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# Regional impacts of ultrafine particle emissions from the surface of the Great Lakes

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## Abstract

Quantifying the impacts of aerosols on climate requires a detailed knowledge of both the anthropogenic and the natural contributions to the aerosol population. Recent work has suggested a previously unrecognized natural source of ultrafine particles resulting from breaking waves at the surface of large freshwater lakes. This work is the first modeling study to investigate the potential for this newly discovered source to affect the aerosol number concentrations on regional scales. Using the WRF-Chem modeling framework, the impacts of wind-driven aerosol production from the surface of the Great Lakes were studied for a July 2004 test case. Simulations were performed for a base case with no lake surface emissions, a case with lake surface emissions included, and a default case wherein large freshwater lakes emit marine particles as if they were oceans. Results indicate that the lake surface emissions can enhance the surface level aerosol number concentration by  $\sim 20\%$  over the remote northern Great Lakes and by  $\sim 5\%$  over other parts of the Great Lakes. These results were highly sensitive to the nucleation parameterization within WRF-Chem; when the nucleation process was deactivated, surface-layer enhancements from the lake emissions increased to as much as  $200\%$ . The results reported here have significant uncertainties associated with the lake emission parameterization and the way ultrafine particles are modeled within WRF-Chem. Nevertheless, the magnitude of the impacts found in this study suggest that further study of this phenomena is merited.

## 1 Introduction

Understanding and quantifying the impacts of aerosols on the atmospheric radiative balance is one of the most significant outstanding problems in climate change science (IPCC, 2007). Aerosols can impact the radiative balance directly through their scattering and absorbing properties, and indirectly by influencing the formation and properties of clouds. Much of the uncertainty associated with these so-called direct and indirect

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aerosol effects results from an insufficient knowledge of the aerosol population itself. A particle's optical properties and ability to form a cloud droplet are each strongly dependent on its physical and chemical properties. Scaling these dependencies up to the aerosol population, i.e., estimating the overall radiative forcing due to the total aerosol present, requires a detailed knowledge of the aerosol size distribution, size-resolved composition, mixing state, and hygroscopic properties. Directly acquiring such detailed knowledge is challenging even during intensive measurement campaigns; amassing a sufficiently complete data set on regional scales or beyond will not be achievable for the foreseeable future. The best available alternative at present is to rely on large-scale models to assimilate the available information of aerosol sources, sinks, and atmospheric transformations and then verify these models with atmospheric observations.

To be successful in building a reliable large-scale atmospheric aerosol model, the natural sources of particulate material must be included as well as the anthropogenic ones. In part this is because after atmospheric aging it is difficult to distinguish anthropogenic and natural aerosol material- validation is most easily approached by considering the sum of all sources. Understanding both the natural and anthropogenic contributions to the aerosol population is especially important when considering aerosol climate impacts, because the most relevant parameters for climate studies are the *changes* to radiative forcing and the subsequent feedbacks. The relationship between an increase in aerosol loading and the resultant radiative forcing is complex and non-linear; understanding the pre-existing natural state of the atmosphere is critical to determining how anthropogenic pollution perturbs the system.

While the sum of natural particle sources far exceeds the total anthropogenic contribution, with some notable exceptions (e.g., volcanoes and forest fires) natural sources tend to be spatially and temporally distributed rather than intense. Natural emissions fluxes are characteristically smaller than anthropogenic ones, but are spread over large areas and can persist for long periods of time. Examples of these "gentle" natural particle sources include marine emissions of sea salt, windblown dust, and pollen releases from forests and grasslands. Despite their low intensity, it is highly likely that all of the

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major sources of natural aerosols – those that have impacts on global scales – have already been identified if not fully quantified.

However, because the emissions intensity of natural sources can be so small, it does still happen that natural particle sources of potential regional importance are discovered from time to time. For example, O'Dowd et al. (1998) observed strong new particle formation events along the Ireland coast under low tide conditions; later work showed conclusively that these particles were the result of the photolysis of iodine-containing compounds emitted from macroalgae exposed during low-tide (O'Dowd et al., 2002). The relatively recent discovery that isoprene can form secondary organic aerosol in the atmosphere is another example. Isoprene is the most abundant volatile organic compound emitted from the biosphere, but for many years the scientific consensus was that its aerosol-forming potential was negligible (Pandis et al., 1991). This consensus was challenged when Claeys et al. (2004) found unexpected evidence of 2-methyltetrols in aerosol samples collected in the Amazon; the most likely mechanism for producing the compounds in the atmosphere is via the photooxidation of isoprene. Focused studies have since shown that isoprene oxidation can yield SOA via both gas-phase and aqueous mechanisms, and it is now believed that isoprene is a significant source of organic aerosol material in many areas of the globe (Henze and Seinfeld, 2006; Kroll et al., 2006). A thorough discussion of these and many other natural sources of particulate material in the atmosphere can be found in the review by Carslaw et al. (2010).

