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LETTER

Increasing influence of Canadian anthropogenic and the Great Lakes Region shipment SO\textsubscript{2} 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 matter 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 CN\textsubscript{10} (condensation nuclei > 10 nm) at rural and remote sites in NYS, we show that Canadian anthropogenic and the Great Lakes Regions shipment SO\textsubscript{2} emission (CAGLESO\textsubscript{2}) significantly influenced CN\textsubscript{10} in NYS. These emissions on average produced a 22\% enhancement of CN\textsubscript{10} in NYS in 2017, varying from 40\% in Northwestern NYS to 10\% in Southeastern NYS. We also found that the impact of CAGLESO\textsubscript{2} on NYS’s CN\textsubscript{10} in 2017 was 2.5 times higher than that in 2005 and 1.6 times higher than that in 2011, which indicated increasing influence of CAGLESO\textsubscript{2} on CN\textsubscript{10} in NYS 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 PM\textsubscript{10} and PM\textsubscript{2.5}. Prior studies found ultrafine particles have several more aggressive health implications than PM\textsubscript{10} and PM\textsubscript{2.5} (Oberdörster et al 1994, Yacobi et al 2010, Schraufnagel 2020). Ultrafine particles contribute little to the total particle mass but comprise an estimated 90\% of particle number concentration (Institute, H E 2013). Its number concentrations can be represented by condensation nuclei measurements (Matson 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\textsubscript{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\textsubscript{2} emission was reduced 83\% from 2005 to 2017. In the meantime, anthropogenic SO\textsubscript{2} emissions decreased from 387,000 tons to 26,000 tons in NYS (93\% reduction), from 2,756,000 tons to 210,000 tons in the Mid-Atlantic states (92\% reduction), from 758,000 tons to 291,000 tons in southeastern Canada (62\% reduction), and from 148,000 tons...
to 141,000 tons in the Great Lakes Region (5% reduction), respectively (figure S1 (available online at stacks.iop.org/ERC/4/071003/mmedia)). The differences between the outcomes of regulations in US and Canada were caused by the differences between US and Canada air quality standards and their implementation time (table S1). As shown in figure S1c and figure S1d, SO2 emission reduction in the Mid-Atlantic States from 2005 to 2009 was 43% which was close to the value of SO2 emission reduction in southeastern Canada (52%). However, in 2010, USEPA revised the primary SO2 standard. This action resulted in a further SO2 emission reduction in US from 2010 to 2017 (83%). At the same time, SO2 emission in southeastern Canada was reduced by 31%. About half of this SO2 emission reduction in southeastern Canada happened in 2017, which was due to the implementation of new and more stringent Canadian Ambient Air Quality Standards for SO2 announced in 2016. Great Lakes shipment SO2 emission only changed 5% from 2005 to 2017. It is because the compounded annual growth rates in marine vessel link-level tonnages in the Great Lakes region were relative stable during these years (ENERCON Services, Inc 2015). In 2017, anthropogenic SO2 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 SO2 emission was 5.4 times higher than that in NYS and 67% of that in the Mid-Atlantic States. As reported by USEPA (https://www.epa.gov/air-trends/sulfur-dioxide-trends, last access July 2022), after 2004, the mean concentration of SO2 observed at 31 EPA Northeast sites has been lower than National standard. After 2011, the 90th percentile SO2 observed at 31 EPA Northeast sites has been lower than National standard. While SO2 pollution is no longer a problem in the region, it is an important precursor of new particle formation which dominates particle number concentration in rural and remote regions (Kulmala et al 2006, Yu and Luo 2009, Merikanto et al 2009, Kulmala et al 2014, Yu et al 2015). SO2 is oxidized into sulfur acid gas which is the primary vapor responsible for atmospheric nucleation (Sipilä et al 2010, Lee et al 2019). Luo and Yu (2011) found that CN10 in the Northeastern US is dominated by secondary particles formed via atmospheric nucleation associated with SO2. It is important to know the potential impacts of Canadian anthropogenic and the Great Lakes Regions shipment SO2 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 (Adams and Seinfeld 2002, Spracklen et al 2005, Zaveri et al 2008, Yu and Luo 2009) 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 Finnace 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. Methods

2.1. Atmospheric chemistry and aerosol microphysics simulation

GEOS-Chem is a widely used global 3D 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 (Bey et al 2001, Keller et al 2014, Luo et al 2019, Holmes et al 2019, Luo et al 2020). In this study, GESO-Chem is driven by MERRA-2 reanalysis data product. NOX–Ox–hydrocarbon–aerosol scheme is run to simulate atmospheric chemistry. The Advanced Particle Microphysics package (Yu and Luo 2009) is used to simulate aerosol microphysics. We run the model with version 12.6.0 at 0.5° × 0.625° horizontal resolution for a nest domain (34°N to 52°N; 97°W to 65°W) whose boundary conditions were provided by a 4° × 5° global simulation. Eight emission sensitivity cases were run to investigate the impact of CAGLESO2 on CN10 in NYS (table 1). Model outputs at PSP and WMS sites by nest simulation are every 10 min.

