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Sensitivity of $PM_{2.5}$ to climate in the Eastern US: a modeling case study

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Abstract. The individual effects of various meteorological parameters on $PM_{2.5}$ concentrations in the Eastern US are examined using the PMCAMx chemical transport model so that these effects and their relative magnitudes can be better understood. A suite of perturbations in temperature, wind speed, absolute humidity, mixing height, cloud cover, and precipitation are imposed individually on base case conditions corresponding to periods in July 2001 and January 2002 in order to determine the sensitivities of $PM_{2.5}$ concentrations and composition to these separate meteorological parameters. Temperature had a major effect on average $PM_{2.5}$ in January ($-170\,\mathrm{ng\,m^{-3}\,K^{-1}}$) due largely to the evaporation of ammonium nitrate and organic aerosol at higher temperatures; increases in sulfate production with increased temperature counteracted much of this decrease in July. Changes in mixing height also had major effects on $PM_{2.5}$ concentrations: $73\,\mathrm{ng\,m^{-3}}$ $(100\,\mathrm{m})^{-1}$ in January and $210\,\mathrm{ng\,m^{-3}}$ $(100\,\mathrm{m})^{-1}$ in July. Changes in wind speed (30 to $55\,\mathrm{ng\,m^{-3}\,\%^{-1}}$) and absolute humidity (15 to $20\,\mathrm{ng\,m^{-3}\,\%^{-1}}$) also had appreciable effects on average $PM_{2.5}$ concentrations. Precipitation changes had large impacts on parts of the domain (a consequence of the base case meteorology), with sensitivities to changing area of precipitation in July up to $100\,\mathrm{ng\,m^{-3}\,\%^{-1}}$. Perturbations in cloud cover had the smallest effects on average $PM_{2.5}$ concentrations. The changes in $PM_{2.5}$ concentrations resulting from changing all eight meteorological parameters simultaneously were approximately within 25% or so of the sum of the changes to the eight individual perturbations. The sensitivities of $PM_{2.5}$ concentrations to changes in these meteorological parameters indicate that changes in climate could potentially have important impacts on $PM_{2.5}$ concentrations.

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1 Introduction

High concentrations of particulate matter (PM), a major constituent of air pollution, have detrimental effects on human health (Godish, 2004). Particulate air pollution has been associated with increases in mortality (Schwartz et al., 1996) and can aggravate respiratory and cardiovascular diseases, damage lung tissue, and lead to premature death (Bernard et al., 2001). The health effects of particulate matter have been associated with both short- and long-term exposure (Kappos et al., 2004).

Concentrations of PM are strongly influenced by meteorology, but there has been little research on how concentrations depend on individual meteorological parameters (Elminir, 2005). PM is comprised of many different species, and meteorology can have complex effects on total PM concentrations due to its impacts on individual species. Aerosol sulfate concentrations depend on the temperature-dependent oxidation of SO_2 in both the gas and aqueous (cloud) phases (Seinfeld and Pandis, 2006). The concentrations of oxidants that react with SO_2 are also dependent on temperature and sunlight intensity (Sweet and Gatz, 1998). Concentrations of semi-volatile nitrate and organic aerosols are temperature and relative humidity dependent; they can also vary with the amount of oxidants present, which is linked to photolysis rates and, therefore, cloud cover. All species have wet deposition as a major sink, so precipitation is expected to have a significant effect on aerosol concentrations. Finally, mixing and dilution influence PM concentrations, so wind speed and mixing height are expected to have an impact as well.

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 PM concentrations; for example, global average temperatures are expected to rise 1.5 to 4.5 K over the next century

(IPCC, 2001). Predictions of wind speed changes in the United States vary depending on the area in question and on the model used. Bogardi and Matyasovszky (1996) predict spatially variable changes in wind speeds in Nebraska under a future climate. Breslow and Sailor (2002) predict wind speed decreases over the United States in the next 50 years. Absolute humidity (water vapor concentration) is generally expected to increase due to the higher saturation vapor pressure of water at higher temperatures (IPCC, 2001). Held and Soden (2000) point out that many models predict that future relative humidity will remain roughly constant with climate change. 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 decreases when temperature increases (Cess et al., 1990). GCM studies also predict minor changes in summer and annual mean precipitation over the eastern United States (Räisänen, 2005). Leung and Gustafson (2005), however, predict significant changes in the number of summer days with precipitation in the Eastern USA. Mickley et al. (2004) and Hogrefe at al. (2004) report increased mixing heights in future climates, though Murazaki and Hess (2006) predict no significant changes in mixing heights in a future climate.

While the response of ozone to changes in meteorology and climate has been examined (Hogrefe et al., 2004; Dawson et al., 2007; Baertsch-Ritter et al., 2004; Racherla and Adams, 2007; Johnson et al., 2001; Brasseur et al., 1998, 2006; Unger et al., 2006; Liao et al., 2006; Muraraki and Hess, 2006; Steiner et al., 2006), there has been relatively little work connecting aerosol concentrations and meteorology. The corresponding studies have generally been statistical observational studies (Elminir, 2005; Wise and Comrie, 2005; Triantafyllou et al., 2002), along with a small number of modeling studies (Unger et al., 2006; Liao et al., 2006). These studies illustrate the difficulty in deriving causal relationships between specific meteorological parameters and measured PM concentrations when the meteorological variables are strongly correlated with one another.

A few process modeling studies have also looked at the connections between meteorology and aerosol concentrations. The results of Mickley et al. (2004) suggest (using black carbon as a tracer) that a warmer future climate could increase the severity of summertime PM episodes in the Northeastern and Midwestern USA. Racherla and Adams (2007) predicted decreases in global burdens and lifetimes of fine PM using the IPCC A2 scenario, though global-average changes at the surface level were small and regional responses were mixed. Increases in aerosol sulfate over the eastern USA were also suggested by this work. In a study that includes both observation and process modeling components, Koch et al. (2003) observed a significant negative correlation between cloud cover and aerosol sulfate due to the longer lifetime of gas-phase-produced sulfate compared to aqueous-phase-produced sulfate. Aw and Kleeman (2003) calculated decreases in $PM_{2.5}$ concentrations due to temperature increases in a modeling study over southern California; this was due to decreases in semi-volatile aerosols, especially ammonium nitrate. The same study predicted increases in non-volatile PM concentrations with temperature. Using a box model, Sheehan and Bowman (2001) predicted an increase in secondary organic aerosol (SOA) yields of 20–150% for a 10 K decrease in temperature due to the temperature-dependent partitioning of the aerosol.

Determining how PM concentrations change as climate changes is an important step toward estimating future air quality. This may allow air quality 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. Observational studies have generally focused on small areas (e.g. one city) and have difficulties in separating the effects of different atmospheric variables; the response of PM concentrations over large regions has been the focus of little research. Additionally few studies have calculated sensitivities of PM concentrations to a comprehensive suite of individual meteorological parameters. The goal of this study is to determine how PM concentrations over the eastern United States respond to changes in meteorological 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 and their relative importance can be better understood.

2 Model description and methods

The PMCAMx model (Gaydos et al., 2007) was the modeling tool used in this study. This model uses the framework of CAMx v. 4.02 (Environ, 2002) 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 RADM method of Chang et al. (1987). Ten aerosol size sections were used, spanning the diameter range from 40 nm to 40 μm. Inorganic aerosol formation was simulated using the bulk equilibrium approach of Capaldo et al. (2000), while aqueous chemistry was modeled using the variable size resolution model (VSRM) of Fahey and Pandis (2003). Equilibrium between the gas and aerosol phases for organics was calculated using the Secondary Organic Aerosol Model (SOAM II) of Strader et al. (1999) as implemented by Koo et al. (2004). In this model, primary organic aerosol (POA) is treated as nonvolatile; SOA is the only organic aerosol component that is treated as semi-volatile. Wet scavenging of aerosols is simulated following the method outlined by Environ (2002) and Seinfeld and Pandis (2006) based on a linear relationship between precipitation rate and scavenging coefficient.

Two periods were modeled using PMCAMx so that both winter and summer could be examined: 12–21 July 2001 and 6–15 January 2002. The first three days, a rather standard spin-up period for regional models in this domain (Hogrefe et al., 2004; Karydis et al., 2007), from each period were used as model initialization days and are excluded from the analysis. The modeling domain was the eastern half of the USA (Fig. 1), with a 36×36 km resolution grid. In the vertical direction, 14 layers in July and 16 layers in January were used, extending from the surface to an altitude of approximately 6 km in July and 14 km in January. Inputs to the model included meteorological conditions, land use data, emissions, and initial and boundary conditions of species. The emissions inventory used was the Midwest Regional Planning Organization's Base E inventory (LADCO, 2003), including BIOME3 biogenics (Wilkinson and Janssen, 2001), which included isoprene and a lumped monoterpene species. Both biogenic species participated in ozone chemistry, however only the monoterpene was included in the SOA mechanism. The emissions are described in more detail in Gaydos et al. (2007) and Karydis et al. (2007). Biogenic emissions were based on the base case meteorology and did not change with perturbations in meteorology. As a result the biogenic VOC emissions were the same in all simulations. The meteorological input into the model was generated by MM5 using assimilated meteorological data. PMCAMx performance for the periods modeled in this study has been evaluated by Karydis et al. (2007) and Gaydos et al. (2007) and was found to vary from fair to excellent depending on the species, period, and area. The most accurate model performance was for ammonium, sulfate, organics, and total $PM_{2.5}$.

