

Sensitivities of NO_x
transformation

H. Lei and J. X. L. Wang

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Sensitivities of NO_x transformation and the effects on surface ozone and nitrate

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

As precursors for tropospheric ozone and nitrate aerosols, Nitrogen oxides (NO_x) in present atmosphere and its transformation in responding to emission and climate perturbations are studied by CAM-Chem model and air quality measurements including National Emission Inventory (NEI), Clean Air Status and Trends Network (CASTNET) and Environmental Protection Agency Air Quality System (EPA AQS). It is found that not only the surface ozone formation but also the nitrate formation is associated with the relative emissions of NO_x and volatile organic compounds (VOC). Due to the availability of VOC and associated NO_x titration, ozone productions in industrial regions increase in warmer conditions and slightly decrease against NO_x emission increase, which is converse to the response in farming region. The decrease or small increase in ozone concentrations over industrial regions result in the responded nitrate increasing rate staying above the increasing rate of NO_x emissions. It is indicated that ozone concentration change is more directly affected by changes in climate and precursor emissions, while nitrate concentration change is also affected by local ozone production types and their seasonal transfer. The sensitivity to temperature perturbations shows that warmer climate accelerates the decomposition of odd nitrogen (NO_y) during the night. As a result, the transformation rate of NO_x to nitrate decreases. Examinations on the historical emission and air quality records on typical pollution areas further confirm the conclusion drawn from modeling experiments.

1 Introduction

Surface ozone and particulate matter (PM) are two major pollutants that affect US air quality (Blanchard and Hidy, 2003; Jacob, 2008) and which have a deleterious effect on the human respiratory system and health in general. The concentrations of surface ozone and PM-related secondary aerosols (nitrate, sulfate, and ammonium) are affected by the precursor emission and transformation rate (Spicer, 1983). Since NO_x

ACPD

13, 21961–21988, 2013

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



is a common precursor for ozone and nitrate aerosols, changes in NO_x emission or the climate induced change in related chemical reactions will affect the concentrations and partitioning of pollutants in the air (Seinfeld, 2006).

Modeling studies have been conducted to diagnose air quality change due to anthropogenic emission change, biogenic emission change and climate change under different scenarios (Wu et al., 2008; Pye et al., 2009; Lei et al., 2013). These studies emphasize the possible long-term changing scenarios, but do not account for the effects of perturbations on the present pollution status and interactions between pollutants, which actually is more meaningful for practical air quality change (Jacob et al., 2008). Considering the vital role of NO_x in ozone and nitrate production, sensitivities of NO_x transformation to ozone/nitrate need to be learned in response to emission and climate perturbations for present air quality.

In addition, long term air quality measurements and well established emission inventories archived significant processes about air quality change in the past and present. For example, the records from Environmental Protection Agency Air Quality System have been used to examine the spatial and temporal variation of pollutants in the United States (Choi et al., 2012; Lei et al., 2012; Lei and Wang, 2013). National Emission Inventory has been used in determining the contributions of emission sources to concentrations of particulate matter and gases to the southeastern United States (Blanchard, et al., 2012). Based on these measurements and inventories, we can assess the changes in concentrations of surface ozone and nitrate aerosols at the present atmospheric condition, and further examine their productions associated with NO_x emissions.

In this study, we comprehensively analyze the sensitivities of NO_x transformations in the present atmosphere through numerical experiments and diagnoses of observational air quality data. First, a set of sensitivity experiments were conducted to evaluate the sensitivities of NO_x transformation pathway against emission or climate perturbations. The emission perturbation was examined by increasing or decreasing the global NO_x emission by 25%. The climate perturbation was examined by increasing or de-

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

is increased (or decreased) by 1 °C to isolate possible influences on transformation rate. The outputs from both positive and negative perturbation runs, representing temperature increases and decreases, are compared with control run results to examine anomalies. These results provide us with information about the tendency and stability of changes in pollutant levels. For instance, if the change in pollutant concentration induced by positive perturbations is larger than that induced by negative perturbations, it means that the transformation tendency to that pollutant is accelerated with positive perturbation. Similarly, if a pair of experiments shows close patterns for changes in pollution levels, the corresponding sensitivity to that perturbation is stable.

