Increasing influence of Canada anthropogenic and the Great Lakes Region shipment SO2 emission on ultrafine particle number concentrations in New York State

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Abstract

The adverse health effects of exposure to high levels of ultrafine particulate number concentration have been widely reported. New York State (NYS) borders southeastern Canada and the Great Lakes Region and is influenced by air pollutants from these upwind source regions. Through comparison of observed and simulated CN10 (condensation nuclei >10 nm) at rural and remote sites in NYS, we show that Canada anthropogenic and the Great Lakes Regions shipment SO_2 emission (CAGLESO2) significantly influenced CN10 in NYS. These emissions on average produced a 22% enhancement of CN10 in NYS in 2017, varying from 40% in Northwestern NYS to 10% in Southeastern NYS. We also found that the impact of CAGLESO2 on NYS's CN10 in 2017 was 2.5 times higher than that in 2005 and 1.6 times higher than that in 2011, which indicated increasing influnece of CAGLESO2 on CN10 in NYS over the last decade.

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Key points

- Simulated CN10 number concentrations were compared with observations at rural and remote sites in New York State (NYS)
- Canada anthropogenic and the Great Lakes Regions shipment SO_2 emission (CAGLESO2) produced a 22% enhancement of CN10 in NYS in 2017
- The impact of CAGLESO2 on CN10 in NYS has become increasingly important due to continuous emission reductions in the US over the last decade

Abstract

The adverse health effects of exposure to high levels of ultrafine particulate number concentration have been widely reported. New York State (NYS) borders southeastern Canada and the Great Lakes Region and is influenced by air pollutants from these upwind source regions. Through comparison of observed and simulated CN10 (condensation nuclei >10 nm) at rural and remote sites in NYS, we show that Canada anthropogenic and the Great Lakes Regions shipment SO_2 emission (CAGLESO2) significantly influenced CN10 in NYS. These emissions on average produced a 22% enhancement of CN10 in NYS in 2017, varying from 40% in Northwestern NYS to 10% in Southeastern NYS. We also found that the impact of CAGLESO2 on NYS's CN10 in 2017 was 2.5 times higher than that in 2005 and 1.6 times higher than that in 2011, which indicated increasing influnece of CAGLESO2 on CN10 in NYS over the last decade.

Plain Language Summary

Human exposure to high levels of ultrafine particulate number concentration can cause adverse health effects. Better understanding of sources, distribution, evolution, and sinks of ultrafine particles are critical knowledge gaps that need to be addressed to understand the role of ultrafine particle exposure on adverse human health effects. In this work, we compared the simulated CN10 (condensation nuclei with diameter larger than 10 nm) with observations at rural and remote sites in New York State (NYS) which are operated by the Atmospheric Sciences Research Center of State University of New York at Albany and found Canada anthropogenic and the Great Lakes Regions shipment SO_2 emission (CAGLESO2) significantly influenced CN10 in NYS. Our study also indicated that the impact of CAGLESO2 on CN10 in NYS has become increasingly important due to continuous reductions of anthropogenic emissions in the US over the last decade.

1. Introduction

Ultrafine particles are particulate matter of nano-scale size (diameter < 100 nm) which are more than an order of magnitude smaller than the sizes of those particles that typically dominate regulated PM10 and PM2.5. Prior studies found ultrafine particles have several more aggressive health implications than PM10 and PM2.5 (Schraufnagel, 2020; Yacobi et al., 2010; Oberdörster et al., 1994). Ultrafine particles contribute little to the total particle mass but comprise an estimated 90% of particle number concentration (HEI, 2013). Its number concentrations can be represented by condensation nuclei measurements (Maston et al., 2004). Better understanding of sources, distribution, evolution, and sinks of ultrafine particles are critical knowledge gaps that need to be addressed to understand the role of ultrafine particle exposure on adverse human health effects.