In addition to a base case scenario for each of the two months, a suite of sensitivity simulations were run in which individual meteorological parameters were perturbed to varying degrees (Table 1). These perturbations are the same as in Dawson et al. (2007). The perturbed variables include temperature, wind speed, absolute humidity, mixing height, cloud liquid water content (LWC) and optical depth (OD), cloudy area, precipitation rate, and precipitating area. Perturbing the meteorological variables individually allows them to be studied in isolation of one another and to be compared to determine their relative impacts on $PM_{2.5}$ concentrations. Vertical wind speeds were calculated from the perturbed horizontal wind speeds to ensure mass conservation. Except for cloud, precipitation, and mixing height changes, perturbations were imposed uniformly in space and time on the modeling domain. Sensitivity to mixing height was tested by simulations in which the mixing height, as determined from vertical diffusivities using 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 or below the original mixing height. Mixing height changes were implemented only when a definitive mixing height could be inferred from a polynomial relation between vertical diffusivity and altitude, as described by O'Brien (1970) (approximately two-thirds of grid cell time steps in July and half of grid cell time steps in January). This corresponded to average changes in mixing height of approximately 150 m. The area of cloud cover and precipitation were changed by growing (or shrinking) existing cloudy or precipitating areas into randomly selected adjacent cells. Cloud cover and precipitation were changed independently of one another so that their effects could be separated. A list of model processes affected by these meteorological changes is also given in Table 1. Emissions of all pollutants, biogenic and anthropogenic, were kept constant in all tests.

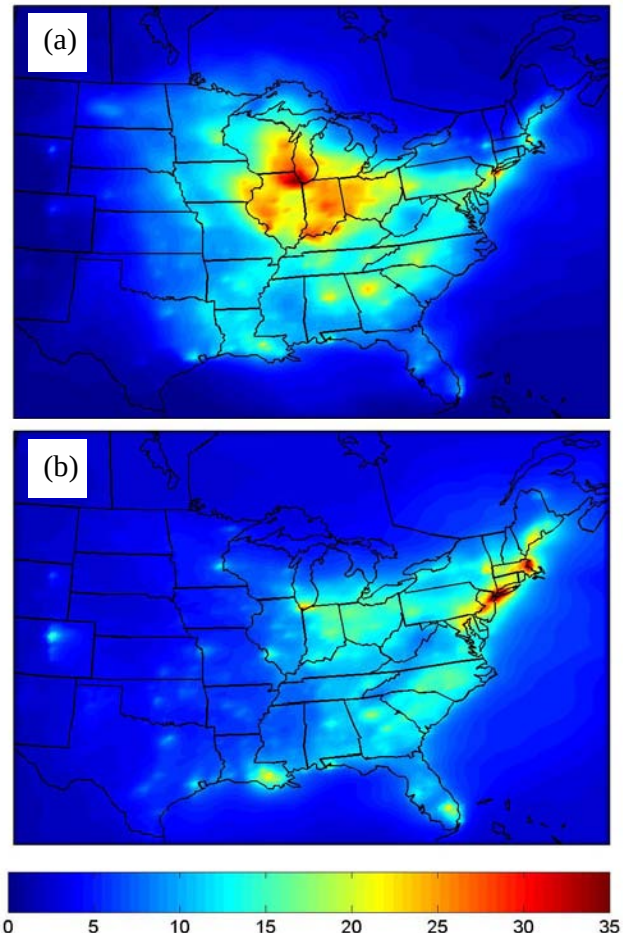

Fig. 1. Average $PM_{2.5}$ concentrations ($\mu g\,m^{-3}$) for the modeled periods of **(a)** July 2001 and **(b)** January 2002.

The model used a fixed concentration of each $PM_{2.5}$ species as boundary conditions. The fixed concentrations indicate an assumption that there is no change in the long range transport of pollution to the US. The elemental carbon boundary condition was $0.1\,\mu g\,m^{-3}$ in both months. In January, the following concentrations were used for boundary conditions: OM, $0.5\,\mu g\,m^{-3}$; sulfate, $0.7\,\mu g\,m^{-3}$; nitrate, $0.3\,\mu g\,m^{-3}$; ammonium, $0.35\,\mu g\,m^{-3}$. July simulations used a different set of boundary concentrations: OM,

Table 1. Meteorological perturbations imposed in this study and adjustments imposed in combined-change simulation.

Meteorological Parameter	Changes in Values Examined	Combined-Change Adjustment	Directly Affected in Simulation
Temperature	+0.5, 1.0, 1.5, 2.5, 4.0, 5.0 K	+2.5 K	Reaction rates, aerosol thermodynamics
Wind speed	±5, 10%	+5%	Vertical velocity/dilution/entrainment, advection, diffusion coefficients, dry deposition resistance
Absolute humidity	±5, 10, 20%	+10%	Reaction rates with H_2O, aerosol thermodynamics
Mixing height	± One model layer	+ One model layer	Vertical diffusivities in layers near mixing height
Cloud LWC & OD	±5, 10, 20%	+10%	Radiation transmittance of clouds, aqueous chemistry
Area of cloud cover			Radiation transmittance of clouds, aqueous chemistry
January	−5.9, −4.7, −2.2, +2.3, +4.7, +6.0%	+6.0%	
July	−3.9, −2.5, +2.2, +4.1%	+4.1%	
Precipitation intensity	±5, 10, 20%	+10%	Wet deposition
Area of precipitation cover			Wet deposition
January	−7.1 −4.8, −2.4, +2.1, +4.9, +7.2%	+4.9%	
July	−4.9, −2.3, +2.4, +4.7%	+4.7%	

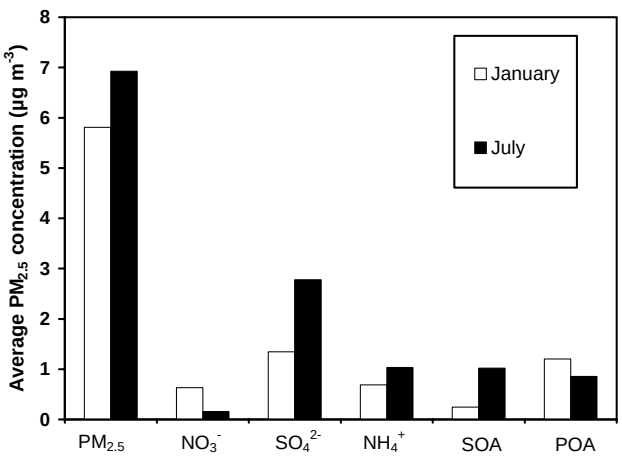

Fig. 2. Simulation-long land cell average concentrations ($\mu g\,m^{-3}$) of total $PM_{2.5}$ and $PM_{2.5}$ nitrate, sulfate, ammonium, secondary organic aerosol (SOA), and primary organic aerosol (POA) in January and July base case simulations.

0.8 $\mu g\,m^{-3}$; sulfate, 0.9 $\mu g\,m^{-3}$; nitrate, 0.1 $\mu g\,m^{-3}$; ammonium, 0.37 $\mu g\,m^{-3}$ (Karydis et al., 2007). Boundary conditions of aerosol species were split equally among the six size bins that comprised $PM_{2.5}$.

Simulation-averaged ground-level concentrations of total $PM_{2.5}$ as well as $PM_{2.5}$ ammonium, sulfate, nitrate, and organics are the species examined in this analysis. The base case predicted concentrations of total $PM_{2.5}$ for both months are shown in Fig. 1, and the land-cell average concentrations for the species investigated for both months are shown in Fig. 2. Average ground-level concentrations of total $PM_{2.5}$ were 5.8 $\mu g\,m^{-3}$ in January and 6.9 $\mu g\,m^{-3}$ in July. In January, the highest simulation-average concentration was 40 $\mu g\,m^{-3}$ in the New York area, due largely to primary organics. In July, the highest average concentrations (up to 44 $\mu g\,m^{-3}$) were in the Midwest, especially the Chicago area; this was largely due to high sulfate concentrations. Nitrate concentrations were relatively high during January (Fig. 2), while sulfate concentrations were high during July. SOA concentrations were higher during July, while POA concentrations changed little with season. SOA comprised 54% of total OM in July, though its contribution was reduced to 17% in January.