Slade et al. (2010) recently described another new source of natural particles that is potentially significant on regional scales – the production of nanoparticles from breaking waves at the surface of freshwater lakes. In Summer 2009, the Purdue University Airborne Laboratory for Atmospheric Research (ALAR) research aircraft conducted several research flights over the northern parts of Lakes Michigan and Huron in support of the Community Atmosphere-Biosphere Interactions Experiment (CABINEX). The intended purpose of the flights was to determine the “background” condition of the regional aerosol prior to air mass advection over the main CABINEX ground site at the Program for Research on Oxidants: Photochemistry, Emissions, and Transport

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(PROPHET) facility at the University of Michigan Biological Station (Carroll et al., 2001). Surprisingly, more than just a regional background aerosol was observed during these flights. On several occasions elevated concentrations of nanoparticles were found over the lakes in a distinct ~15–40 nm mode that was usually not present at the PROPHET site. The available evidence strongly suggests that these particles were the result of natural, wave breaking processes at the lake surface. As described by Slade et al. (2010), the particles were only observed when whitecap waves were present, the concentration of the 15–40 nm mode was in most cases largest just above the surface, and the concentration of that mode increased exponentially with surface wind speed. Furthermore, a chemical analysis of the lake water indicated that the concentrations of trace constituents were cumulatively sufficient to produce a dry particle mode in the 15–40 nm size range if the size distribution of water-containing droplets emitted from the lake surface were similar to that observed in marine environments.

While the particle production mechanism proposed by Slade et al. (2010) is interesting, it has not yet been definitively established – that would require a more targeted experiment with a more robust set of observations. Moreover, it is not yet clear that the proposed lake surface aerosol source is large enough to be atmospherically relevant on a regional scale. As noted in that initial study, a reasonable estimate of the particulate mass expected from a lake surface source would only be a tiny fraction of the primary emissions from the upwind region; there would be a similarly small relative contribution to specific hazardous pollutants to the atmosphere. These are intuitive results – the particles resulting from the proposed lake surface source have very small diameters and thus have very little mass.

On the other hand, Slade et al. (2010) did indicate that the lake surface source was potentially significant in terms of particle number, comparable to the low end of the range of atmospheric new particle formation rates. Nilsson et al. (2001) measured marine particle fluxes on the order of  $10^6 \text{ m}^{-2} \text{ s}^{-1}$  at atmospherically relevant conditions (wind speed =  $10 \text{ m s}^{-1}$ , height = 10 m); averaged through the lowest 100 m of the atmosphere, this would correspond to a new particle source rate of  $\sim 0.01 \text{ cm}^{-3} \text{ s}^{-1}$ . This

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“equivalent” source rate is only a rough estimate, but it is consistent with the low end of range of new particle formation rates reported by Kulmala et al. (2004) and it is reasonable to speculate that wind-driven surface emission events could last longer and thus have similar overall impact to chemically-driven new particle formation events. This is key, because several studies have demonstrated that atmospheric new particle formation can influence the aerosol indirect climate effect by increasing cloud condensation nuclei (CCN) concentrations (e.g., Kuang et al., 2009; Merikanto et al., 2009; Pierce and Adams, 2009; Yu and Luo, 2009). The mechanism for the nanoparticles derived from lake surfaces to impact climate would be similar – they would potentially impact how the aerosol population evolves due to condensational growth and its cloud forming potential. If the lake surface emissions mechanism is valid, these impacts would exist, but it is not clear whether they would be important on the temporal and spatial scales relevant to regional climate.

This study examines the potential for a lake-surface nanoparticle source to affect the aerosol population on regional scales. With finer spatial and temporal scales than those of global climate models, mesoscale models are better suited for a detailed analysis of regional aerosol emissions and processing. Using the WRF-Chem mesoscale model, we looked at the impacts of wind-driven lake surface nanoparticle emissions on total aerosol number concentrations for an exemplary case in the Great Lakes region. First, for a detailed analysis, a period was selected that is characterized by high surface winds over an extended (~several hour) time period, a relatively clean background condition, and low precipitation. Then, a two-week period was evaluated to characterize the persistence of wind-driven lake surface nanoparticle emissions on a regional scale. Cases were simulated both with and without parameterized emissions of nanoparticles from the lake surface, and analysis focused on the changes to the aerosol population on the regional scale. The study concludes with a discussion of the potential impacts of the modeled changes to the aerosol population and of the model improvements needed to enable a more rigorous study of the impact of wind-generated aerosols on aerosol-cloud interactions.

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## 2 Method

### 2.1 WRF-Chem

For the study we employed version 2.2 of the mesoscale Weather Research and Forecasting model with online Chemistry (WRF-Chem). The model is based on version 2 of the non-hydrostatic WRF community model, developed at the National Center for Atmospheric Research in collaboration with several other research institutions (<http://www.wrf-model.org>). WRF-Chem is an operational forecasting model that is designed to be computationally efficient and flexible for scientific research. One of the most advanced aspects of WRF-Chem is that its architecture allows for consistency between the chemical and meteorological modeling components such that they both use the same time step, horizontal and vertical resolutions, transport schemes, and physics schemes for subgrid-scale processes. This “online” feature within WRF-Chem allows for the study of aerosol-cloud-radiation-meteorology interactions and feedbacks (e.g., Gustafson et al., 2007; Ntelekos et al., 2009; Zhang et al., 2010b; Zhao et al., 2010). The general performance of WRF-Chem has been well-documented (e.g., McKeen et al., 2009; Zhao et al., 2010), including for the northeastern United States for the period used for this study (McKeen et al., 2007).

To accurately simulate all of the meteorological, chemical, and aerosol processes over urban to regional scales that can affect aerosol-cloud-radiation interactions is still a challenge, despite the recent advances in incorporating aerosol dynamics models into three-dimensional regional and global models. Nevertheless, the actively supported community WRF-Chem model provides a useful tool for the systematic analysis of aerosol impacts on radiation and clouds and for evaluating our current understanding of aerosol formation and transformation processes in the atmosphere. For this initial investigation on the potential significance of lake-surface nanoparticles, we have not considered aerosol-cloud-radiation interactions; instead, this study focuses on the impact wind-generated ultrafine particles from the Great Lakes on the aerosol number concentrations as a proxy for their potential impact on regional climate.