New York State (NYS) is one of the Mid-Atlantic states (New York, New Jersey, Pennsylvania, Delaware, Maryland, Washington, D. C., Virginia, and West Virginia) and borders southeastern Canada (Ontario and Quebec) and the Great Lakes Region (Lakes Superior, Michigan, Huron, Erie, and Ontario). Because of planetary winds and monsoon, NYS is primarily influenced by northwesterly winds in winter and southerly winds in summer (Turner, 1900). Wind carries air pollutants from upwind source regions and impacts air pollution level in NYS (Emami et al., 2018). In the last decades, the Clean Air Act has led to remarkable air pollutant emission cuts and air quality improvements in the United States. Due to emission reduction in the Mid-Atlantic states, SO_2 emissions in southeastern Canada and the Great Lakes Region became comparable with local emissions in NYS and surrounding regions. As reported by the United States Environmental Protection Agency (USEPA)'s bottom-up emission estimations, the US anthropogenic SO_2 emission was reduced 83% from 2005 to 2017. In the meantime, anthropogenic SO_2 emissions decreased from 387 thousand tons to 26 thousand tons in NYS (93% reduction), from 2756 thousand tons to 210 thousand tons in the Mid-Atlantic states (92% reduction), from 758 thousand tons to 291 thousand tons in southeastern Canada (62% reduction), and from 148 thousand tons to 141 thousand tons in the Great Lakes Region (5% reduction), respectively (Figure S1). In 2017, anthropogenic SO_2 emission in southeastern Canada was 11 times higher than that in NYS and 1.4 times higher than that in the Mid-Atlantic states. The Great Lakes Region shipment SO_2 emission was 5.4 times higher than that in NYS and 67% of that in the Mid-Atlantic states. SO_2 is an important precursor of new particle formation which dominates particle number concentration in rural and remote regions (Yu et al., 2015; Kulmala et al., 2014; Yu and Luo, 2009; Merikanto et al., 2009; Kulmala et al., 2006). It is important to know the potential impacts of Canada anthropogenic and the Great Lakes Regions shipment SO_2 emission (CAGLESO2) on CN10 in NYS which provide useful information to the public and policymakers. Atmospheric chemistry models with detailed aerosol microphysics which let them successfully predict particle number concentrations have been developed (Yu and Luo, 2009; Zaveri et al., 2008; Spracklen et al., 2005; Adam and Seinfeld, 2002) and can be

used to access the impact of individual surface sources on CN10 via the zero-out emissions method which assumes emissions from testing regions/sources to be zero.

In this study, we simulated CN10 in the Northeastern US with the 3-D chemical transport model GEOS-Chem coupled with the Advanced Particle Microphysics (APM) package (Yu and Luo, 2009). Using the long-term CN10 observations at Pinnacle State Park (PSP) and Whiteface Mountain Summit (WMS) in NYS which are operated by the Atmospheric Sciences Research Center of State University of New York at Albany (Schwab et al., 2016), model performance was evaluated. Then the model was used to investigate the impacts of CAGLESO2 on CN10 in NYS by the zero-out emissions method in the two regions.

2. Observed and simulated CN10 at NYS rural and remote sites

GEOS-Chem is a widely used global 3-D model of atmospheric chemistry with numerous updates to keep the model a state-of-the-art tool for the investigation of a wide range of atmospheric composition problems (Luo et al., 2020; Luo at al., 2019; Holmes et al., 2019; Keller et al., 2014; Bey et al., 2001). We run the model at $0.5^o \times 0.625^o$ horizontal resolution for a nest domain (34^oN to 52^oN; 97^oW to 65^oW) whose boundary conditions were provided by a $4^o \times 5^o$ global simulation. Eight emission sensitivity cases were run to investigate the impact of CAGLESO2 on CN10 in NYS (Table 1). In this study, we used CN10 observations at PSP and WMS sites to evaluate model performance. The PSP site (42.09^oN and 77.21^oW) is located in Addison, NY, a village in southwestern NY. The WMS site (42.09^oN and 77.21^oW) is located in Wilmington, NY, one of the High Peaks of the Adirondack Mountains in northeastern NY, and is above the forest canopy at about 1490 m (Schwab et al., 2016). CN10 number concentration was measured with TSI model 3783 CPCs.