After the numerical experiments, the measurements of surface ozone and nitrate aerosol concentrations over typical mega cities will be analyzed for the variability in recent several years. The variability will be associated with the variation of NO_x and other precursor emissions to evaluate and better understand the conclusions drawn from numerical studies. Measurements of pollutants include the CASTNET observations of nitrate and the EPA AQS records of surface ozone. Emission analysis will use the emission budgets from US EPA National Emissions Inventory.

The CAM-Chem model used in this study includes a simulation of O₃-NO_x-CO-VOC chemistry and aerosol chemistry module, and its ability in reproducing global ozone and aerosol levels has been established in a suite of tropospheric ozone or aerosol studies (e.g., Tie et al., 2005; Murazaki and Hess, 2006; Emmons et al., 2010; Lei et al., 2012, 2013). Emissions of ozone precursors are based on the present-day inventory (for the year 2000) as described by Emmons et al. (2010). The meteorology data that drives the CAM-Chem model is the National Centers for Environmental Prediction reanalysis II 1° × 1° fields from 1999 to 2002 (Kanamitsu et al., 2002). The six-hour interval data were re-gridded to a horizontal resolution of 1.9° latitude by 2.5° longitude and 26 vertical levels extending up to 0.01hPa. Evaluation run and sensitivity experiments are performed during the four-year period. After one year's spinning up, the model result for the last three years is used in the analysis and comparisons.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

3 Results and discussion

3.1 Evaluation for the current atmosphere and emissions

To evaluate CAM-Chem's ability to reproduce the surface ozone and nitrate aerosol concentrations, we compare the model-derived concentrations over the contiguous United States around 2000 relative to the US Environmental Protection Agency Air Quality System site measurements. The model is driven by CCSM3 meteorology and running for 2000–2002 is compared against the same observations by using 3 yr average data. All measurements which include at least a 75 % record of valid operations at the rural sites during the whole comparison period are selected within a given model grid cell. Then the selected measurements are averaged to get a grid cell mean value that can be directly compared with the modeled value for that grid cell.

Since summer is the active period for ozone production and the availability of ozone measurements are the best in summer, we therefore compare the summer average surface ozone concentrations to evaluate the model performance. Figure 2 shows the CAM-Chem derived summer average surface ozone concentration over the contiguous US and the corresponding observations from EPA AQS. We find that the model is able to reproduce the summer ozone concentrations over most area of the contiguous United States, with overall biases smaller than 20 ppb. Similar spatial patterns and values of biases in the comparison of daily 8 h maximum ozone concentrations over the contiguous United States have been reported in Lei et al. (2012).

Nitrate aerosols exist in the atmosphere in various phases and forms. Direct measurements of the air concentrations of nitrate aerosols are not as easy as other pollutants. As a result, present observations of nitrate aerosol concentrations are derived from depositions. The CASTNET observations has systematically recorded the total nitrate deposition in the past decade and derived total nitrate concentrations based on it. Therefore, we compare the simulated total nitrate (nitrate aerosols + nitric acid) concentrations with CASTNET concentration map to evaluate the CAM-Chem simulations. Figure 3 shows the CAM-Chem simulated (right) and CASTNET derived (left)

annual-mean concentration of total nitrate ($\mu\text{g m}^{-3}$) over the contiguous US in 2002. The CAM-Chem model captures the pattern of total nitrate concentrations. The main difference is on the coastal region of Texas, where the lack of observations would be the main reason. The general biases on other area are within $1 \mu\text{g m}^{-3}$, which basically convinced the ability of CAM-Chem model in following sensitivity experiments.

3.2 Transformation rate change response to emission perturbations

Figure 4 shows the annual mean change ratio of surface nitrate aerosol concentration simulated from the experiment with 25% increase (or decrease) of global NO_x emission. The patterns are similar between the two experiments, which indicate a stable sensitivity in response to emission perturbations. Comparing magnitudes of two diagrams in Fig. 4, it is clear that the tendency of transformation from NO_x to nitrate increases with NO_x emission increase. In the industrial regions of the United States (the east, the coastal area of Texas, and southern California), the change rate of nitrate is smaller than the change rate of NO_x emissions. Over the majority of the non-industrial area of the United States, the change rate of nitrate is generally higher than the change rate of emission. This result suggests that the NO_x transformation rate over industrial regions is much lower, which may be caused by the limitation of available base precursors of nitrate aerosols.