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The meteorological dynamics and physics options included in WRF have been described by Skamarock et al. (2005). Unless otherwise noted, WRF physics options used for this study include the following: the Goddard shortwave radiation scheme (Chou and Suarez, 1994), the RRTM longwave radiation scheme (Mlawer et al., 1997), the Yonsei University (YSU) planetary boundary layer (PBL) scheme (Hong and Pan, 1996), the Noah land-surface model (Chen and Dudhia, 2001), the Grell-Devenyi cumulus cloud parameterization (Grell and Devenyi, 2002), and the WSM 5 microphysics scheme (Hong et al., 2004). For WRF-Chem, several modules for gas- and aerosol-phase chemistry and dynamics have been incorporated into WRF; a general description of WRF-Chem is provided by Grell et al. (2005), and the incorporation of the chemistry and aerosol modules used for this study is explained in Fast et al. (2006). Here, only the components of WRF-Chem relevant for this study will be discussed.

The WRF-Chem gas-phase chemistry and aerosol modules were used to simulate aerosol concentrations and size distributions for July 2004 in the eastern United States so that we could characterize the potential for particle emissions from the Great Lakes surface to impact the regional aerosol population. The gas-phase chemistry and aerosol modules employed for this study are the Carbon Bond Mechanism Z (CBM-Z) (Zaveri and Peters, 1999) and the Model for Simulation Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2008), respectively. Both modules had been previously integrated into the WRF framework (Fast et al., 2006). This version of WRF-Chem with CBM-Z and MOSAIC includes inorganic aerosol components and primary organic and elemental carbon; secondary organic aerosol (SOA) is not included.

To explicitly model the aerosol size distribution, the MOSAIC aerosol module in WRF-Chem uses a two-moment sectional approach based on the work of Tzivion et al. (1989) for aerosol coagulation, condensational growth and evaporation. In the two-moment sectional algorithm, concentrations of both aerosol number and mass are simulated prognostically for each bin so that total mass (gas + aerosol) and aerosol number are conserved simultaneously during condensation and evaporation. The MOSAIC aerosol module within WRF-Chem models particles with dry diameters between



40 nm and 10  $\mu\text{m}$ , with the size range divided into either four or eight size bins. The option of eight size bins is chosen for this study, with the dry particle diameters doubling for each successive bin boundary. Within each bin, particles are assumed to be internally mixed and can be composed of sulfate, nitrate, ammonium, other (unspecified) inorganic material, primary organic carbon, elemental carbon, and water. Rates of condensation and evaporation for trace gases ( $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$ , and  $\text{NH}_3$ ) are based on particle size and equilibrium concentrations determined from MTEM (Zaveri et al., 2005a) and MESA (Zaveri et al., 2005b) thermodynamics models. MOSIAC simulates Brownian coagulation using the method of Jacobson et al. (1994) For new particle formation, the model uses the  $\text{H}_2\text{SO}_4$ - $\text{H}_2\text{O}$  binary nucleation scheme of Wexler et al. (1994); newly-formed particles are assumed to have the smallest diameter tracked by the model (40 nm) and  $\text{H}_2\text{SO}_4$  is assumed to be completely neutralized by  $\text{NH}_3$  if a sufficient amount exists in the gas phase.

## 2.2 Baseline emissions

The emissions inventory used in this study was described by Kim et al. (2006). The portion of the inventory covering United States emissions was updated from the US EPA NEI99 (National Emissions Inventory, base year 1999) versions 3 (<http://www.epa.gov/ttn/chief/net/1999inventory.html#final3.crit>). Details of the NEI99 inventory and its implementation within WRF-Chem are discussed in detail by Frost et al. (2006). The emissions inventory was initially constructed to model a typical 1999 ozone-season day using data provided by the EPA. The updated inventory includes the reported point source emissions of  $\text{NO}_x$  (64% of the total NEI99  $\text{NO}_x$  point emissions) and  $\text{SO}_2$  (80% of NEI99  $\text{SO}_2$  point emissions) from nearly 1000 individual monitoring locations within the Continuous Emissions Measurement System (CEMS) network for the summer of 2004. Emissions from point sources included in NEI99 but not in the CEMS dataset were kept at their 1999 levels. On-road, off-road, and area emissions from NEI99, as well as the point source emissions of compounds other than  $\text{NO}_x$  and  $\text{SO}_2$ , were not modified and remain at 1999 ozone season

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day levels. The revised inventory developed by Frost et al. (2006) also included the 1995 Canadian province- and census-division-level area and mobile emissions available from EPA (<ftp://ftp.epa.gov:EmisInventory/canadaSMOKE>) and the 1999 Mexican state-level area source emissions north of 24° N compiled for the BRAVO study (<http://www.epa.gov:/ttn/chief/net/mexico.html>).