Figure 1a shows the model simulated horizontal distribution of annual mean CN10 in NYS. CN10 is high in Western NYS and Southern NYS and is low in Northeastern NYS. The values are varied from ~2000 cm^{-3} to above 4000 cm^{-3}. PSP is located at Southwestern NYS with higher values of CN10, while WMS is located at Northeastern NYS where CN10 are lower than other parts of NYS. The two sites are ideal for characterizing CN10 in NYS. Time series of observed and simulated CN10 by US+CG case and US case (Table 1) in 2017 at the two sites are compared in Figure 1 (b-c). At PSP, CN10 is high in spring and fall and low in summer and winter. Low CN10 values resulted from low new particle formation rates which are limited by high temperature in summer and low level of sulfuric acid gas produced by oxidization of SO_2 during winter. Normalized mean biases (NMB) of simulated CN10 by US+CG case and US case are 2.9% and -20.6%, respectively. At the absence of CAGLESO2, simulated CN10 is obviously lower than oberservation in March. Some peaks of CN10 observed during spring are missed by the US case. The correlation coefficient between observed and simulated daily CN10 by US+CG case is high up to 0.59. Due to CAGLESO2 zero-out, the correlation coefficient between observation and simulation drops to 0.39, indicating that CAGLESO2 does not only affect CN10

values at PSP but also CN10 time variations at PSP. We also compare observed and simulated CN10 at WMS (Figure 1c). WMS is perched high atop the Whiteface Mountain summit, where observation is sensitive to air mass carrying air pollutions. To reflect the observed air mass at WMS, we sample model results at the layer where the temperature is closest to observation. Different from the seasonal variation of CN10 at PSP which shows a clear drop of CN10 during summer, WMS's CN10 is continuously high from spring to fall. It is because unstable boundary layer conditions during summer transport more pollutants from the surface to the mountain summit. NMBs of CN10 at WMS by US+CG case and US case are 65.5% and 37.8%, respectively. The model overestimates CN10 at WMS. Correlation coefficients between observed and simulated CN10 are 0.48 for US+CG case and 0.37 for US case, respectively. It is clear that the absence of CAGLESO2 significantly reduces correlation coefficients between observation and simulation at both PSP and WMS sites. It indicates that Canada anthropogenic SO_2 emission and the Great Lakes Region shipment SO_2 emission play important roles in CN10 time variations at the two sites.

3. Impact of CAGLESO2 on CN10 in NYS

Figure 2 shows the simulated impacts of CAGLESO2 on SO_2 volume mixing ratio ($[SO_2]$), sulfur acid gas volume mixing ratio ($[H_2SO_4]$), nucleation rate (J), number concentration of secondary particles larger than 10 nm (CN10SP), and CN10 in NYS. To focus on the impact of CAGLESO2 in NYS, we exclude grids out of NYS from domain mean calculation and further analysis. As shown in Figure 2a, the domain mean of relative changes of $[SO_2]$ in NYS due to inclusion of CAGLESO2 is 78.0%. The impact is reduced from 150-200% in Northwestern NYS to 15-30% in Southeastern NYS. SO_2 does not directly impact new particle formation and CN10 but is involved via its oxidation product sulfuric acid gas. Sulfuric acid gas is critical for new particle formation and growth in most rural and remote regions (Yu et al., 2015; Kulmala et al., 2006). Figure 2b shows that $[H_2SO_4]$ is enhanced 47.4% in NYS and has a similar spatial pattern to that of $[SO_2]$. Increased $[H_2SO_4]$ in NYS significantly enhances nucleation rate at corresponding regions and results in a 134.5% enhancement of nucleation rate in NYS (Figure 2c). The impact of CAGLESO2 on CN10SP in NYS is shown in Figure 2d. CN10SP in NYS is increased 26.8% which is much lower than the increase in nucleation rate. It is because CN10SP number concentration is controlled not only by new particle formation but also by the transport of CN10SP from surrounding regions. Background CN10SP reduces the changes of CN10SP corresponding to the changes of nucleation rate. Figure 2e shows the changes of CN10. CN10 in NYS includes both secondary and primary particles. Although CN10 in NYS is dominated by secondary particles (Luo et al., 2011), we still find 16.5% of these CN10 are primary particles (Figure S2). Due to the influence of primary particles, CN10 in NYS is increased 22.4% which is ~80% of that of CN10SP. The high impacts of CAGLESO2 on CN10 in NYS are found at Rochester (43.16ºN, 77.61ºW), a city located at Western NYS nearby Lake Ontario, and surrounding regions with the values of 30-50%, while low impacts on CN10 are found at the downstate region with the values of 5-15%.

