)	0 to 1%
Precipitation rate	−20 to +20%[f]	+0.002 ppb $\%^{-1}$ (-5.3×10^{-6}, $+9.2\times10^{-3}$)	−0.04 ppb to +0.04 ppb (−0.2 ppb to +0.2 ppb)	−1 to +2%
Precipitating area	−10 to +10%[h]	+0.005 ppb $\%^{-1}$ (-3.1×10^{-6}, +0.018)	−0.07 to +0.07 ppb (−0.3 ppb to +0.3 ppb)	−2 to +2%

[a]The $[O_3]$ metric used is simulation-average daily maximum 8 h average $[O_3]$ in land grid cells.
[b]Land grid cells exceeding AQS at any point during simulation.
[c]IPCC, 2001.
[d]Based on IPCC temperature projections and constant 80% RH. http://ipcc-ddc.cru.uea.ac.uk/sres/scatter_plots/scatterplots_region.html.
[e]Breslow and Sailor, 2002.
[f]Especially speculative; included to enable intercomparison among all parameters.
[g]Cess et al., 1990.
[h]IPCC Data Distribution Centre.

4 K temperature increase (Cess et al., 1990). Leung and Gustafson (2005) predict changes in the number of summer days with precipitation decreases of 6–8 days per season in Texas, and increases of 5 days per season in the Midwest. Murazaki and Hess (2006), however, report no significant changes in precipitation. Other expected climate changes have been characterized less thoroughly.

Sensitivities of ozone to changes in absolute humidity were calculated using only positive humidity changes, while sensitivities to cloudy area were calculated using only negative cloudy area changes (due to the nonlinear overall responses to these parameters and given consensus regarding the sign of their future changes). Estimated future meteorological changes are average changes corresponding to doubled CO_2 concentrations for temperature and absolute humidity, 2050 projections for wind speed and precipitating area, and a 4 K sea surface temperature perturbation for cloudy area. The precipitating area change was inferred from predicted changes in total precipitation over the eastern USA (Leung and Gustafson, 2005). Changes in mixing height, cloud LWC and OD, and precipitation rate were chosen so that somewhat liberal estimates of the ozone sensitivity could be calculated and compared to the sensitivities to other parameters.

The mean, 5th and 95th percentile values for sensitivities of daily maximum 8 h average $[O_3]$ were included in the analysis to account for variability from location to location and day to day. The expected effects on this ozone metric were calculated by multiplying each of these three sensitivity values

with the expected range of changes in the meteorological parameter. Predicted changes in exceedances were calculated using simple regressions of changes in exceedances versus the imposed meteorological perturbation. Using the expected effects on the daily maximum 8 h average O_3 metric, temperature appears to have the strongest effect on ozone concentrations, with absolute humidity having a smaller but appreciable effect, and wind speed, mixing height, and cloud and precipitation changes having relatively small effects. The variability in the response to changes in wind speed, mixing height, and cloud LWC and OD appears to make the interactions between ozone and these parameters appreciable (~0.1–1 ppb), though sensitivities to the other meteorological parameters still appear rather small even with variability taken into account.

The response of the exceedance metric was somewhat different from the response of the 8 h average metric. Temperature again had the largest effect, while wind speed and mixing height had a smaller effect, and absolute humidity and cloud and precipitation changes had very small effects on exceedances. Especially interesting is the role of absolute humidity increases, which had an appreciable effect on the 8 h average ozone metric, but practically no effect on exceedances (Fig. 4(c) and Table 3). It appears that absolute humidity increases did not have enough of an effect on the 8 h average in areas near the ozone standard to push them above or below the standard (Fig. 8).

5. Conclusions

The results of this study indicate that there are important links between changes in summertime meteorology and ozone concentrations. Changes in temperature, absolute humidity, wind speed, mixing height, cloud liquid water content and optical depth, cloud extent, precipitation rate, and precipitation extent are all expected to lead to changes in ozone, though to varying degrees. Using both daily maximum 8 h average concentration and exceedances of the air-quality standard as metrics, ozone was most affected by temperature in this episode. The average daily maximum 8 h average ozone concentration over land grid cells increased with temperature by $0.34\,\mathrm{ppb\,K^{-1}}$. Additionally, temperature increases in the range predicted by IPCC (2001) led to increases in the area exceeding the air-quality standard of 20–60%, compared to base case conditions. Absolute humidity had a somewhat smaller but appreciable effect on the daily maximum 8 h average metric. These results are similar to Racherla and Adams (2006), in which a global modeling study predicts increases in water vapor as the dominant factor decreasing global ozone, mostly in remote areas, but predict summertime ozone increases in polluted areas. The increase in ozone with increased temperature, and the decreases in ozone with increased wind speed and mixing height are similar to the results of Baertsch-Ritter et al. (2004), though the two studies use different ozone metrics. These results are also consistent with Murazaki and Hess (2006) who see increases in ozone (attributed to increases in temperature and water vapor) in high-NO_x areas. Wind speed and mixing heights had appreciable effects on the exceedance metric. Taking into account the variability of responses in time and space, the response of the daily maximum 8 h average ozone concentration to changes in wind speed, mixing height, and cloud LWC and OD was also appreciable (~0.1–1 ppb), even though the average response to these parameters was rather small (<0.1 ppb).

Changes in climate can potentially increase concentrations of ozone, and air pollution episodes could potentially become more severe under a changed climate scenario, which could increase pollution-related health effects. Changes in other meteorological parameters may reduce the effect of temperature on ozone, and the change in ozone that results from a combination of meteorological changes does not necessarily equal the sum of the ozone changes that would result from individual meteorological perturbations. In the one combined-change case examined, the ozone change due to combined meteorological perturbations was less than the sum of the resulting changes from the individual meteorological perturbations.

In order to gain a more accurate understanding of how future climate will affect air quality, projections of future climate must predict accurately the meteorological factors that have important links to air quality. The large uncertainties in predictions of future climate currently make the task of beginning to predict future air quality rather difficult, though the most uncertain aspects of future climate, especially cloud cover and precipitation, appear to have rather minor effects on ozone concentrations. The uncertainties associated with changes in large-scale dynamics, convection, and stratosphere–troposphere exchange add to this complexity.

Additionally, the lack of predictions of expected changes in mixing heights further complicates the issue. For an actual prediction of future air quality, projections of future emissions of pollutants and precursors are also necessary.

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