### 2.3 Lake surface emissions

At present, there are not enough data available to develop a parameterization for estimating the wind-driven particle emissions from freshwater lake surfaces; the measurements described by Slade et al. (2010) are insufficient. However, a reasonable approximation of the lake surface particle emissions rate can be drawn from marine particle flux measurements. As has been described in detail by recent reviews of marine aerosol production (Lewis and Schwartz, 2004; O'Dowd and De Leeuw, 2007), the great majority of the particle number production from the ocean surface is due to the wind-driven entrainment and subsequent bursting of small air bubbles in breaking waves. The bursting of these bubbles result in the production of numerous small droplets from the rupture of the bubble film, and the ejection of a smaller number of somewhat larger droplets from a pressure-induced jet. After emission, droplets produced by either mechanism will rapidly equilibrate to the ambient relative humidity, resulting in a emissions spectra whose concentration is related chiefly to wind speed and whose size distribution is related chiefly to the concentration of trace species at the water surface.

While evidence suggests that the presence of surface-active organic species in the surface layer has some effect on wind-driven droplet production, to a first approximation the number of droplets produced from a freshwater surface should be very similar to the number produced from a marine surface. This is true even though the size distribution of the resultant equilibrated particles would be quite different from a freshwater lake because of the vastly different concentrations of dissolved material in saltwater and fresh water. If we assume that the droplet number production does not depend on the

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lake surface composition, we can model the particle number flux from the Great Lakes using the particle number flux parameterizations derived by Geever et al. (2005):

$$\log(F_{10\text{nm}}) = 0.099U_{22} - 0.73 \quad (1)$$

$$\log(F_{100\text{nm}}) = 0.109U_{22} - 1.19 \quad (2)$$

Here,  $U_{22}$  is the horizontal wind speed at 22 m (in  $\text{m s}^{-1}$ ) and  $F_{10\text{nm}}$  and  $F_{100\text{nm}}$  are the particle source fluxes (in  $10^6 \text{ m}^{-2} \text{ s}^{-1}$ ) for particles with diameters larger than 10 nm and larger than 100 nm, respectively. These parameterizations were based on measurements conducted at Mace Head, Ireland; they were consistent with previous measurements in the north Atlantic (Nilsson et al., 2001) and an earlier parameterization proposed by Mårtensson et al. (2003) (O'Dowd and De Leeuw, 2007). O'Dowd et al. (2008) recently suggested an updated parameterization for  $F_{100\text{nm}}$  based on the same data, but no complementary update is available for  $F_{10\text{nm}}$ .

For this study we have used the  $F_{10\text{nm}}$  parameterization developed by Geever et al. (2005) (Eq. 1) to describe the particle number emissions flux from the Great Lakes. We chose to use the  $F_{10\text{nm}}$  parameterization rather than the one for  $F_{100\text{nm}}$  with the assumption that the former estimate best represents the total number of wind-driven droplets released by the water surface, and is therefore a better upper bound estimate of the total number flux of particles from the surface after water vapor equilibration. This parameterization requires 22-m horizontal wind speed, which is not a standard diagnostic parameter within WRF-Chem; for this study wind speeds at 22 m have been calculated using similarity theory and the surface-level wind speeds, following the methodology for calculating 10-m wind fields by the land-surface models in WRF.

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## 2.4 Simulations

The goal of this study is to characterize the potential for lake-derived particles to impact the regional aerosol population. These potential impacts can be evaluated both on short time-scales after a meteorological “event” and by examining the average impacts over longer time periods. The greatest short-term impacts should occur when two meteorological conditions are met: (1) the presence of high winds over the lake surface, which would lead to the most intense lake surface emissions; and (2) the near-absence of precipitation in the region, which would minimize the effectiveness of wet deposition as a removal mechanism for the freshly-emitted particles. To find a suitable case that met these conditions, we revisited a set of 24-h retrospective simulations that had been performed for 12–31 July 2004 for a domain covering the eastern United States at 27-km × 27-km horizontal resolution with 34 vertical layers (McKeen et al., 2007) (Fig. 1a). These simulations were initialized at 00:00:00 UTC each day with National Centers for Environmental Prediction (NCEP) Global Forecast System (GFS) model analysis fields. The WRF results from these existing simulations were reanalyzed for a sub-domain that included the most of the Great Lakes region (Fig. 1b). Lake Ontario was not included in the analysis domain because of its small size relative to the northern Great Lakes and because it is more heavily influenced by anthropogenic sources, suggesting that the impact of lake aerosols would be small. In our reanalysis 14 July 2004 emerged as a suitable test case – on this day there was an extended period of strong surface winds over the northern Great Lakes, especially Lake Huron, and limited regional precipitation (Fig. 2). Moreover, the wind event on 14 July was relatively isolated- winds were calmer on the preceding and following days which allowed to more easily focus on the impacts of the event for analysis.

Several sets of simulations were performed, with three cases for each set labeled DEFAULT, BASE, and LAKE (Table 1). The DEFAULT cases use the emissions parameterizations already included in version 2.2 of WRF-Chem and that are still used in the most recent publicly released version of the model (version 3.3). In its publicly released

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form, the WRF model does not distinguish between ocean and freshwater bodies, and so in the DEFAULT cases all water surfaces are treated as if they were ocean. As a result, the Great Lakes are emissions sources for large sea-salt particles based on the parameterization of Gong et al. (1997). For the BASE cases, WRF-Chem was modified so that there are no emissions from the lakes whatsoever by turning the sea-salt parameterization for the Great Lakes sub-domain off in the model (any sea-salt emissions from the Atlantic Ocean were unaffected). In the LAKE cases, ultrafine-particle emissions from the surface of the Great Lakes were estimated using the  $F_{10\text{nm}}$  parameterization developed by Geever et al. (2005) (Eq. 1). All particles emitted from the lake surface were assumed to have dry diameters of 40 nm and were initially placed in the smallest of the eight size bins included in the MOSAIC aerosol dynamics module in WRF-Chem. This approximation implicitly assumes that the emitted particles are somewhat larger than what was observed by Slade et al. (2010), but this simplification is not expected to significantly impact the aerosol number concentration results.