Figure 3 shows seasonal variations of the impacts of CAGLESO2 on CN10 in NYS, at Rochester, PSP, and Albany, the capital of NYS. Maximum domain mean impact of CAGLESO2 on CN10 in NYS appears in March with the value of 55.6%, while minimum domain mean impact of CAGLESO2 on CN10 in NYS appears in October with the value of 16.0%. At Rochester, the impact of CAGLESO2 on CN10 is increasing from January to June and then is decreasing until September with a minimum value of 27.5%. It reaches its maximum value of 56.9% in March and then slightly decrease in April and then reaches its second high value of 55.7% in June. At PSP, the impact of CAGLESO2 on CN10 is high in spring which varies from 38.1% to 74.3% and low in late summer and early fall which varies from 14.2% to 23.7%. At Albany, the first peak of the impact of CAGLESO2 on CN10 appears in March with a value of 44.3%, and the second peak appears in December with a value of 28.1%. The minimum impact of CAGLESO2 on CN10 appears in April with a value of 8.6%. Figure 3 indicates that the impact of CAGLESO2 on CN10 shows significant seasonal changes whose amplitude of variation can be high up to a factor of 5. The enhancements of CN10 in NYS, at Rochester, PSP, and Albany caused by Canada anthropogenic SO_2 emission are 2.2, 2.5, 2.2, and 3.2 times higher than those caused by Great Lakes Region shipment SO_2 emission, respectively. It indicates anthropogenic SO_2 emission in Canada have a stronger impact on CN10 in NYS than SO_2 shipment emission in Great Lakes Region.

To explore the changes associated with emission reduction in the last decade, we also studied the impacts of CAGLESO2 on CN10 in NYS in 2005 and 2011. The model is run under the same settings for US case and US+CG case but using meteorology fields and emissions for years 2005 and 2011 (Table 1). As shown in Figure 4a-c, the relative changes of CN10 in NYS due to CAGLESO2 in 2005, 2011, and 2017 are 8.9%, 13.6%, and 22.4%, respectively. Comparing to 2005, the relative changes of CN10 due to CAGLESO2 are increased 1.5 times in 2011 and 2.5 times in 2017, respectively. Our analysis on the changes of absolute changes of CN10 in 2005, 2011, and 2017 due to CAGLESO2 found that the absolute changes of CN10 due to CAGLESO2 in the three years are 360.5 cm^{-3}, 435.5 cm^{-3}, and 533.5 cm^{-3}, respectively (Figures 4d-f). Comparing to 2005, the absolute changes of CN10 due to CAGLESO2 are increased 1.2 times in 2011 and 1.5 times in 2017, respectively. It indicates the increasing influnce of CAGLESO2 on CN10 in NYS is not only reflected in relative changes but also reflected in absolute changes. We also noticed that the increasing of absolute changes of CN10 from 2005 to 2017 is smaller than that of relative changes. It is because CN10 concentrations in Western NY, Finger Lakes region, Southern Tier region, and Central NY, where exhits obvious increasing of absolute changes, are decreased from 2005 to 2017 over there due to emission reduction in the Mid-Atlantic states. As shown in Figure 4g-i, CN10 concentrations in Western NY, Finger Lakes region, and Southern Tier region are decreased from 4000-5500 cm^{-3} in 2005 to 2000-3000 cm^{-3} in 2017, while CN10 concentrations in Central NY are decreased from 4000-5000 cm^{-3} in 2005 to 2000-2500 cm^{-3} in 2017. Reduced CN10 concentrations enhanced the ratio of relative change of

CN10 in 2017 to those in 2005 and 2011.