For the transformation pathway from NO_x to ozone, Fig. 5 shows the annual mean change ratio of surface ozone concentration. Over the major industrial regions of the eastern United States, the ozone concentration decreases in response to emission increase, which means less active NO_x is transformed. Hydrocarbon-limited surface ozone production and associated NO_x titration are responsible for the converse relationship in the industrial regions. For the United States, the overall change rate of ozone is also considered very low, in comparison to the 25% change rate in NO_x emissions, which is attributable to the limitation of hydrocarbons.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



positive to negative tendencies. One is around April and the other is around September. For September transition, the tendency changing for the experiments of increasing NO_x emission sign earlier than that for the emission reduction ones, while it is very close for April transition. In summary, an increase of NO_x emissions results in an earlier transfer to hydrocarbon-limited type. This pattern is a result of the variation of hydrocarbon emissions, which has strong seasonality. The relative level of hydrocarbon (hydrocarbon/ NO_x) determines the ozone production type and thus affects the transfer time in each case.

3.3 Transformation rate change response to temperature perturbations

Figure 8 shows the annual mean change ratio of surface nitrate aerosol concentration from the experiments with a temperature increase or decrease of 1°C . Generally, nitrate aerosol concentration over the United States decreases as a response to temperature increase at current emission level. Increase in temperature tends to depress the chemical and physical processes relevant to nitrate aerosol formation and thus decrease nitrate concentration. These results are explainable by temperature impact on N_2O_5 formation. Warmer temperature tends to accelerate the decomposition of NO_y ($\text{N}_2\text{O}_5 \rightarrow \text{NO}_2 + \text{NO}_3$ and $\text{NO}_3 + h\nu \rightarrow \text{NO} + \text{O}_2$), and thus results in less nitrate production (Seinfeld et al., 2006). In addition, active NO_x concentration is also affected by the NO_x -to-ozone transformation pathway, which in turn affects nitrate formation.

For the temperature impacts on the NO_x -to-ozone transformation pathway, Fig. 9 shows the annual mean change ratio of surface ozone concentration. Surface ozone production is directly affected by climate and precursor emission change due to the rapid photochemical reactions for ozone formation (Seinfeld et al., 2006). The ozone concentration increases generally over continental United States, except mountain regions and northern border areas where the NO_x concentration is mainly a result of transport from remote sources. Since higher temperatures tend to increase the ozone production efficiency, more NO_x is consumed in ozone production over source regions. Therefore, less NO_x produced in industrial regions is transported to farming regions

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Sensitivities of NO_x
transformation**

H. Lei and J. X. L. Wang

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



with relatively low emissions, and thus ozone concentration in the farming regions decreases. Conversely, it may enhance the transport of hydrocarbons to industrial regions by increasing the gradient of hydrocarbon concentrations. According to the nitrate aerosol concentration change, increased NO_x involvement in ozone production results in less active NO_x available for nitrate formation (e.g. affecting N₂O₅ formation) and thus contributes to the decrease in nitrate concentration.

In addition, the chemical reactions that occur during the night are important in aiding the transformation of NO_x into nitrate (Seinfeld et al., 2006), since the medium chemicals for nitrate formation are formed nocturnally (Fig. 1). Temperature is an important factor affecting nocturnal chemistry. Therefore, it is necessary to examine the diurnal change in the sensitivity of NO_x transformation pathways to temperature change. Figure 10 shows the diurnal cycle of the annual mean concentration change averaged over the industrial region (defined in Fig. 6). The ozone concentration is slightly increased by about 2 % over present value as a response to the 1 °C temperature increase. Although the change in transformation rate is near constant for the whole day, it is relatively larger during the daytime than the nighttime. Nitrate concentration decreases during the day and sharply increases at night, with a change ratio from –0.2 to 0.3. This also indicates that ozone production is relatively independent from nitrate production. The daytime decrease of nitrate is mainly caused by the accelerated NO_y decomposition and also the increased consumption of active NO_x in ozone formation. During the night, ozone is consumed and contributes to nitrate formation (shown in Fig. 1). As a result, NO_x transformation to nitrate is enhanced. However, nitrate formation is also influenced by other factors including humidity, the availability of base precursors, and concentrations of other acids. As a result, potential responses of nitrate aerosol concentration to a changing climate and emission are fairly complex. Observations and microphysical studies of its formation may further improve our understanding.