The DEFAULT, BASE, and LAKE cases were repeated with several combinations of aerosol dynamics processes (nucleation, coagulation, and dry deposition) turned on and off in order to determine the sensitivity of the results to other processes affecting aerosol number concentration. Because modeled wind speeds can be sensitive to the boundary-layer scheme used, simulations using the Mellor-Yamada-Janjic (MYJ) (Mellor and Yamada, 1982; Janjic, 2002) instead of the YSU boundary layer scheme were also performed. Most WRF-Chem simulations were conducted for 14 July 2004 only, to investigate the short-term impact of lake aerosols (with 12–13 July simulations of the BASE cases used for model spin-up). Some of the simulations were extended to 27 July to investigate the longer-term average impacts.

### 3 Results

#### 3.1 Episodic Impacts on 14 July 2004

14 July 2004 was chosen for the episodic event study because the WRF model results for that day meet the two criteria that were presumed necessary for the lake surface source to contribute significantly to the aerosol population – high surface wind speeds and the absence or near-absence of precipitation over and downwind of the Great Lakes. Figure 2 shows the modeled 22-m wind speed and aerosol number flux, with wind vector overlays, for 14 July 2004 at 12:00:00 GMT (08:00 a.m. Eastern Daylight Time). This was the time of highest modeled wind speeds during this day. The results indicate that during this period winds were generally northerly to northwesterly over most of the Great Lakes region except for over Lake Erie, where the winds were turning westerly. Wind speeds were greater than  $8 \text{ m s}^{-1}$  over most of the Great Lakes and greater than  $15 \text{ m s}^{-1}$  over Lake Huron (Fig. 2a). These wind speeds led to particles being emitted from the lake surface at rates on the order of a few  $10^6 \text{ m}^{-2} \text{ s}^{-1}$ . The areas of greatest particle emissions during the 14 July 2004 event were the southeast part of Lake Superior, western Lake Huron, and over Lake Erie (Fig. 2b).

Figure 3 shows the surface-layer aerosol number concentrations and the percent difference between the LAKE0 and BASE0 cases for 14 July 2004 at 12:00:00 GMT – the same time as is represented in Fig. 2. The results shown in Fig. 3 are based on simulations with nucleation, coagulation, and dry deposition processes all active in the model. The spatial distribution of aerosol number concentrations shown in Fig. 3a indicates that contributions to aerosol number concentrations within the analysis domain were dominated by urban and industrial emissions from Chicago, Detroit, and the Ohio River Valley region. Because the prevailing winds are northerly to northwesterly, most of the Great Lakes region was upwind of the major emissions sources during this period, with the exception of Lake Erie where surface level concentrations were  $\sim 8000 \text{ cm}^{-3}$ . Air over Lake Superior was the most pristine among all the Great Lakes, as expected since it is also the most remote. Concentrations over Lake Superior were

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generally less than  $\sim 2000 \text{ cm}^{-3}$  in the BASE0 case. Concentrations over Lakes Michigan and Huron were generally between  $\sim 2000$  and  $\sim 4000 \text{ cm}^{-3}$ .

Figure 3b shows relative increases in surface-level aerosol concentrations when wind-generated emissions from the lake surface are included in the model. The percentage change was greatest over southeastern Lake Superior, where the lake emissions increased the surface layer concentrations by  $\sim 20\%$ . This large percentage increase resulted from a combination of a relatively high lake surface emissions rate and a very low pre-existing aerosol concentration. Lake surface aerosol emission rates were actually higher over Lakes Erie and Huron than over Lake Superior (Fig. 2b), but because of the higher relative amounts of pre-existing aerosol in those regions, the percentage increases over the former lakes were less than  $\sim 8\%$  and  $\sim 4\%$ , respectively. The results in Fig. 3b also suggest that the impacts of the lake surface aerosol emissions are located predominantly within the source region. Percentage increases in aerosol concentrations over land downwind of the Great Lakes were generally smaller than  $2\%$  and persisted for only  $\sim 100 \text{ km}$ ; the exception to this result was over Michigan's Upper Peninsula where surface-layer aerosol enhancement was  $6\text{--}14\%$  for much of the area between Lakes Superior and Michigan.

The relationship between the lake surface aerosol flux and the surface-layer aerosol concentration can be better understood by looking at their temporal relationship. For simplicity we will focus this comparison on the region with the greatest relative increase in aerosol concentration – the southeast part of Lake Superior. Figure 4a shows the area of comparison – those grid cells over and adjacent to Lake Superior where the percentage difference between the LAKE0 and BASE0 cases at 12:00:00 GMT on 14 July 2004 was greater than  $10\%$ . Time series of the surface-layer aerosol number concentrations for the two cases, the percentage difference between them, and lake surface aerosol number fluxes are shown for this area in Fig. 4b. On this day, the wind speeds increased continuously from 0 to 12 GMT; as the winds picked up, the aerosol number concentrations decreased in the BASE0 case as dilution occurred. The same temporal trend was predicted in the case for which lake surface aerosol

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emissions were included in the model (the LAKE0 case), but in the latter case the rate of decrease in aerosol concentration was smaller. Both cases had an average surface layer concentration of  $\sim 4500 \text{ cm}^{-3}$  at 00:00 GMT, but by the 12:00 GMT minimum the LAKE0 average concentration was  $\sim 200\text{--}300$  particle per  $\text{cm}^3$  greater than the BASE0 case ( $\sim 2500 \text{ cm}^{-3}$  and  $\sim 2000 \text{ cm}^{-3}$  for the LAKE0 and BASE0 cases, respectively). In general, the percent contribution increase of lake surface-derived aerosols was more strongly correlated with the changing background particle concentration rather than increased lake surface emissions from the increased wind speed.