4. Conclusion

Measurements and reliable modeled concentrations of CN10 with high enough spatial and temporal resolutions are essential for the health effects studies. It is important to understand key factors controlling the spatial and temporal variations of CN10. In the Mid-Atlantic states, anthropogenic SO_2 emission was reduced from 2756 thousand tons in 2005 to 210 thousand tons in 2017 which were comparable to those emitted in Canada and Great Lakes Region. Our study found Canada anthropogenic SO_2 emission and the Great Lakes Regions shipment emission significantly enhanced CN10 in NYS by 25% and shown obvious impact on time variations of observed CN10 at NY rural and remote sites. Due to emission reduction at NYS and surrounding regions in the last decade, the impact of Canada anthropogenic SO_2 emission and the Great Lakes Regions shipment emission in NYS was increased from 8.9% in 2005 to 22.4% in 2017. As announced by White Paper on Clean Energy Standard Procurements to Implement New York's Climate Leadership and Community Protection Act, the State's electricity will be 70% from renewable sources by 2030 and 100% from renewable sources by 2040 (New York Energy Research and Development Authority and New York Department of Public Service, 2020). This action will further reduce the emission level within NYS. Our study highlights that the impact of Canada anthropogenic SO_2 emission and the Great Lakes Regions shipment emission on CN10 in NYS is expected to continuously increase in the future.

Open Research. The GEOS-Chem model is available to the public at https://geos-chem.seas.harvard.edu/. Observations at PSP and WMS are available at University at Albany Atmospheric Science Research Center's Air Quality Monitoring Products (http://atmoschem.asrc.cestm.albany.edu/~aqm/). Registration for AQM products is required at http://atmoschem.asrc.cestm.albany.edu/.

Author contributions. GL and FY developed the project idea, updated the model, and carried out the numerical simulations. Observations are from JS's group.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1. Model emission sensitivity simulation descriptions for 8 cases run in this study.

Case Name	Anthropogenic SO_2 emission in Canada	Shipment SO_2 emission in Great Lakes Region	Year
US	**No**	**No**	**2017**
US+C	**Yes**	**No**	**2017**
US+G	**No**	**Yes**	**2017**
US+CG	**Yes**	**Yes**	**2017**
$\mathbf{US_{2005}}$	**No**	**No**	**2005**
$\mathbf{(US+CG)_{2005}}$	**Yes**	**Yes**	**2005**
$\mathbf{US_{2011}}$	**No**	**No**	**2011**
$\mathbf{(US+CG)_{2011}}$	**Yes**	**Yes**	**2011**