3.4 Evidences in observations

In order to get evidences from observations and convince the findings drawn from sensitivity experiments, we further examine the NO_x transformation in existing emission and air quality records. These datasets include NEI emission datasets, CASTNET nitrate observations and EPA AQS ozone observations. Two typical cities are selected to represent the two types of ozone production environment in the analysis. The first is Los Angeles. Los Angeles is a heavy polluted city where the ozone production type is strongly VOC-limited (e.g. Harley et al., 1993; Steiner et al., 2006). Nitrate observations on the site (JOT403) located in Los Angeles from CASTNET will be used in the analysis. Ozone data in the surrounding area of site-JOT403 will be averaged based on EPA AQS records. The other is Atlanta city in Georgia. Ozone production in Atlanta is known to be a typical NO_x -limited type (Sillman et al., 1995). Similarly, we use data on the site (GAS153) located on the suburban area of Atlanta from CASTNET to do the analysis.

Figure 11 shows the historical variations of emissions and pollutant concentrations in Los Angeles from 1996 to 2012. Here, we show the change rates of surface ozone and total nitrate on the study area. The reason to choose total nitrate (nitrate aerosols + nitric acid) is to avoid the influence of sulfate and availability of ammonia on nitrate formation (Lei and Wuebbles, 2013), so that the analyzed change can largely represent the specific effect of NO_x transformation. Generally, both NO_x and anthropogenic VOC emissions slowly decrease. NO_x decrease rate is around 20 % by the year 2012. Considering the large amount of VOC emitted from biogenic sources in summer time, the VOC decreasing rate may be less than the trend shown in Fig. 11. It is shown that both summer time surface ozone concentration and nitrate concentration shows decreasing trends. Nitrate decreasing rate is larger than that for surface ozone, which echoes the results from the model sensitivity analysis (in Fig. 7). Due to the reduction of biogenic VOC emissions in winter time, the surface ozone production in Los Angeles is strongly VOC-limited. As a result, the total nitrate concentration decreases as much as 60 %

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

in response to the NO_x decrease, while the ozone concentration does not show clear decreasing trend. According to the analyses in sensitivity study, more active NO_x can be produced for the nitrate formation with a rate above the NO_x increasing rate under the VOC-limited ozone production environment. Now, under the same environment, the nitrate decreasing rate is larger than the decreasing rate of NO_x. This observational variability convinced the analyses in modeling sensitivity studies. It indicates that nitrate production is associated with local ozone production type.

Figure 12 shows the other observational results from a typical NO_x-limited region-Atlanta, Georgia. Although ozone production in Atlanta is reported to be a typical NO_x-limited type (Sillman et al., 1995), the analyses in suburban regions of Atlanta would further ensure this type (Pierce et al., 1998). The summer time nitrate and ozone both show decreasing trends in response to NO_x emission reduction in the past decade. Nitrate shows a decreasing rate that is larger than the decreasing rate of ozone. The winter time ozone production in the suburban of Atlanta is still be NO_x-limited type. Therefore, the change rate of nitrate is much lower than the change rate under VOC-limited environment (e.g. Los Angeles). Although the winter time ozone data is not abundant in Atlanta area, available data also shows a clear decreasing trend.

4 Conclusions

As a precursor for tropospheric ozone and nitrate aerosols, Nitrogen oxides (NO_x) and its transformation are heavily related to their concentrations and the total pollution level. Climate and emission changes are two key factors that dictate NO_x transformation. In this study, the Community Atmosphere Model with Chemistry is used for sensitivity simulations to study NO_x transformations in responding to emission and climate perturbations. In the emission perturbation experiments, ozone concentration over the industrial regions is relatively stable, and its causes are attributable to the limitation of available hydrocarbon precursors and nitrate titration. The decrease or small increase in ozone concentration in response to a large increase in NO_x emission leads to more

face ozone or nitrate concentration would be valuable topics to further understand the complicate processes associated with the ozone/nitrate formation and their practical changes.