New particle formation is usually not a significant source of aerosol number over regions of high pre-existing aerosol loading; in those circumstances the competition for condensable material favors condensation onto pre-existing particles. However, in more pristine environments new particle formation events can be common when the concentrations of precursor gases are sufficiently high. In many regions atmospheric new particle formation can be the dominant mechanism for increasing the particle number concentration. This appears to be true for the northern Great Lakes region- a comparison of the BASE case with nucleation turned on (BASE0, Fig. 3a) and off (BASE1, Fig. 5a) indicates that surface-layer aerosol number concentrations were 3–4 times higher over most of Lake Superior in the model when nucleation was active. For Lakes Michigan, Huron, and Erie, aerosol number was reduced by  $\sim 50\%$  when nucleation was turned off in the model. Without nucleation, the percent increase in surface layer aerosol number concentration would have been up to  $\sim 200\%$  over Lake Superior and as high as  $40\%$  over Lakes Michigan and Huron (Fig. 5b). This implies that the importance of the lake surface emissions in the region is highly sensitive to the strength and frequency of new particle formation events in the region (and to the accuracy of the model in representing those events).

In addition to the surface-layer results, column burdens and vertical profiles of aerosol number were also examined to determine whether lake surface aerosol emissions could have significant impacts on aerosol number concentrations above the surface layer. Figure 6a shows the percent difference in aerosol column burdens between

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the LAKE0 and BASE0 cases; this comparison includes the six lowest layers in the model, from the surface up to  $\sim 200$  m. The maximum difference in column burden was  $\sim 10\%$ , and occurred slightly downwind of Lake Superior along the southeastern shore. Comparing Figs. 4a and 6a suggests that while the percentage increase in aerosol loading was greatest in the surface layer, the impact of lake surface aerosol emissions was carried further downwind above the surface layer. Figure 6b shows the vertical profiles of the aerosol number concentrations averaged over the grid cells highlighted in Fig. 6a (i.e., the grid cells in which the percent difference between the LAKE0 and BASE0 cases was greater than 5%). These vertical profiles indicate that the impact of lake aerosols decreased quickly with increasing altitude and was confined to only the  $\sim 200$  m closest the surface; this result is consistent with the limited observations of Slade et al. (2010). Figure 6b also illustrates the impact of nucleation (BASE1 and LAKE1) and coagulation plus dry deposition (BASE2 and LAKE2) on the aerosol vertical profiles in the model as well as the impact when the YSU PBL scheme is replaced with the MYJ PBL scheme (BASE3 and LAKE3). As was noted for the surface-level figures, nucleation was the dominant source of particles in this region for the model period. Without nucleation, the impact of lake surface aerosol emissions would be much greater on a percentage basis. On the other hand, without coagulation and dry deposition the pre-existing (BASE2) aerosol number concentrations would increase and thus the impact of lake-derived aerosol becomes smaller on a percentage basis. The results were similar when the MYJ PBL scheme was used rather than the YSU PBL scheme.

### 3.2 Averaged impacts for 14–27 July 2004

Averaged simulations results for the two-week period of 14 July– 27 July 2004 for the cases with all aerosol processes included in the model are shown in Figs. 7 and 8. Figure 7 shows the time-averaged lake surface aerosol emissions rate based on the modeled 22-m wind speed and the parameterization of Geever et al. (2005) (Eq. 1). The time-averaged emission rate for the two-week period in mid-summer was on the

order of  $10^6 \text{ m}^{-2} \text{ s}^{-1}$ , with the highest values over Lake Erie ( $\sim 1 \times 10^6 \text{ m}^{-2} \text{ s}^{-1}$ ) and the lowest over Lake Superior ( $\sim 6 \times 10^5 \text{ m}^{-2} \text{ s}^{-1}$ ). Figure 8a shows the two-week average of aerosol number concentration for the BASE0 case; and Fig. 8b shows the percent difference between the LAKE0 and BASE0 cases. As was seen for the 14 July 2004 episodic case, the greatest relative impacts from the lake surface aerosol emissions occurred over Lake Superior, even though emission rates were higher over the other Great Lakes. Aerosol number concentrations increased by  $\sim 8\%$  over central Lake Superior when lake aerosol emission is included, by  $\sim 5\%$  over Lake Huron and northern Lake Michigan, and by less than  $2\%$  over southern Lake Michigan and Lake Erie.

### 3.3 “Freshwater Oceans” – standard WRF-Chem emissions

As discussed in Sect. 2.4, WRF does not distinguish between ocean and freshwater lake water bodies, and so the publicly released versions of WRF-Chem (including the most recent release) treat all water surfaces as if they were oceans. Thus in standard WRF-Chem the Great Lakes are emissions sources for large sea-salt particles; specifically, the parameterization of Gong et al. (1997b) is used to model sea-salt emissions and is applied identically to both oceans and any other water bodies large enough to appear in the model grid. Several DEFAULT simulations (Table 1) were carried out that left this obvious error in place to investigate how this simplification impacted the aerosol population dynamics within the model.