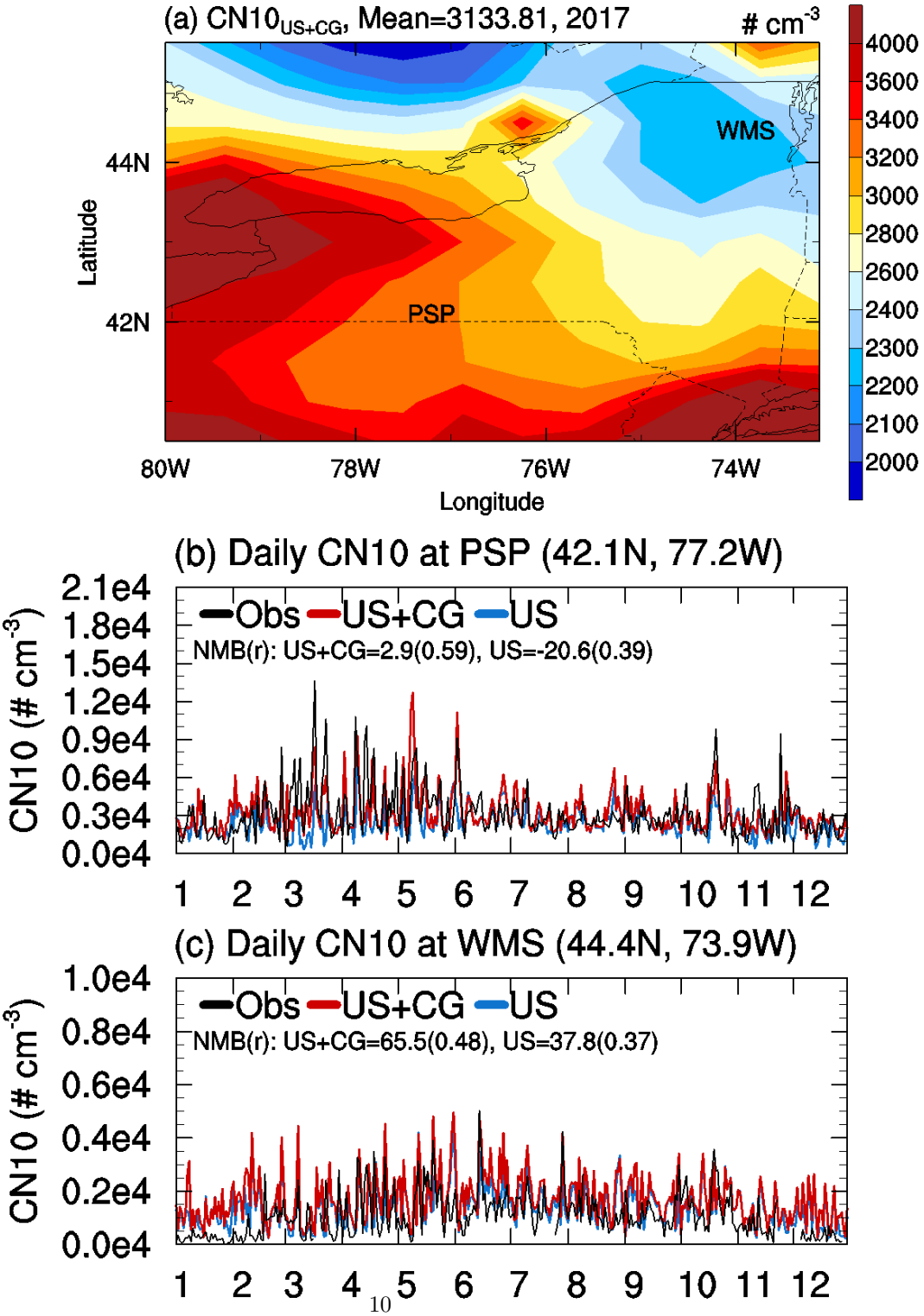
(a) CN10US+CG, Mean=3133.81, 2017
cm-3
4000
3600
3400
3200
3000
2800
2600
2400
2300
2200
2100
2000
WMS
PSP
44N
42N
Latitude
80W
78W
76W
74W
Longitude
(b) Daily CN10 at PSP (42.1N, 77.2W)
Obs US+CG US
NMB(r): US+CG=2.9(0.59), US=-20.6(0.39)
CN10 (# cm-3)
2.1e4
1.8e4
1.5e4
1.2e4
0.9e4
0.6e4
0.3e4
0.0e4
1 2 3 4 5 6 7 8 9 10 11 12
(c) Daily CN10 at WMS (44.4N, 73.9W)
Obs US+CG US
NMB(r): US+CG=65.5(0.48), US=37.8(0.37)
CN10 (# cm-3)
1.0e4
0.8e4
0.6e4
0.4e4
0.2e4
0.0e4
1 2 3 4 5 6 7 8 9 10 11 12

Figure 1. (a) Horizontal distribution of annual mean CN10 in NYS and time serieses of observed and simulated daily CN10 at (b) PSP and (c) WMS sites in 2017.