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5 ship and NOAA air resources laboratory.

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Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Sensitivities of NO_x
transformation**

H. Lei and J. X. L. Wang

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- Jacob, D. J. and Winner, D. A.: Effect of climate change on air quality, *Atmos. Environ.*, 43, 51–63, 2008.
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ACPD

13, 21961–21988, 2013

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

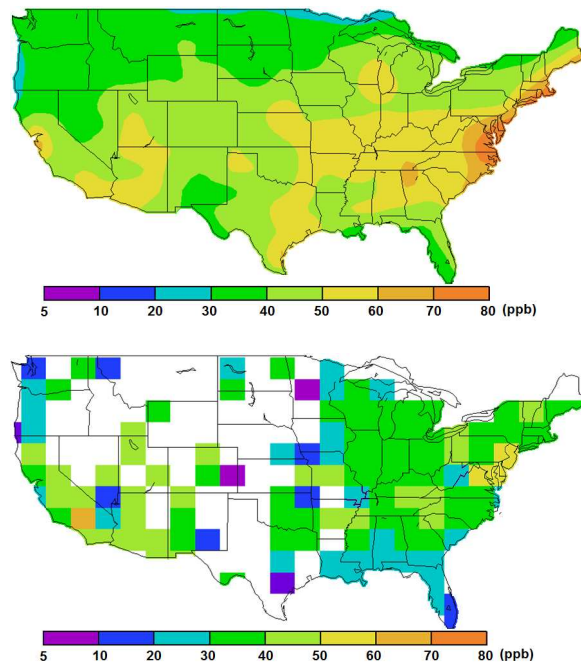
[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Fig. 2. Summer average surface ozone concentrations over the contiguous United States during 2000–2002. The CAM-Chem simulated results (Left) and the US EPA AQS site measurements (Right) (Unit: ppb).

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

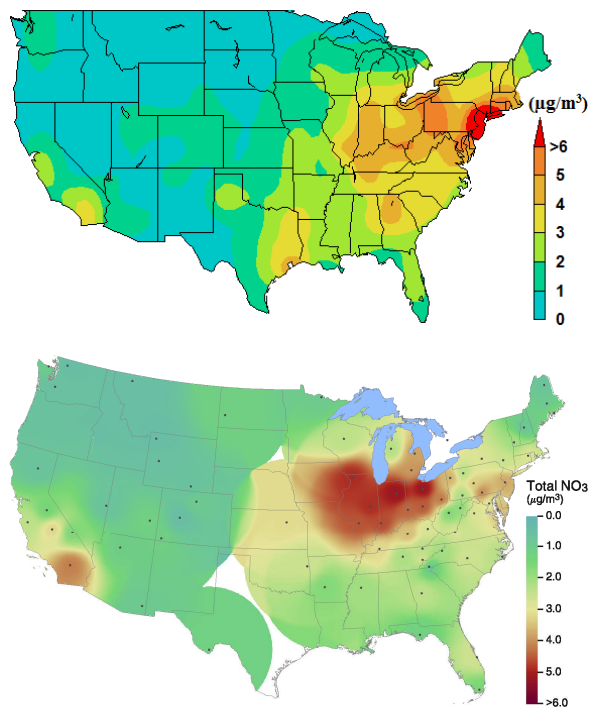
[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Fig. 3. Annual average total nitrate concentrations over the contiguous United States in 2002. The CAM-Chem simulated results (Left) and the CASTNET observations (Right, image credit CASTNET) (Unit: $\mu\text{g m}^{-3}$).