3 Results and discussion

3.1 Temperature

The response of $PM_{2.5}$ concentrations to temperature was largely the result of competing changes in sulfate and nitrate concentrations with a smaller role played by organics. In January, average $PM_{2.5}$ concentrations over land grid cells decreased by 170 $ng\,m^{-3}\,K^{-1}$ (2.9% K^{-1}), while average concentrations in July decreased by 16 $ng\,m^{-3}\,K^{-1}$ (0.23% K^{-1}). In January, when nitrate concentrations were high, the response of total $PM_{2.5}$ was stronger than in July, when nitrate concentrations were low. Total $PM_{2.5}$

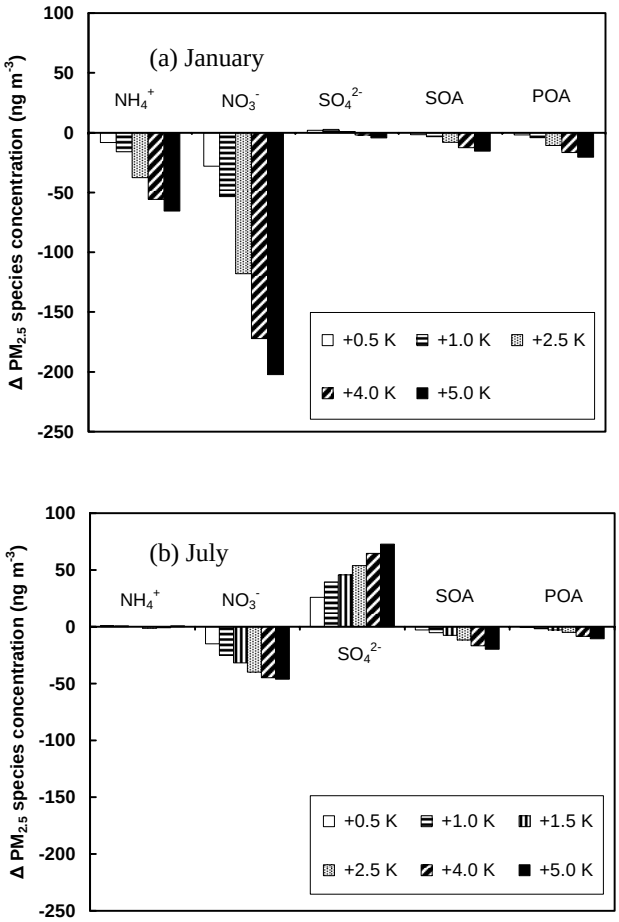

Fig. 3. Average differences in simulation-averaged ground-level $PM_{2.5}$ species concentrations in **(a)** January and **(b)** July for perturbed temperature cases.

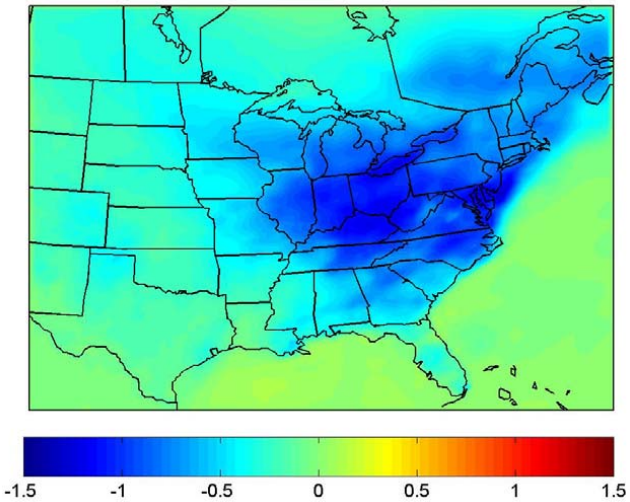

Fig. 4. Average changes in total $PM_{2.5}$ ($\mu g\,m^{-3}$) January for a 2.5 K temperature increase.

concentrations decreased by $2.9\%\,K^{-1}$ in January and by $0.23\%\,K^{-1}$ in July, resulting in an average reduction of $1.6\%\,K^{-1}$.

In July, temperature increases led to increases in sulfate concentrations and simultaneous decreases in nitrate and organic concentrations. In January, however, average nitrate and organic concentrations still decreased as temperature was increased, but average sulfate concentrations were rather insensitive to temperature changes. Changes in ammonium were a consequence of the changes in nitrate and sulfate. These average changes are shown in Fig. 3. Average $PM_{2.5}$ nitrate concentrations over land cells decreased by $120\,ng\,m^{-3}\,K^{-1}$ ($19\%\,K^{-1}$) and $26\,ng\,m^{-3}\,K^{-1}$ ($17\%\,K^{-1}$) in January and July respectively. This is mostly due to the volatilization of ammonium nitrate, which partitions to the gas phase at higher temperatures (Seinfeld and Pandis, 2006). Average $PM_{2.5}$ sulfate concentrations over land cells increased by $1.6\,ng\,m^{-3}\,K^{-1}$ ($0.12\%\,K^{-1}$) and $34\,ng\,m^{-3}\,K^{-1}$ ($1.3\%\,K^{-1}$) in January and July respectively. This link between sulfate concentrations and temperature is due to the increased rate of oxidation of SO_2 at higher temperature, caused by temperature-dependent rate constants and higher concentrations of oxidants. Average concentrations over land grid cells of total organic $PM_{2.5}$ decreased by $13\,ng\,m^{-3}\,K^{-1}$ ($0.90\%\,K^{-1}$) and $14\,ng\,m^{-3}\,K^{-1}$ ($0.75\%\,K^{-1}$) in January and July respectively. This is the net effect of increased gas-phase partitioning and faster gas-to-particle conversion at higher temperatures (Strader et al., 1999). In January, SOA accounted for 17% of organic mass over land cells and 42% of the response of organic $PM_{2.5}$ mass to a 2.5 K temperature increase; in July, SOA accounted for 54% of organic mass and 59% of the corresponding temperature response. The stronger effect of temperature on nitrate than on organics was also suggested by Aw and Kleeman (2003). Average nitrate concentrations decreased by $15\%\,K^{-1}$ on average, organic concentrations decreased by $1.0\%\,K^{-1}$, and sulfate concentrations increased by $0.12\%\,K^{-1}$ in January and $4.2\%\,K^{-1}$ in July.

The sensitivities to temperature changes were nonuniform throughout the domain. In some places, the response of total $PM_{2.5}$ was dominated by decreases in nitrate, while in other places increases in sulfate were dominant (Figs. 4 and 5). In January, the response of total $PM_{2.5}$ (Fig. 4) was very similar to that of $PM_{2.5}$ nitrate. The response in January was rather homogeneous throughout the domain (Fig. 4). In July, the response of total $PM_{2.5}$ (Fig. 5a) reflected the combined responses of nitrate (Fig. 5b) and sulfate (Fig. 5c). The increases in sulfate and decreases in nitrate offset each other to lead to a small response in average total $PM_{2.5}$. The response of $PM_{2.5}$ concentrations in July was much more variable spatially than the response in January. Changes in organics tended to be rather small (averaging $-14\,ng\,m^{-3}\,K^{-1}$ with a maximum sensitivity of $-100\,ng\,m^{-3}\,K^{-1}$ in July), and changes in $PM_{2.5}$ ammonium appear to have been largely influenced by the changes in nitrate and sulfate.

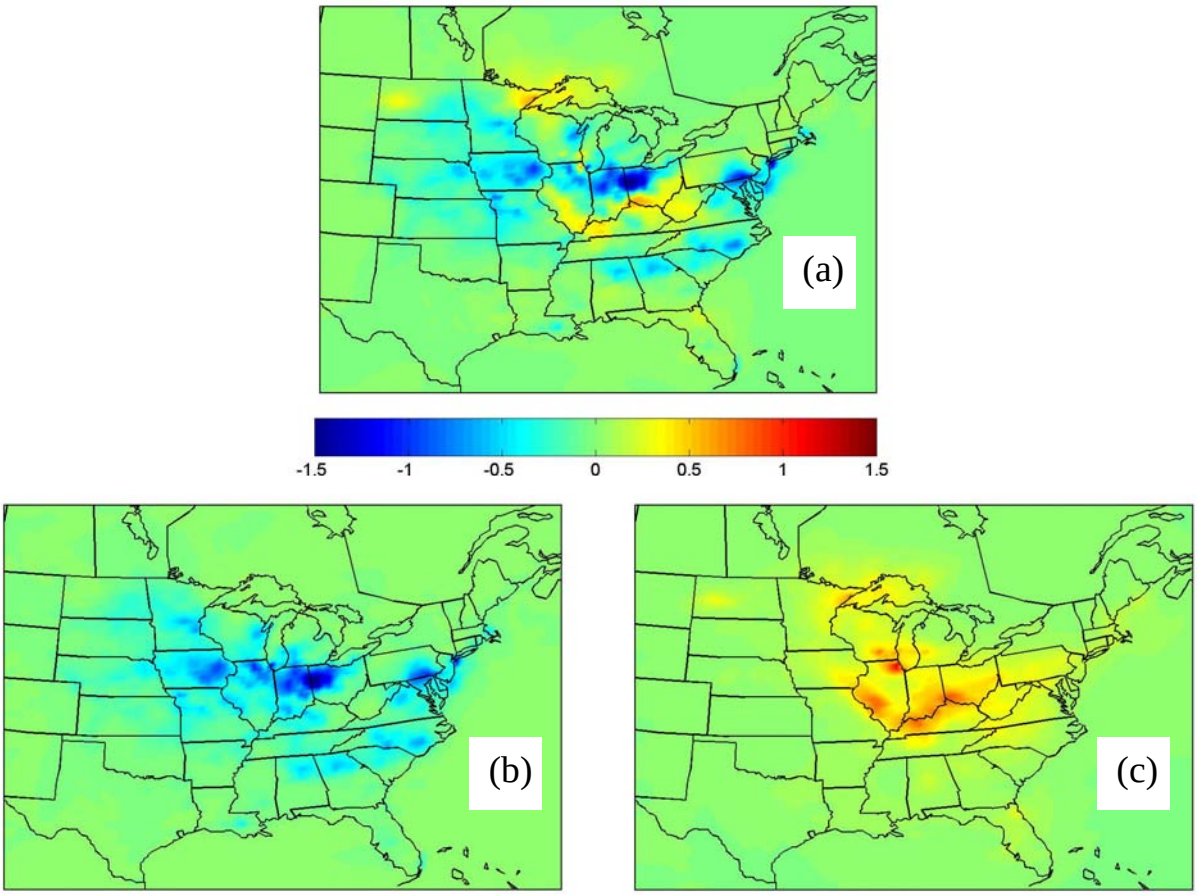

Fig. 5. Average changes in **(a)** total $PM_{2.5}$ ($\mu g\,m^{-3}$), **(b)** $PM_{2.5}$ nitrate, and **(c)** $PM_{2.5}$ sulfate in July for a 2.5 K temperature increase.