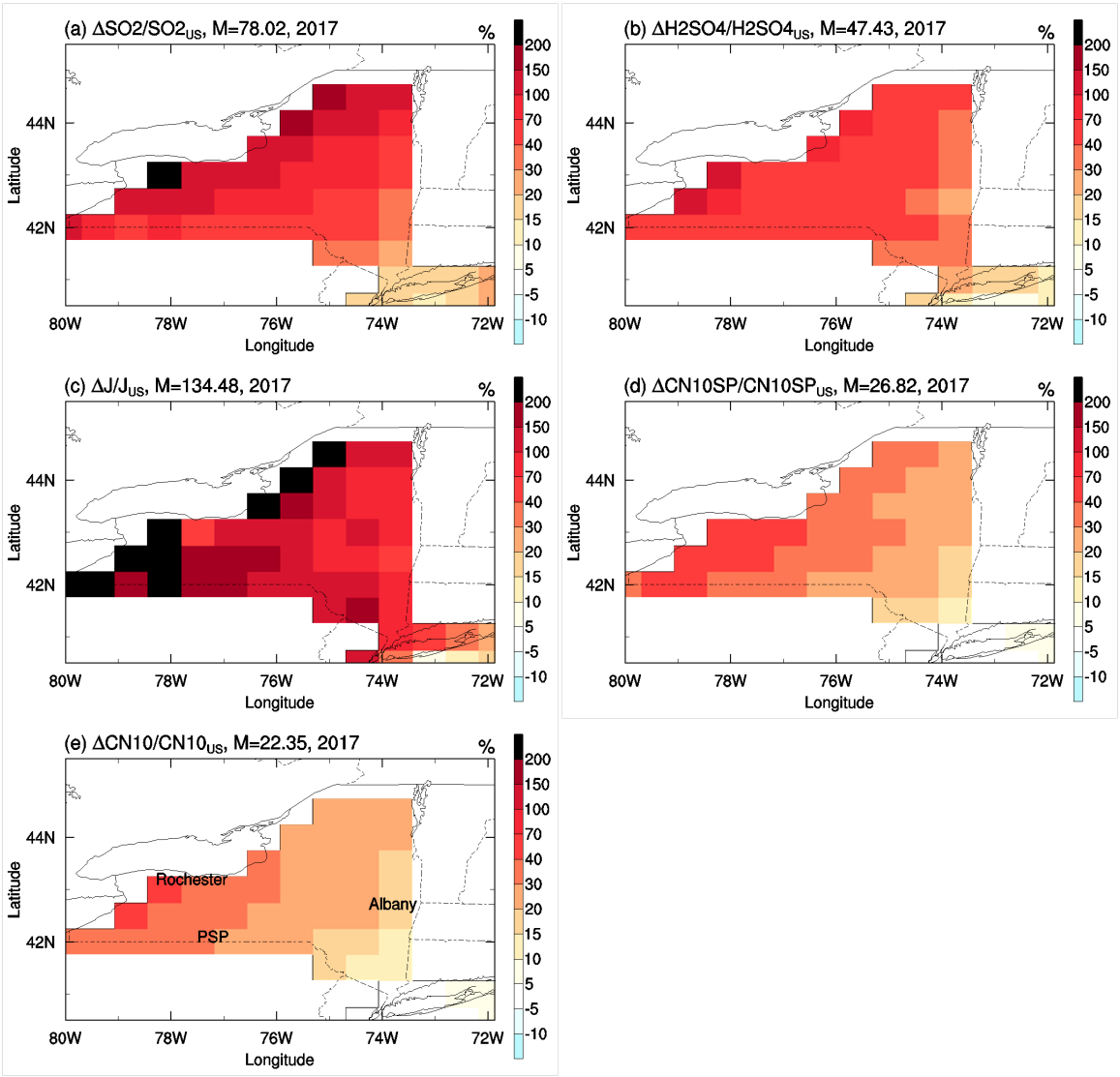

Figure 2. Horizontal distributions of relative changes of (a) SO_2, (b) sulfur acid gas, (c) nucleation rate, (d) number concentration of secondary particles larger than 10 nm, and (e) number concentration of all particles larger than 10 nm in NYS due to Canada anthropogenic SO_2 emission and Great Lakes Regions shipment SO_2 emission. Grids out of NYS are excluded.