2.2. CN 10 number concentration observation

In this study, we used CN10 observations at PSP and WMS sites to evaluate model performance. The PSP site (42.09°N and 77.21°W) is located in Addison, NY, a village in southwestern NY. The WMS site (44.37°N and 73.9°W) 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.
3. Observed and simulated CN10 at NYS rural and remote sites

Figure 1 (a) 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 figures 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 oxidation of SO$_2$ during winter. Mean absolute errors (MAE) of simulated CN10 by US + CG case and US case are 1137 cm$^{-3}$ and 1222 cm$^{-3}$, respectively. MAE of US + CG case is 7% lower than MAE of US case. 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 observation in March. Some peaks of CN10 observed during spring are missed by the US case. Standard deviations (SD) of CN10 by observations, US + CG case, and US case are 1893 cm$^{-3}$, 1667 cm$^{-3}$, and 1171 cm$^{-3}$, respectively. SD of US + CG case is 1.4 times higher than SD of US case and is closer to SD of observation. 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 1(c)). WMS is perched high atop the Whiteface Mountain summit, where observation is sensitive to air mass carrying air pollutants. 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. MAEs of CN10 at WMS by US + CG case and US case are 922 cm$^{-3}$ and 781 cm$^{-3}$, respectively. MAE of US + CG case is 18% higher than MAE of US case. 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. SD of CN10 at WMS by observation, US + CG case, and US case are 831 cm$^{-3}$, 914 cm$^{-3}$, and 781 cm$^{-3}$, respectively. SD of US + CG case is 10% higher than SD of observation, while US case and is 6% lower than SD of observation. 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 Canadian anthropogenic SO$_2$ emission and the Great Lakes Region shipment SO$_2$ emission play important roles in CN10 time variations at the two sites.

4. 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$_2$SO$_4$]), 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 2(a), 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

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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 |
| US$_{2005}$ | No                                     | No                                            | 2005 |
| (US + CG)$_{2005}$ | Yes                                    | Yes                                           | 2005 |
| US$_{2011}$ | No                                     | No                                            | 2011 |
| (US + CG)$_{2011}$ | Yes                                    | Yes                                           | 2011 |
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 (Kulmala et al 2006, Yu et al 2015). Figure 2(b) shows that \([\text{H}_2\text{SO}_4]\) is enhanced 47.4% in NYS and has a similar spatial pattern to that of \([\text{SO}_2]\). Increased \([\text{H}_2\text{SO}_4]\) in NYS significantly enhances nucleation rate at corresponding regions and results in a 134.5% enhancement of nucleation rate in NYS (figure 2(c)). The impact of CAGLESO2 on CN10SP in NYS is shown in figure 2(d). 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 2(e) shows the changes of CN10. CN10 in NYS includes both secondary and primary particles. Although CN10 in NYS is dominated by secondary particles (Luo and Yu 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 (43.16°N, 77.61°W), 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 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 Canadian 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), Canadian anthropogenic SO$_2$ emission (orange), and both of the two (red).
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 decreases 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 at Rochester, PSP, and Albany caused by Canadian anthropogenic SO2 emission are 2.5, 2.2, and 3.2 times higher than those caused by Great Lakes Region shipment SO2 emission, respectively. Rochester and PSP are close to Great Lakes. Contributions of Great Lakes Region shipment SO2 emission to CN10 enhancement at the two places are 28.6% and 31.3%, respectively. Although Rochester is located at the lakeside of Lake Ontario, due to large Canadian anthropogenic SO2 emission source at Hamilton and Toronto, contribution of Great Lakes Region shipment SO2 emission at Rochester is lower than that at PSP. At Albany, where is far away from Great Lakes, contribution of Great Lakes Region shipment SO2 emission to CN10 enhancement is 23.8% which is lower than the contributions at Rochester and PSP. The enhancements of CN10 in NYS caused by Canadian anthropogenic SO2 emission is 2.2 times higher than those caused by Great Lakes Region shipment SO2 emission. It indicates anthropogenic SO2 emission in Canada has a stronger impact on CN10 in NYS than SO2 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 annual averaged 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 figures 4(a)–(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 361 cm$^{-3}$, 436 cm$^{-3}$, and 534 cm$^{-3}$, respectively (figures 4(d)–(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 influence 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 exits 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 figures 4(g)–(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. 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 State Energy Research and Development Authority and New York Department of Public Service 2020). This action will further reduce the emission level within NYS. When CN10 concentrations in NYS is reduced continuously due to these new energy and emission policies, CAGLESO2 may become a major source of CN10 concentrations in NYS. It implies the combined action via the Canada-United States Air Quality Agreement to reduce CAGLESO2 can help to reduce ultrafine particle exposure level and then benefit residents’ health in NYS.

5. 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 2756,000 tons in 2005 to 210,000 tons in 2017 which were comparable to those emitted in Canada and Great Lakes Region. Our study found Canadian 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 Canadian 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. Our study shown that upwind SO$_2$ emission source can impact downwind region ultrafine particle number concentration level. When health risk of exposure to high levels of ultrafine particles becomes an issue, people need to pay attention to both local and upwind region’s SO$_2$ emissions. Our study highlights that the impact of Canadian anthropogenic SO$_2$ emission and the Great Lakes Regions shipment emission on CN10 in NYS is expected to continuously increase in the future. The combined action via the Canada-United States Air Quality Agreement to reduce CAGLESO2 can help to reduce ultrafine particle exposure level and then benefit residents’ health in NYS.

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Supplementary material for this article is available online

Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

Author contributions

GL and FY developed the project idea, updated the model, and carried out the numerical simulations. Observations are from JS’s group.
Conflict of interest

The authors declare no competing interests.

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