Table 2. Simulation-average sensitivities to meteorological perturbations in Pittsburgh in Atlanta.

	January		July		Units
	Pittsburgh	Atlanta	Pittsburgh	Atlanta	
Temperature	−2.2	−2.1	0.26	−0.68	$\%\,K^{-1}$
Wind speed	−0.83	−0.71	−0.73	−0.93	$\%\,\%^{-1}$
Absolute humidity	0.12	0.15	0.06	0.22	$\%\,\%^{-1}$
Mixing height	−0.9	−1.1	−1.2	−1.5	$\%\,(100\,m)^{-1}$
LWC and OD	−0.02	−0.02	−0.002	−0.003	$\%\,\%^{-1}$
Cloudy area	−0.1[a]	−0.04	−0.09	−0.2	$\%\,\%^{-1}$
Precipitation rate	−0.01	−0.01	−0.1	−0.2	$\%\,\%^{-1}$
Precipitation area	−0.001	−0.001	−0.1	−0.3	$\%\,\%^{-1}$

[a] For an increase in cloudy area. Smaller sensitivity for decrease in cloudy area (Sect. 3.6).

In January, simulation-averaged concentrations of total $PM_{2.5}$ decreased by 300 $ng\,m^{-3}\,K^{-1}$ (−2.1% K^{-1}) in Atlanta and 400 $ng\,m^{-3}\,K^{-1}$ (−2.2% K^{-1}) in Pittsburgh (Table 2). In contrast, July concentrations of $PM_{2.5}$ decreased by 150 $ng\,m^{-3}\,K^{-1}$ (−0.68% K^{-1}) in Atlanta and increased by 60 $ng\,m^{-3}\,K^{-1}$ (+0.26% K^{-1}) in Pittsburgh. The January responses in both cities were dominated by decreases in nitrate, while the July responses were the results of the combined responses of nitrate, sulfate, and organics. Western Ohio and the Great Lakes region, where nitrate concentrations were relatively high in both January and July, experienced the largest decreases with increased temperature due

to nitrate decreases, while the Ohio River Valley experienced the largest increases in July due to increases in sulfate. The response to a temperature increase in Chicago was dominated by increases in sulfate in July and decreases in nitrate in January.

3.2 Wind speed

Wind speed changes affected all species that comprised $PM_{2.5}$, with increases in wind speed generally leading to decreases in $PM_{2.5}$ concentrations, and decreases in wind speed generally leading to increases in $PM_{2.5}$. The simulation-long average $PM_{2.5}$ concentration over land grid cells decreased with increasing wind speed by $30\,ng\,m^{-3}\,\%^{-1}$ ($0.56\%\,\%^{-1}$) and $50\,ng\,m^{-3}\,\%^{-1}$ ($0.77\%\,\%^{-1}$) in January and July respectively. Changes in concentrations were greatest in the populated and polluted areas of the domain and smaller (or nearly zero) in more remote areas. The largest decrease in concentrations in January was in the New York area ($270\,ng\,m^{-3}\,\%^{-1}$ or $0.68\%\,\%^{-1}$), while the largest concentration decrease in July was near Chicago ($340\,ng\,m^{-3}\,\%^{-1}$ or $0.77\%\,\%^{-1}$). Concentrations in both Atlanta and Pittsburgh also decreased with increased wind speed (Table 2). These results are consistent with the observed association between high PM concentrations and stagnation (and, therefore, low wind speed) (Triantafyllou et al., 2002).

The above changes in concentrations are largely due to changes in advection and dispersion with wind speed, with changes in dry deposition playing a relatively small role. Because westerly winds are most common over the Eastern US, increased wind speeds carry additional PM out to the ocean. The resultant absolute changes in $PM_{2.5}$ concentrations appear to be minor in areas with low PM concentrations, but appreciable in more polluted areas. The relative sensitivities were roughly uniform, between -0.5 and $-0.9\%\,\%^{-1}$, indicating an important impact of changes in wind speed on $PM_{2.5}$ concentrations.

3.3 Absolute humidity

Changes in absolute humidity had the largest effects on concentrations of ammonium nitrate aerosol with concentrations increasing with increased absolute humidity (Fig. 6). This effect was somewhat stronger during the summer, when water vapor, nitric acid, and ammonia concentrations are highest.

The simulation-long average $PM_{2.5}$ concentration over land grid cells increased with water vapor concentration by $14\,ng\,m^{-3}\,\%^{-1}$ ($0.24\%\,\%^{-1}$) and $20\,ng\,m^{-3}\,\%^{-1}$ ($0.29\%\,\%^{-1}$) in January and July respectively, while nitrate concentrations changed by $11\,ng\,m^{-3}\,\%^{-1}$ ($1.7\%\,\%^{-1}$) and $23\,ng\,m^{-3}\,\%^{-1}$ ($15\%\,\%^{-1}$) respectively. Changes in average concentrations for a 10% increase in water vapor are shown in Fig. 7. Increases in humidity shift the equilibrium of the ammonia-nitric acid system toward the aerosol phase, resulting in higher concentrations of ammonium nitrate aerosol

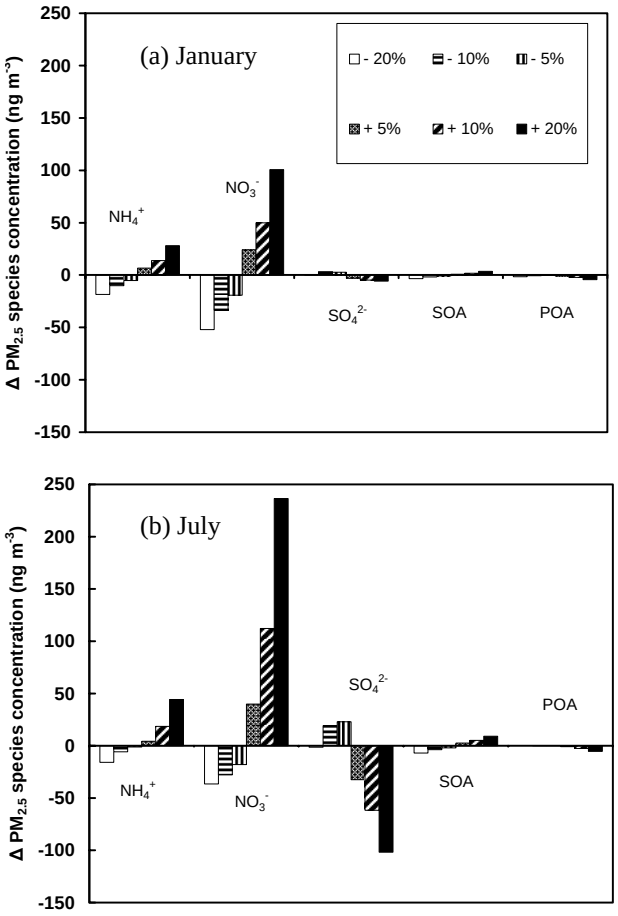

Fig. 6. Changes in simulation-long ground-level average $PM_{2.5}$ species concentrations in **(a)** January and **(b)** July perturbed absolute humidity simulations.

(Seinfeld and Pandis, 2006). Changes in sulfate aerosol were relatively small in summer (roughly half the changes in average nitrate) and practically negligible in winter, and changes in organics were negligible in both seasons. Ammonium concentrations appear to have been influenced by the changes in nitrate and, to a lesser extent, sulfate. The spatial distribution of changes of average total $PM_{2.5}$ strongly resembled the changes in $PM_{2.5}$ nitrate. The areas of increased total $PM_{2.5}$ (Figs. 7a and b) corresponded to the areas of increased $PM_{2.5}$ nitrate.

Changes in nitrate accounted for most of these total changes in Atlanta in Pittsburgh (Table 2); other species changed little compared to nitrate aerosol. These changes and the changes over the entire domain indicate that the effects of absolute humidity on $PM_{2.5}$ concentrations are potentially important, especially the effect on $PM_{2.5}$ nitrate.