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

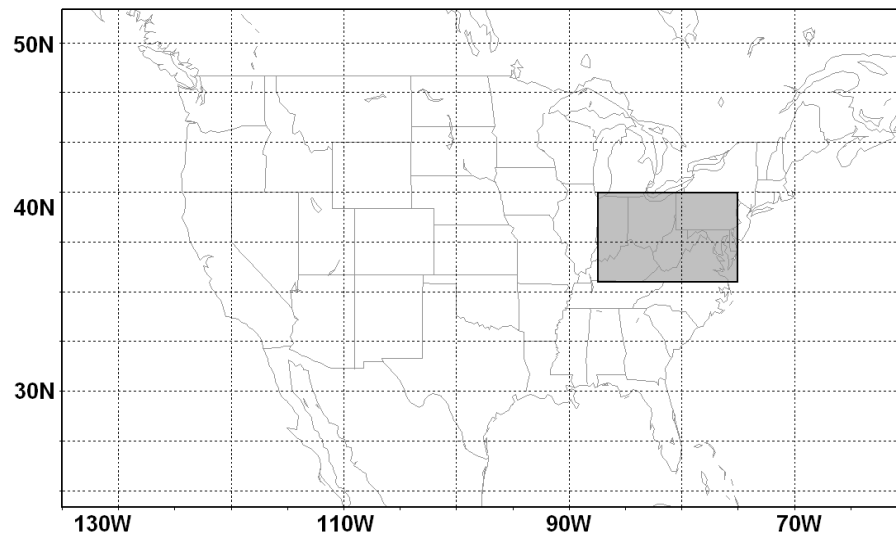


Fig. 6. The shaded rectangle region is the sensitive region used for analysis. Surface aerosol concentrations are averaged over this area.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Sensitivities of NO_x
transformation**

H. Lei and J. X. L. Wang

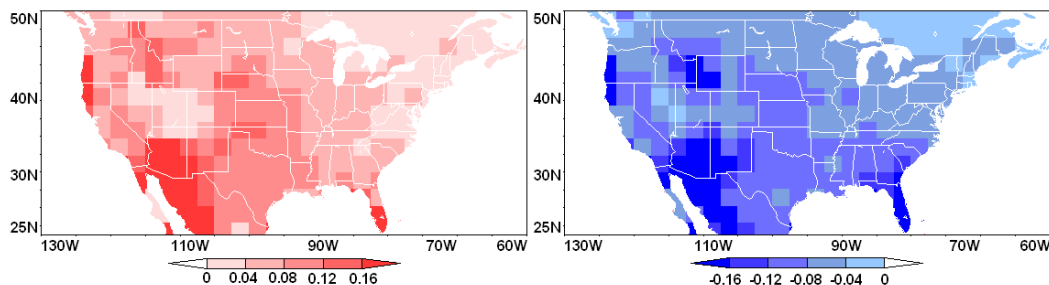


Fig. 8. Annual mean change ratio of surface nitrate aerosol concentration ((Experiment-Control run)/Control run, Left: result of 1°C temperature decrease; Right: result of 1°C temperature increase.).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

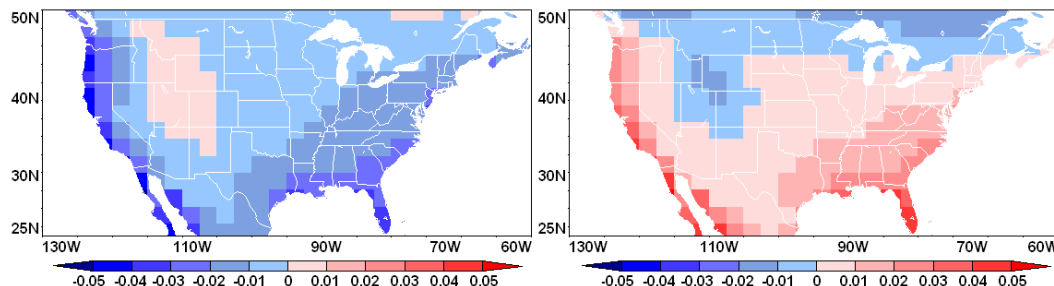


Fig. 9. Annual mean change ratio of surface ozone concentration ((Experiment-Control run)/Control run, Left: result of 1°C temperature decrease; Right: result of 1°C temperature increase.).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Sensitivities of NO_x transformation