For most of the simulation results, having sea-salt particles emitted over the Great Lakes introduced only small differences in aerosol number concentrations, but there were exceptions. Figure 9 illustrates how the erroneous introduction of sea-salt emissions from the Great Lakes can cause a significant modeling error. Figure 9a shows the percent difference in surface-layer aerosol number concentrations between the DEFAULT0 and BASE0 cases for 14 July 2004 at 20:00:00 GMT. Here, the inclusion sea-salt emissions over the Great Lakes has resulted in an overall decrease in total aerosol number concentrations. This seemingly counterintuitive result can be explained by the

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fact that the presence of large sea-salt particles in the aerosol population can greatly increase the aerosol condensational sink, leading to reduced nucleation rates and hence a net decrease in the total number of particles even though there is an increase in primary emissions. This link between the emission of sea-salt particles over the Great Lakes and the suppression of new particle formation is confirmed in Fig. 9b, which shows that when nucleation was turned off in the model, sea-salt emissions over the Great Lakes led to the more expected result of increased aerosol number concentrations (Fig. 9b). These results highlight the non-linearity of aerosol dynamics and the importance of including all sources of aerosol particles in the model correctly in order to model aerosol number (and hence CCN) concentrations properly. Averaged over the 14–27 July 2004 period, the impact of erroneously introducing sea-salt particles over the Great Lake regions was to reduce surface-level aerosol number concentrations by ~1 % over Lake Huron; impacts over other regions were minimal when time-averaged over this two-week period (Fig. 10).

## 4 Discussion

This is the first modeling study to investigate the impact of lake surface aerosol emissions on regional aerosol number concentrations over the Great Lakes region. The simulation results suggest that these wind-generated aerosol emissions can have significant impacts on number concentrations over the source region at least on an episodic basis, and potentially over longer timescales. However, there are significant uncertainties associated with emissions rate of lake aerosols, new particle formation rates, and several other model limitations. Even though the results here suggest that the impacts of the lake emissions on aerosol number concentrations exist largely within the source regions, the consequent effects of these sources on aerosol-cloud interactions could potentially impact meteorology downwind of the Great Lakes by changing the CCN distributions and cloud formation and propagation. To understand whether and how strongly these emissions would have an impact on regional meteorology,

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additional modeling work is needed using a fully coupled aerosol-cloud regional meteorology model. While the most recent publicly-released version of WRF-Chem (version 3.3) is able to model aerosol-cloud interactions, considerable additional improvement is still needed in the modeling of aerosol size distributions for us to have confidence in the results.

The first required improvement would be to extend the modeled aerosol size bin to below the current minimum diameter of 40 nm. Slade et al. (2010) found that the lake-derived aerosols exist primarily in the 15–40 nm diameter size range, and it was a significant assumption in this work to place the lake surface aerosol emissions in the 40–80 nm size bin. Newly formed particles from atmospheric nucleation are likewise placed in this 40–80 nm bin immediately upon formation. The inclusion of smaller particles in the modeled size distribution would mean faster coagulation rates with larger particles, resulting in fewer particles overall. Particles with  $D_p < 40$  nm also have faster dry deposition velocities than particles with  $D_p = 40$  nm over water surfaces (Seinfeld and Pandis, 2006). Thus, assuming the emitted particles to be larger leads to aerosol number concentrations being slightly overestimated. Furthermore, properly modeling the size distribution is critical for determining CCN concentrations- smaller particles are less likely to grow to large enough to be CCN. The specified size of the lake-derived particles will also affect the condensational sink, and thereby impact the dynamics of nucleation and condensational growth, which also has the potential to affect CCN concentrations.

A second needed improvement in the modeling of aerosol size distributions is in the parameterization of the new particle formation rate. The different parameterizations used in 3-D models can result in predicted new particle formation rates that span several orders of magnitude; this is a major source of uncertainty when estimating indirect radiative forcing and modeling aerosol-cloud interactions (Pierce and Adams, 2009; Zhang et al., 2010a, b). This study also highlights the need for accurate new particle formation parameterizations, since our simulation results suggest that new particle formation is the dominant source of particle number in the northern Great Lakes

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region and is the single most critical factor in determining the relative importance of the lake surface emissions source in determining the surface level aerosol concentration. As discussed earlier, the MOSAIC aerosol module in WRF-Chem applies the H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O binary homogeneous nucleation scheme of Wexler et al. (1994). Ambient measurements have suggested that binary nucleation theory underestimate observed nucleation rates (e.g., Weber et al., 1999), but these results are not conclusive for all regions. Another weakness of the model used is that it uses the H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O binary homogeneous nucleation to produce particles at  $D_p = 40$  nm, even though the particle formation parameterizations were derived for  $D_p \sim 3$ –10 nm. Again, a lower modeled minimum particle diameter is needed to properly account for the growth of particles from a few nanometers to CCN sizes.