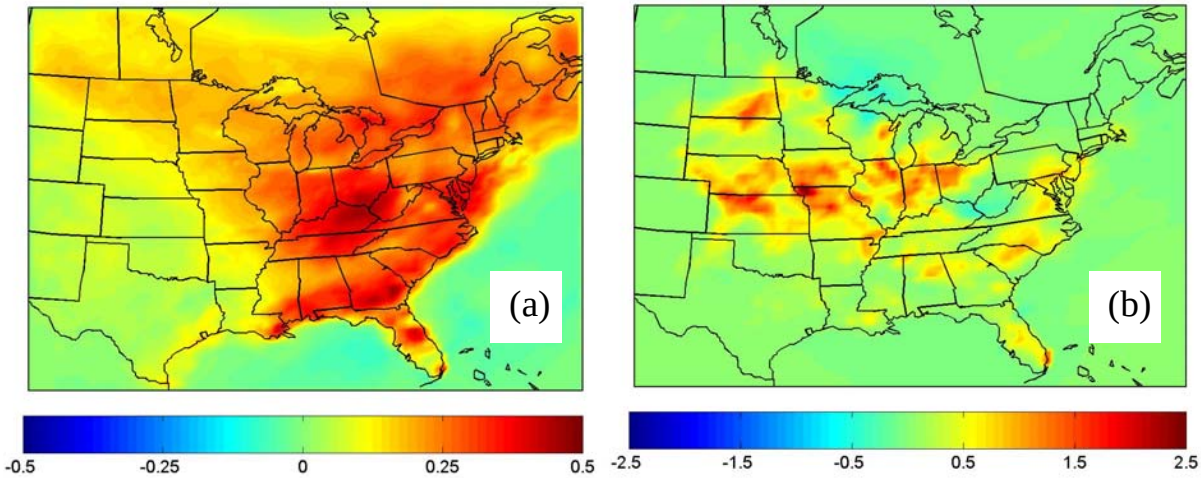

Fig. 7. Average changes in total $PM_{2.5}$ ($\mu g\,m^{-3}$) in **(a)** January and **(b)** July for a 10% increase in absolute humidity.

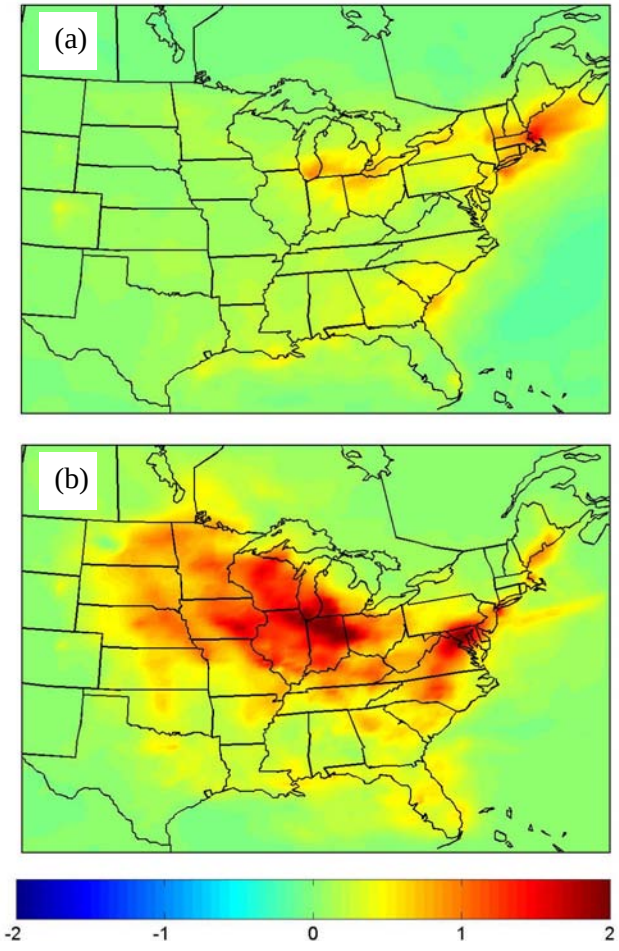

Fig. 8. Average changes in $PM_{2.5}$ concentrations ($\mu g\,m^{-3}$) due to a one-layer decrease (approximately 150 m) in mixing height in **(a)** January and **(b)** July.

3.4 Mixing height

Changes in mixing height had effects on all aerosol species. As expected, increases in mixing height led to decreases in $PM_{2.5}$ concentrations. Species were affected roughly in proportion to their relative concentrations, indicating that the mixing height effect is a simple dilution effect that does not induce major chemical feedbacks. In January and July, the average land cell $PM_{2.5}$ concentration decreased by $73\,ng\,m^{-3}$ $(100\,m)^{-1}$ (-1.3% $(100\,m)^{-1}$) and $210\,ng\,m^{-3}$ $(100\,m)^{-1}$ (-3.0% $(100\,m)^{-1}$) respectively. The difference between seasons is mainly due to lower mixing heights in July during the period modeled. The simulation-average base-case mixing height was 620 m in January and 420 m in July, the lower mixing height in July being a consequence of the periods selected. Generally, mixing heights tend to be lower in winter than in summer, meaning that changes in mixing height would affect winter concentrations more strongly than summer concentrations.

The simulation-averaged changes in $PM_{2.5}$ due to an increase in mixing height are shown in Fig. 8. The effect of mixing height on $PM_{2.5}$ concentrations in Atlanta and Pittsburgh was significant in both seasons (Table 2). The effect of mixing height on $PM_{2.5}$ concentrations, therefore, appears to be rather important, especially in polluted areas.

3.5 Cloud liquid water content and optical depth

Neither total $PM_{2.5}$ concentrations nor any aerosol species showed a strong sensitivity to changes in cloud LWC and OD (at constant cloudy area). Base case cloud cover and rain are shown in Fig. 9. In January and July, the land-cell average $PM_{2.5}$ concentration decreased with increased LWC and OD by $0.9\,ng\,m^{-3}\,\%^{-1}$ and $1.7\,ng\,m^{-3}\,\%^{-1}$ ($-0.02\%\,\%^{-1}$ for both seasons) respectively. The average sensitivities of all species during both months were less than $1\,ng\,m^{-3}\,\%^{-1}$.

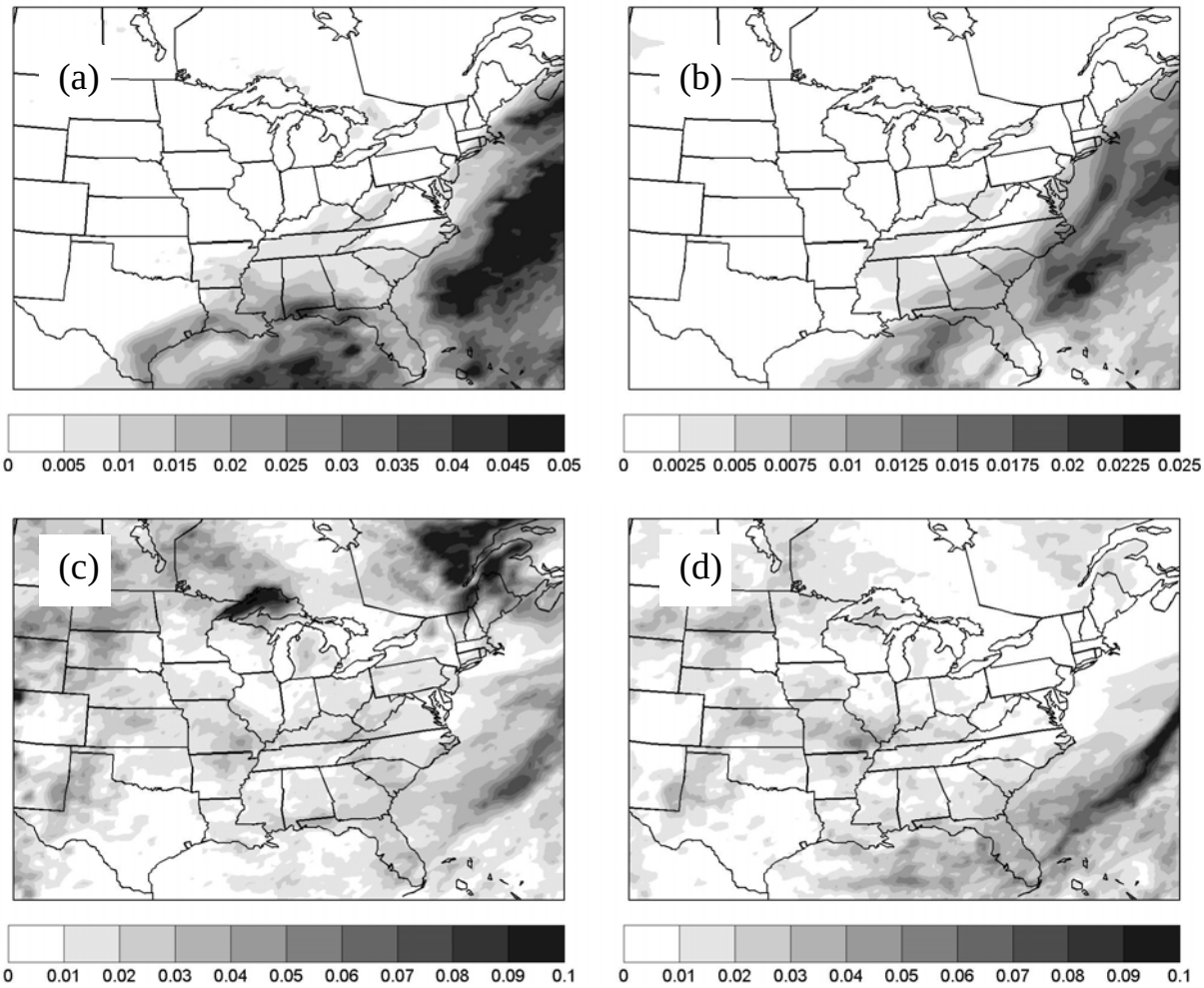

Fig. 9. Column- and simulation-averaged base case **(a)** January cloud water content ($g\,m^{-3}$), **(b)** January precipitation water content ($g\,m^{-3}$), **(c)** July cloud water content ($g\,m^{-3}$), and **(d)** July precipitation water content ($g\,m^{-3}$).