H. Lei and J. X. L. Wang

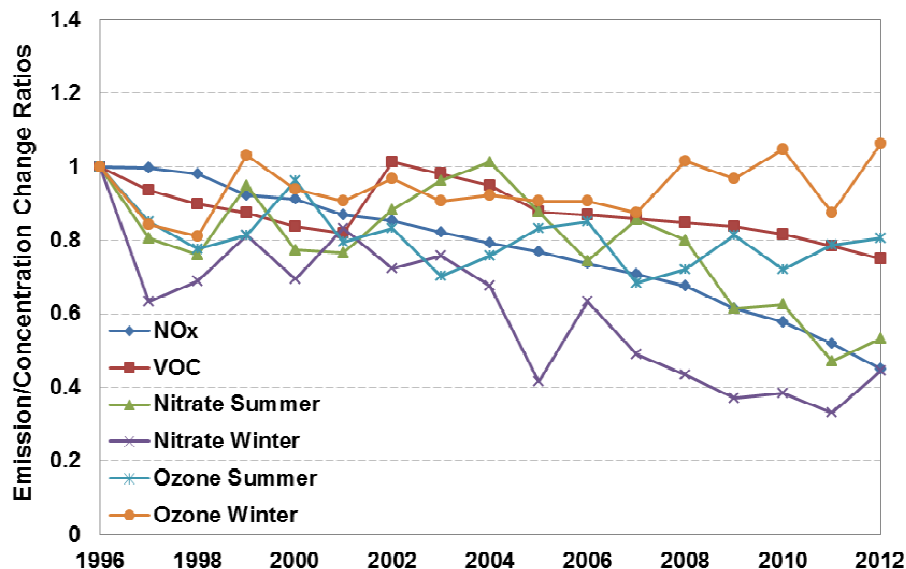


Fig. 11. Historical variations of emissions and pollutant concentrations in Los Angeles, CA during 1996 and 2012. The data for NO_x and VOC emissions are from NEI dataset. The change rate of total nitrate in summer and winter are calculated from CASTNET measurements at site JOT403. The change rate of ozone is calculated from daily 8 h maximum concentrations of surface ozone which are averaged from US EPA AQS records at the same county where the site JOT403 is located. The change rate is calculated based on the first year concentration (e.g. 1996).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Sensitivities of NO_x transformation

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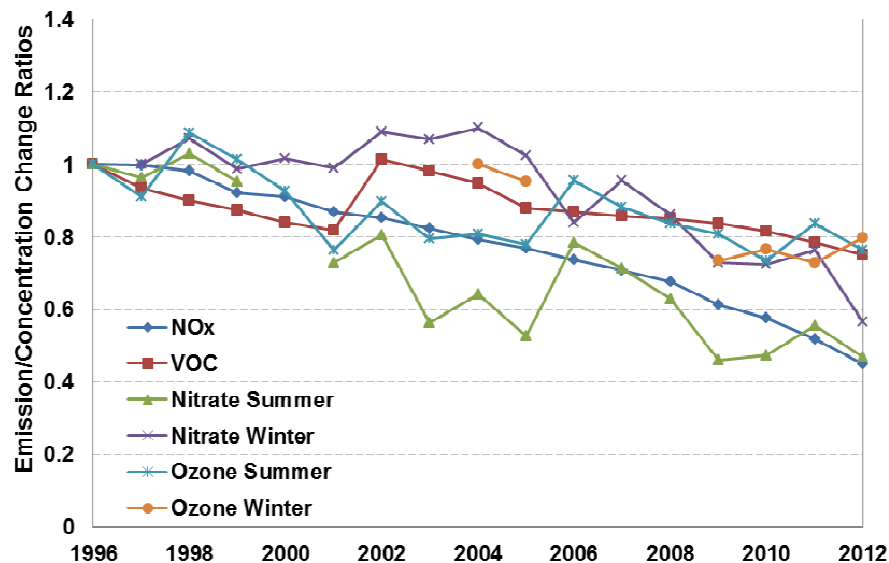


Fig. 12. Historical variations of emissions and pollutant concentrations in suburban of Atlanta, GA during 1996 and 2012. The data for NO_x and VOC emissions are from NEI dataset. The change rate of total nitrate in summer and winter are calculated from CASTNET measurements at site GAS153. The change rate of ozone is calculated from daily 8 h maximum concentrations of surface ozone which are averaged from US EPA AQS records at the same county where the site GAS153 is located. Winter ozone in this area is not recorded until recent three years. The change rate is calculated based on the first year concentration (e.g. 1996).