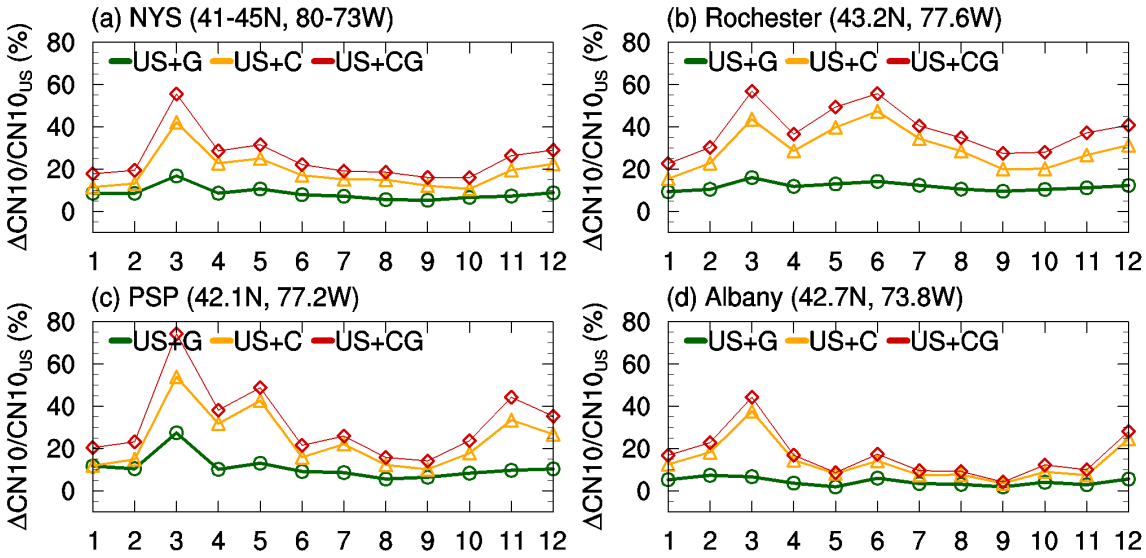

Figure 3. Monthly mean of relative changes of CN10 in (a) NYS (domain mean within the ranges of 40.5-45.5°N and 80-72°W excluding grids out of NYS), at (b) Rochester, (c) PSP, and (d) Albany due to Great Lakes Region shipment SO_2 emission (green), Canada anthropogenic SO_2 emission (orange), and both of the two (red).

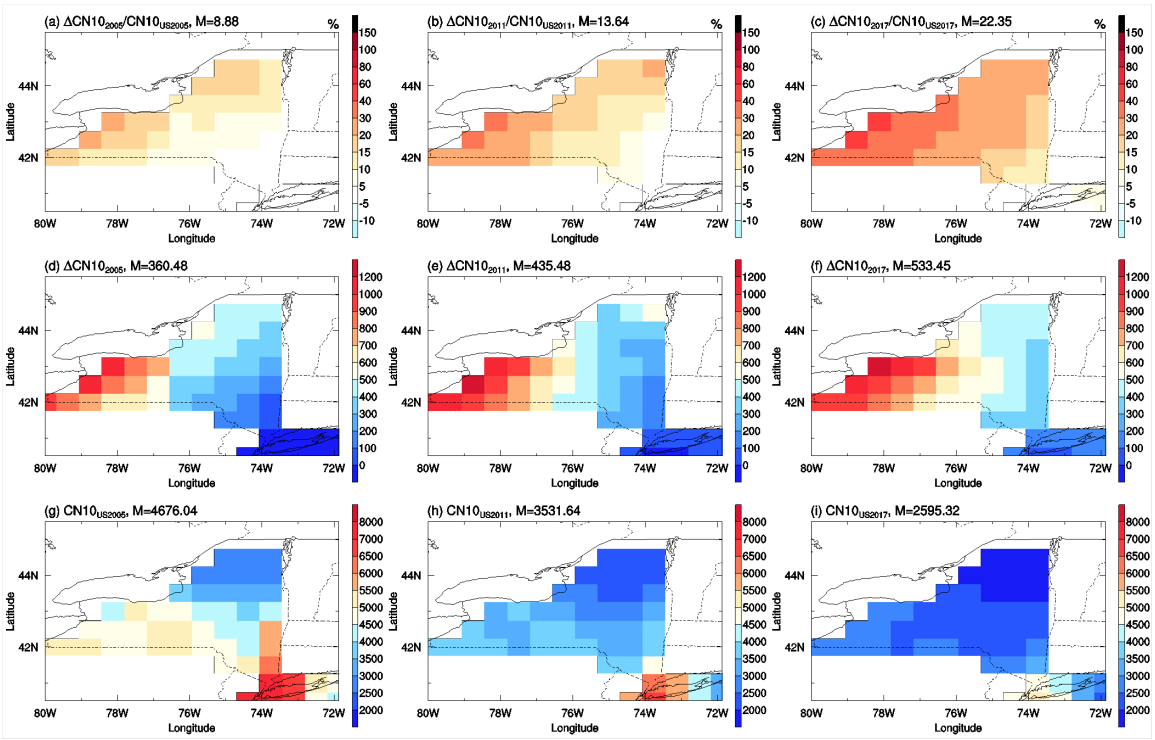

Figure 4. Horizontal distributions of relative change of CN10 in (a) 2005, (b) 2011, and (c) 2017 due to Canada anthropogenic SO_2 emission and Great Lakes Regions shipment SO_2 emission. (d-f) the same as (a-c) but for absolute change of CN10. Horizontal distributions of CN10 in (g) 2005, (h) 2011, and (i) 2017. Grids out of NYS are excluded.