Pandis and Seinfeld (1989) calculated a relatively small change in total aqueous sulfate for an increase in LWC inside a single cloud. The net effect of these small changes in sulfate chemistry during cloudy periods several hundred meters aloft is a minor change in average $PM_{2.5}$ at ground level.

Average concentrations of total $PM_{2.5}$ in both Atlanta and Pittsburgh changed rather little with cloud LWC and OD (Table 2). The largest sensitivity of total $PM_{2.5}$ concentrations in July was $-70\,ng\,m^{-3}\,\%^{-1}$ ($-0.28\%\,\%^{-1}$) near St. Louis, and the largest sensitivity in January was $-50\,ng\,m^{-3}\,\%^{-1}$ ($-0.13\%\,\%^{-1}$) near Boston. Even these extreme values are rather small, indicating that the effects of cloud LWC and OD (at fixed cloudy area) on $PM_{2.5}$ concentrations are of minor importance. The location-specific responses, especially for the cloud and rain parameters, are largely a consequence of the period modeled and the relatively short duration of the study. These differences between location-specific responses do not necessarily mean that one location is inherently more sensitive to changes in clouds and precipitation, but they do give an estimate of the range of sensitivities.

3.6 Cloudy area

The influence of cloudy area on $PM_{2.5}$ concentrations varied by season and location, and all simulation-average changes, both domain-wide and in specific locations, were rather small. The mechanisms by which changes in cloudy area affect $PM_{2.5}$ concentrations is essentially the same as the mechanism by which cloud LWC and OD affect concentrations. In both January and July, increases in cloudy area led to decreases in simulation-averaged $PM_{2.5}$ over land grid cells. This average decrease was $2\,ng\,m^{-3}\,\%^{-1}$ ($-0.03\%\,\%^{-1}$) in January and $14\,ng\,m^{-3}\,\%^{-1}$ ($-0.2\%\,\%^{-1}$) in July. The differences in simulation-average ground-level concentrations of major $PM_{2.5}$ species due to changes

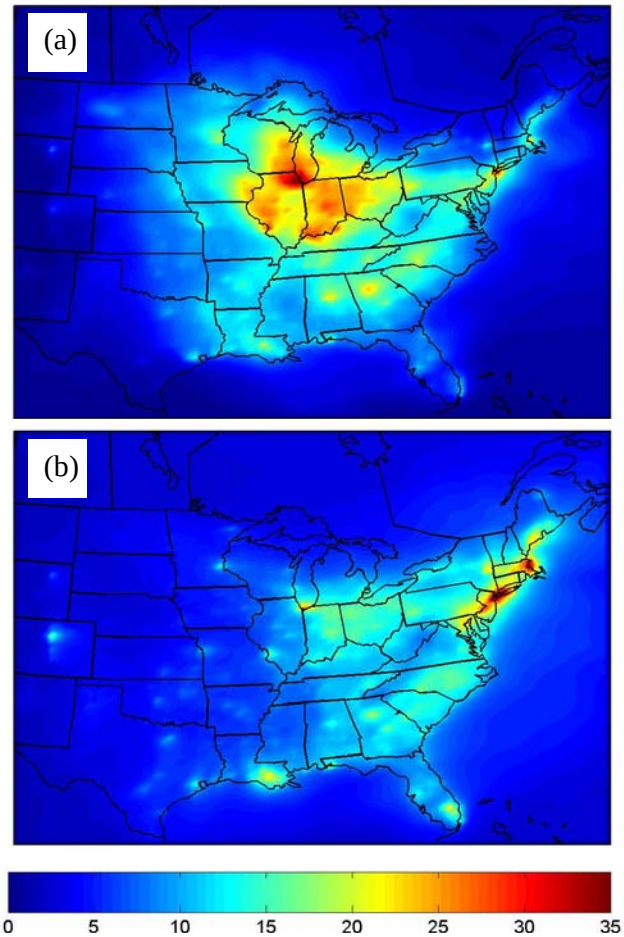

Fig. 10. Changes in simulation-long ground-level average concentration of major $PM_{2.5}$ species with changing cloudy area in **(a)** January and **(b)** July.

in cloudy area are shown in Fig. 10. In January, average nitrate ($-2\,ng\,m^{-3}\,\%^{-1}$ or $-0.3\%\,\%^{-1}$) and organics ($-1\,ng\,m^{-3}\,\%^{-1}$ or $-0.1\%\,\%^{-1}$) decreased with increased cloud cover, while average sulfate increased ($2\,ng\,m^{-3}\,\%^{-1}$ or $0.1\%\,\%^{-1}$). In July, all species decreased with increased cloud cover, with both sulfate and organics decreasing by $5\,ng\,m^{-3}\,\%^{-1}$ ($-0.2\%\,\%^{-1}$ and $-0.3\%\,\%^{-1}$, respectively). The difference between seasons in the sulfate response is due to the greater relative importance of aqueous sulfate production in January than in July.

The responses in Pittsburgh and Atlanta $PM_{2.5}$ concentrations to changes in cloud cover were mixed and rather small (Table 2). In January, the Pittsburgh $PM_{2.5}$ concentration increased by $0.3\,ng\,m^{-3}\,\%^{-1}$ ($-0.002\%\,\%^{-1}$) for the 5.9% cloud cover decrease, and decreased by $20\,ng\,m^{-3}\,\%^{-1}$ ($-0.1\%\,\%^{-1}$) for a 6% cloud cover increase. January $PM_{2.5}$ concentrations in Atlanta, however, were affected little by either a 5.9% cloud cover decrease ($-7\,ng\,m^{-3}\,\%^{-1}$ or $-0.05\%\,\%^{-1}$) or a 6.0% cloud cover increase ($5\,ng\,m^{-3}\,\%^{-1}$ or $-0.03\%\,\%^{-1}$). In both cities in January, average nitrate and ammonium decreased as cloud cover was increased. $PM_{2.5}$ concentration in July decreased with increased cloud cover by $20\,ng\,m^{-3}\,\%^{-1}$ ($-0.09\%\,\%^{-1}$) in Pittsburgh and $50\,ng\,m^{-3}\,\%^{-1}$ ($-0.2\%\,\%^{-1}$) in Atlanta. In both cities, the July sensitivity to cloud cover changes was dominated by changes in sulfate, which decreased as cloud cover was increased. The changes in $PM_{2.5}$ resulting from cloud cover changes were rather small, and it appears that they are of secondary importance to $PM_{2.5}$ concentrations.

3.7 Precipitation rate

Changes in the rate of precipitation affected $PM_{2.5}$ concentrations more strongly in July than in January. Changes in simulation-average $PM_{2.5}$ for a 10% decrease in precipitation rate in July are shown in Fig. 11. Sensitivities in much of the Midwest and Southeast were between 0.3 and $0.5\%\,\%^{-1}$. Sensitivities larger than $0.3\%\,\%^{-1}$ covered a large portion of the domain. The changes in $PM_{2.5}$ resulted even in areas with little or no base-case precipitation (Fig. 9d), such as northern Indiana (Fig. 11b), indicating that changes in precipitation in upwind areas affected $PM_{2.5}$ concentrations in downwind areas.

In both Pittsburgh and Atlanta, the sensitivity of total $PM_{2.5}$ to changes in precipitation rate was over an order of magnitude larger in July than in January (Table 2). This is due in part to the differences in precipitation between the two months (Fig. 9) causing a percentage adjustment in precipitation to represent a different amount of rainfall in each month. There is also an effect of the differences in the type of precipitation between the two seasons. In the Eastern USA, large-scale precipitation tends to dominate in winter, while convective precipitation is important in summer. Since convective storms tend to be short-lived, changes in precipitation rate help them more fully wash out aerosols. The overall wet removal by large-scale systems, which generally have a longer lifetime, is less sensitive to the precipitation rate since there is more time to fully wash out aerosols from the air. Areas with heavy base-case precipitation, such as the southeastern section of the domain, southern Missouri, Kansas, and the Dakotas (Fig. 9d), had small sensitivities to precipitation changes (Fig. 11). The areas with the largest sensitivities (Fig. 11) were the areas with smaller amounts of base-case precipitation, such as the Great Lakes region and West Virginia (Fig. 9d).

The sensitivities of $PM_{2.5}$ concentrations to precipitation rate (with fixed area of precipitation) were rather large over much of the domain. Overall land-average sensitivities were comparable to those of other meteorological parameters: $-0.02\%\,\%^{-1}$ in January and $-0.2\%\,\%^{-1}$ in July. The small sensitivity in January is partly a consequence of the lack of modeled precipitation in areas such as the Great Lakes region and New England (Fig. 9b) that tend to receive substantial precipitation in January. Precipitation in July compared more

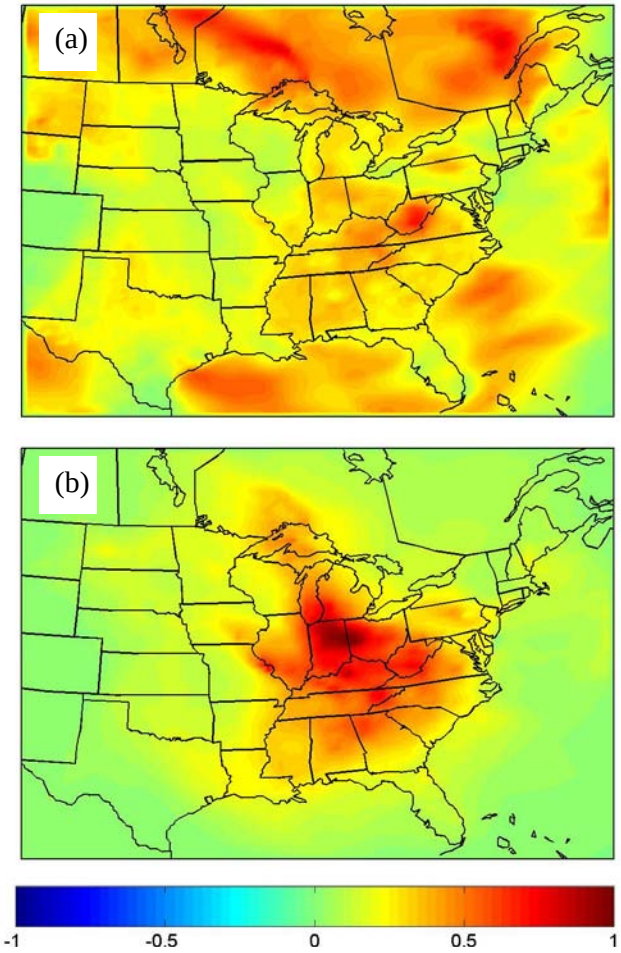

Fig. 11. Percent changes in simulation-average $PM_{2.5}$ concentrations divided by percent decrease in precipitation rate ($\%\,\%^{-1}$) **(a)**, and changes in simulation-average $PM_{2.5}$ concentration ($\mu g\,m^{-3}$) **(b)**, both calculated for a 10% reduction in precipitation rate in July.

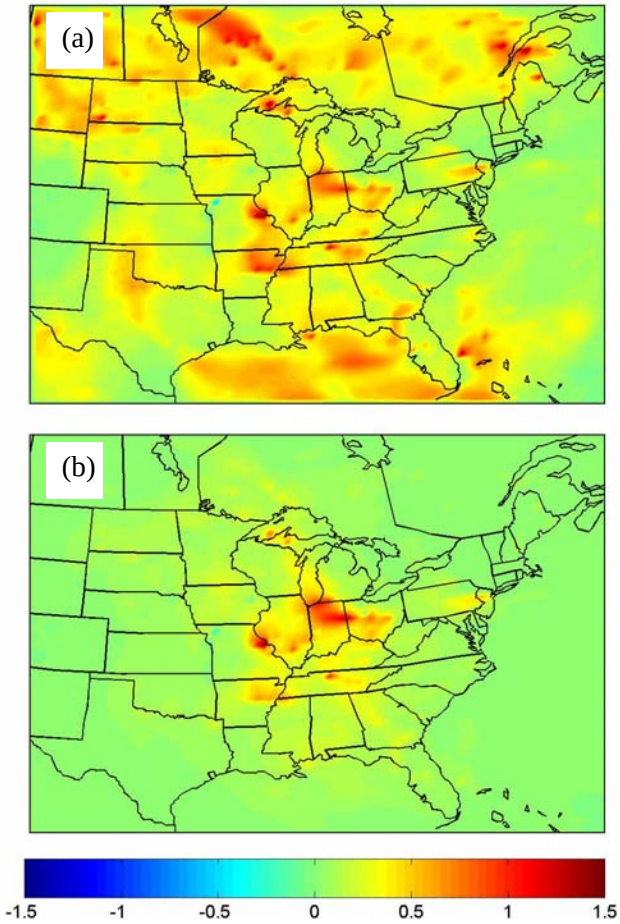

Fig. 12. Percent changes in simulation-average $PM_{2.5}$ concentrations divided by percent decrease in precipitating area ($\%\,\%^{-1}$) **(a)**, and changes in simulation-average $PM_{2.5}$ concentration ($\mu g\,m^{-3}$) **(b)**, calculated for a 4.9% decrease in area of precipitation in July.

favorably to measured precipitation. Additionally, snow and ice were assumed not to remove pollutants in the model. These results indicate that there is a moderate effect of precipitation rate (with fixed area of precipitation) on $PM_{2.5}$ concentrations, with the strongest relative effect (in terms of percent change in $PM_{2.5}$) in areas receiving light to moderate rainfall and in their downwind areas.

3.8 Precipitation area

The effects of changes in area of precipitation were again more pronounced in July than in January. The area of precipitation was defined as the average fraction of grid cells over which precipitation at any hour during the simulation. The simulation-average changes due to a 4.9% decrease in precipitating area in July are shown in Fig. 12. These changes were substantial over a large portion of the domain. Simulation-average sensitivities more negative than $-0.45\%\,\%^{-1}$ covered much of the domain in July (Fig. 12), though in January the differences in nearly all areas were between $-0.3\%\,\%^{-1}$ and zero. The average land-cell sensitivity of total $PM_{2.5}$ to the change in precipitating area in July was $-15\,ng\,m^{-3}\,\%^{-1}$ ($-0.2\%\,\%^{-1}$) while the average sensitivity in January was a factor of 50 smaller.

Pittsburgh and Atlanta were both more greatly affected by changes in precipitating area in July than in January (Table 2). In both cities in January, sensitivities were on the order of $0.1\,ng\,m^{-3}\,\%^{-1}$ ($0.001\%\,\%^{-1}$). In July, concentrations in Atlanta decreased by $60\,ng\,m^{-3}\,\%^{-1}$ ($-0.3\%\,\%^{-1}$), while concentrations in Pittsburgh decreased by $20\,ng\,m^{-3}\,\%^{-1}$ ($-0.1\%\,\%^{-1}$). These two cities, however, were affected less by changes in precipitation than much of the Midwest due to the location of precipitation in these cases (Fig. 9). Near St. Louis, absolute values of the sensitivities were near $350\,ng\,m^{-3}\,\%^{-1}$ (approximately $-1.5\%\,\%^{-1}$). The impact of the area of precipitation on $PM_{2.5}$ concentrations therefore appears to be a moderately

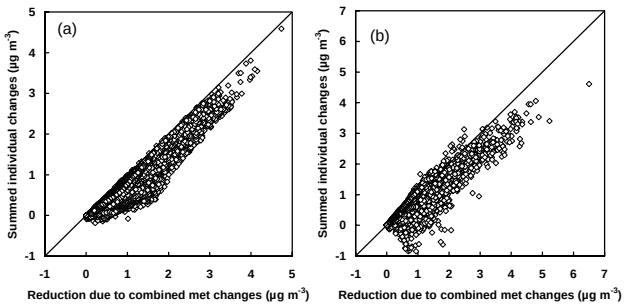

Fig. 13. Sum of changes in $PM_{2.5}$ concentrations from separate meteorological perturbations versus changes due to combined meteorological perturbations in **(a)** January and **(b)** July. Each data point represents a simulation-average concentration in one grid cell. All values have been multiplied by −1 for easier viewing. Lines are 1:1 lines.

important one, due to the large response in a rather small area, rather than a large mean response.

4 Additivity of effects

Two additional simulations were run in which perturbations in all eight meteorological parameters were imposed in both months. These perturbations are listed in Table 1. The resulting changes in average concentrations were compared to the sum of the changes that resulted when the perturbations were imposed individually. For simulation-long land-cell averages in July of total $PM_{2.5}$, ammonium, sulfate, and organics, the signs of the two predictions agreed. Both methods yielded predicted changes in simulation- and land-cell-average nitrate close to zero. The predicted changes in simulation- and land-cell-average sulfate were within 20% (or 0.03 $\mu g\,m^{-3}$) of one another, and predicted changes in organics differed by only 0.01 $\mu g\,m^{-3}$. Predicted changes in total $PM_{2.5}$ differed by 22%, or 0.15 $\mu g\,m^{-3}$. In January, both methods predicted the same signs for changes in total $PM_{2.5}$, ammonium, sulfate, nitrate, and organics. Predicted changes in total average $PM_{2.5}$ in January differed by 0.28 $\mu g\,m^{-3}$, or 32%. Predicted changes in average organics and nitrate were within 10% of each other, while there was a factor of 6 difference (0.21 $\mu g\,m^{-3}$) in predicted sulfate changes in January

A plot of the sum of individual changes in $PM_{2.5}$ concentrations versus the changes resulting from the combined-change simulation are shown in Fig. 13. The two changes were rather well correlated (R=0.95 in January and 0.94 in July), and the slope of the linear fit of the summed individual changes versus the combined changes was 0.73 in January and 0.77 in July. The summed individual changes were on average 27% smaller than the predicted changes from the combined-change simulation in January and 23% smaller in July. This is a reasonable agreement between the two methods, at least in this case.

5 Relative importance of meteorological parameters

The relative importance of the various meteorological parameters was estimated by taking into account the average sensitivities of total $PM_{2.5}$ to the meteorological perturbations, the spatial variability of sensitivities, and potential future changes in the meteorological parameters. The mean sensitivities were multiplied by climate-model-predicted meteorological changes to yield estimates of changes in total $PM_{2.5}$ concentrations due to each parameter. The mean sensitivities were calculated using the highest- and lowest-perturbation simulations for each variable, over which the sensitivities were roughly linear. These changes are summarized in Table 3. The work predicting these meteorological changes is summarized in Dawson et al. (2007).

Sensitivities of $PM_{2.5}$ 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). Expected 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 (SST) perturbation for cloudy area. The projections for 2050 are more modest than the doubled CO_2 and 4 K SST increase, so changes in wind speed and precipitating area may be underrepresented. 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 intensity were chosen so that somewhat liberal estimates of the total $PM_{2.5}$ sensitivity could be calculated and compared to the sensitivities to other parameters. The mean and 1st and 99th percentile values for sensitivities of total $PM_{2.5}$ concentrations were included so that the spatial variability of sensitivities could be taken into account. Temperature, absolute humidity, wind speed, and mixing height led to the largest $PM_{2.5}$ changes in January (with mean predicted responses on the order of hundreds of $ng\,m^{-3}$), while in July, absolute humidity, wind speed, mixing height, precipitation intensity, and precipitating area all had potentially major effects on $PM_{2.5}$ (Table 3). Temperature had little impact on mean July concentrations due to the competing effects on nitrate, sulfate, and organics; in January, the volatility of nitrate aerosol became dominant, causing larger decreases in $PM_{2.5}$ with increasing temperature. The range for the mean predicted effect of temperature on $PM_{2.5}$ concentrations was −24 to −71 $ng\,m^{-3}$ in July; in January the range was −260 to −770 $ng\,m^{-3}$. Neither cloud nor precipitation changes had a major impact on mean January $PM_{2.5}$ (with mean predicted changes less than or equal to 20 $ng\,m^{-3}$), though variability in the response to precipitation intensity indicates that it may be a somewhat important variable (with predicted responses up to 150 $ng\,m^{-3}$). In July, temperature,

Table 3. Summary of expected meteorological changes and their effects on $PM_{2.5}$ concentrations in January and July. (Major sensitivities in bold).

Meteorological Parameter	Predicted Change of Parameter	Sensitivity Mean (1%, 99%)	Predicted Effect ($ng\,m^{-3}$) Mean (1%, 99%)	Sensitivity Mean (1%, 99%)	Predicted Effect ($ng\,m^{-3}$) Mean (1%, 99%)
Temperature	+1.5 to +4.5 K[a]	$-171\,ng\,m^{-3}\,K^{-1}$ (−463, −6.83)	**−770 to −260** (−2100 to −10)	$-16\,ng\,m^{-3}\,K^{-1}$ (−238, 101)	−71 to −24 (−1100 to 450)
Absolute humidity	+7 to +21%[b]	$14\,ng\,m^{-3}\%^{-1}$ (0.078, 36.3)	**99 to 300** (0.55 to 760)	$20\,ng\,m^{-3}\%^{-1}$ (−28.2, 133)	**140 to 410** (−590 to 2800)
Wind speed	−1.4 to −4.5%[c]	$-33\,ng\,m^{-3}\%^{-1}$ (−142, 12.3)	**45 to 150** (−55 to 640)	$-53\,ng\,m^{-3}\%^{-1}$ (−215, 3.88)	**75 to 240** (−17 to 970)
Mixing height	−1 layer to +1 layer[d] (−150 m to +150 m)	$-73\,ng\,m^{-3}\,(100\,m)^{-1}$ (−629, 58.6)	**−110 to 110** (−630 to 630)	$-210\,ng\,m^{-3}\,(100\,m)^{-1}$ (−1290, 3.67)	**−310 to 310** (−1300 to 1300)
Cloud LWC and OD	−15 to +15%[d]	$-0.9\,ng\,m^{-3}\%^{-1}$ (−5.28, 0.185)	−14 to 14 (−80 to 80)	$-1.8\,ng\,m^{-3}\%^{-1}$ (−14.1, 2.28)	−26 to 26 (−210 to 210)
Cloudy area	−4.4 to −0.2%[e]	$-1.7\,ng\,m^{-3}\%^{-1}$ (−17.5, 20.8)	0.34 to 7.6 (−92 to 77)	$-14\,ng\,m^{-3}\%^{-1}$ (−99.4, 10.0)	2.9 to 64 (−44 to 440)
Precipitation rate	−20 to +20%[d]	$-1.0\,ng\,m^{-3}\%^{-1}$ (−7.51, 0.082)	−20 to 20 (−150 to 150)	$-17\,ng\,m^{-3}\%^{-1}$ (−68.2, −0.037)	**−330 to 330** (−1400 to 1400)
Precipitating area	−10 to +10%[f]	$-0.4\,ng\,m^{-3}\%^{-1}$ (−4.13, 0.381)	−3.7 to 3.7 (−41 to 41)	$-15\,ng\,m^{-3}\%^{-1}$ (−97.5, −0.0016)	**−150 to 150** (−980 to 980)

[a] IPCC, 2001
[b] Based on IPCC temperature projections and constant 80% RH.
[c] Breslow and Sailor, 2002
[d] Especially speculative; included to enable intercomparison among all parameters.
[e] Cess et al., 1990
[f] IPCC Data Distribution Centre: http://ipcc-ddc.cru.uea.ac.uk/sres/scatter_plots/scatterplots_region.html

cloudy area, and cloud LWC and OD had smaller but potentially important effects if variability is taken into account (with responses up to several hundred $ng\,m^{-3}$).

6 Conclusions

The strongest of the effects of changes in meteorology on $PM_{2.5}$ concentrations were the effects of temperature, wind speed, absolute humidity, mixing height, and precipitation. Wind speed, mixing height, and precipitation affected all PM species. Temperature increased average sulfate concentrations and decreased average nitrate and organics concentrations. The main effect of increased absolute humidity was increased nitrate aerosol. These effects could lead to appreciable changes in $PM_{2.5}$ concentrations under a changed future climate.

The response of $PM_{2.5}$ concentrations to changes in meteorology was the net effect of the changes in individual aerosol species. The qualitative behavior of the key processes responsible for these sensitivities should not generally be very sensitive to choice of modeled time periods, even though the calculated sensitivities are dependent on the time period and base case meteorology. $PM_{2.5}$ concentrations had a rather small response to temperature changes in summer ($-16\,ng\,m^{-3}\,K^{-1}$ on average), due largely to increases in sulfate canceling decreases in nitrate and organics, while $PM_{2.5}$ concentrations in winter decreased more strongly ($-170\,ng\,m^{-3}\,K^{-1}$ on average) because of reductions in nitrate and organics. $PM_{2.5}$ concentrations increased with increased absolute humidity in both winter ($14\,ng\,m^{-3}\,\%^{-1}$) and summer ($20\,ng\,m^{-3}\,\%^{-1}$), driven largely by increases in nitrate concentrations. Mixing height changes led to mixing and dilution effects, with $PM_{2.5}$ concentrations generally decreasing as mixing height was increased. The mean effect of mixing height changes was nearly 3 times larger in July than in January, due to lower average mixing heights during the simulated July period and somewhat lower concentrations in January. Increases in wind speed led to changes in advection and transport resulting in decreases in $PM_{2.5}$ concentrations of $33\,ng\,m^{-3}\,\%^{-1}$ in January and $53\,ng\,m^{-3}\,\%^{-1}$ in July.

Cloud LWC and OD and cloudy area led to small changes in $PM_{2.5}$ on average, but there were some areas with appreciable responses. Nitrate and organics generally decreased with increased cloud cover in both seasons; the same was true for sulfate in July, but not in January. As expected, $PM_{2.5}$ concentrations decreased with increased precipitation rate and precipitating area, though the sensitivities to changes in these precipitation parameters were over a factor of 10 larger in July than in January. The differences between seasons can be due to the differences in the dominant types of precipitation between the two seasons (large-scale in winter versus convective in summer), the rather small amount of precipitation in the modeled January period (Fig. 9), and the lack of scavenging by snow in the model. The largest mean expected changes for the imposed precipitation changes were between 0.1 to $0.8\,\mu g\,m^{-3}$ (Table 3), though the spatial variability in responses could mean precipitation-driven changes in $PM_{2.5}$ of up to approximately $3\,\mu g\,m^{-3}$ (Table 3).

The potential for changes in average concentrations of several $\mu g\,m^{-3}$ indicates that changes in meteorology can have important impacts on $PM_{2.5}$ concentrations. The changes in concentrations caused by changes in meteorology should, therefore, be taken into account in long-term air quality planning.

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