The parameterization of the lake surface emissions flux is itself another significant source of uncertainty in this work. For the simulations presented here, it was assumed that the total number of particles emitted at the freshwater lake surfaces could be reasonably described by the parameterization of Geever et al. (2005), even though the latter was based on data collected over ocean rather than fresh water. To some extent uncertainty associated with the choice of emissions flux parameterization is unavoidable—many models for estimating wind-driven marine aerosol fluxes have been proposed, and they can result in very different flux estimations (Lewis and Schwartz, 2004). However, adapting a marine parameterization for fresh water requires some major assumptions about the nature of the aerosol production mechanism over marine and fresh water bodies. In its most basic conceptualization, marine particles are formed when droplets are ejected from the ocean surface and subsequently evaporate to leave a residual particle. If the ejection process is completely independent of surface chemistry, then applying a marine number flux parameterization to fresh water conditions is reasonable – the residual particles produced should be smaller in the latter case but they should be equally abundant. However, recent studies suggest strongly that ocean surface chemistry does play a significant role in regulating marine emissions (O’Dowd and De Leeuw, 2007). In this circumstance the marine flux parameterizations would

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not be transferable to fresh water conditions. At present no better option is available but this source of uncertainty should be studied more closely in the future.

Despite the limitations of the current model, our simulation results suggest that lake-generated ultrafine particles have the potential to impact aerosol number concentrations in the Great Lakes region. Further studies should be carried out to both better characterize the lake surface source via in-situ measurements and to model the impacts of these particles on the regional aerosol population throughout the year. This study was based on only a two-week period in July of 2004. Monitoring data indicates that July 2004 was a normal July, and that wind speeds over the Great Lakes during summer months are relatively low compared to other seasons (National Data Buoy Center, <http://www.ndbc.noaa.gov/>). During fall and late spring, when wind speeds are higher and the lakes are not covered by ice, the contribution of lake aerosols is likely to be greater. Moreover, the anticipated climatic changes for the Great Lakes region in the coming decades could further enhance the lake surface aerosol source. Summertime (July–September) wind speeds over Lake Superior have been increasing since 1985, likely due to climate warming (Desai et al., 2009). This increasing trend in wind speeds combined with reduced ice cover (also predicted under global warming scenarios) means that the emissions of lake aerosols will increase in intensity in the future and that its contributions will extend further into the early winter and late spring seasons when historically the northern Great Lakes have been frozen over.

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**Table 1.** Summary of WRF-Chem simulations performed.

Simulation	Emissions Over Lakes	Aerosol Processes	PBL Scheme	Period*
DEFAULT0 BASE0 LAKE0	Sea-salt None Ultrafine	All	YSU	14–27 July 2004 14–27 July 2004 14–27 July 2004
DEFAULT1 BASE1 LAKE1	Sea-salt None Ultrafine	Nucleation Off	YSU	14 July 2004 14 July 2004 14 July 2004
DEFAULT2 BASE2 LAKE2	Sea-salt None Ultrafine	Coagulation Off & Dry Deposition Off	YSU	14 July 2004 14 July 2004 14 July 2004
BASE3 LAKE3	None Ultrafine	All	MYJ	14 July 2004 14 July 2004

\* 12–13 July 2004 BASE-case simulations are used as chemical spin-up for each set.

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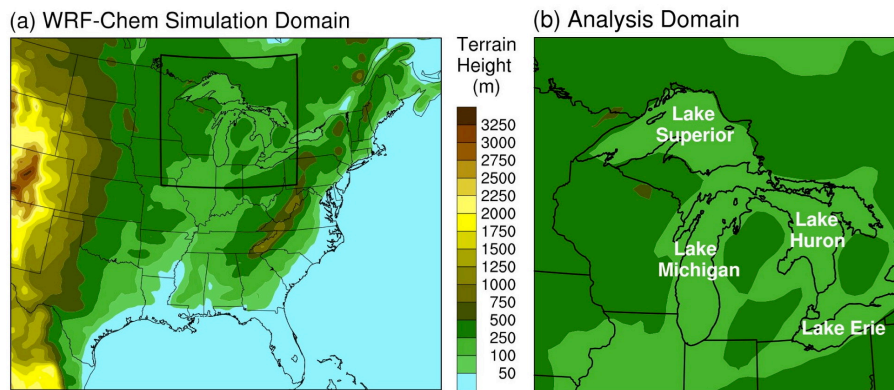
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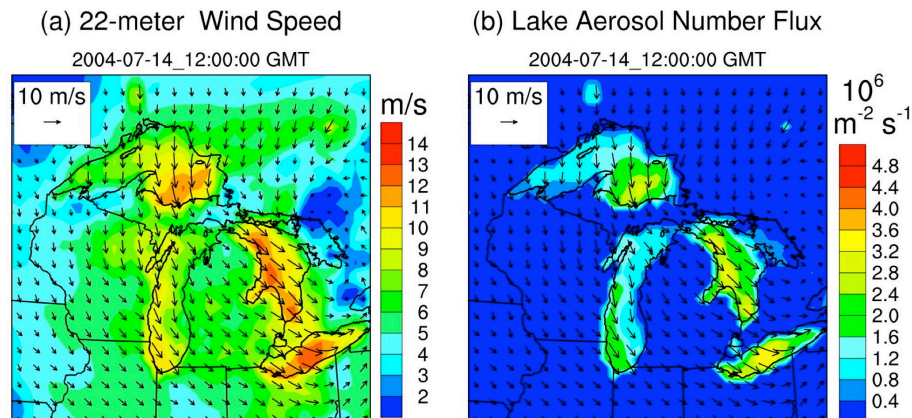


**Fig. 1.** (a) Terrain height (m) of the WRF-Chem simulation domain and (b) analysis domain focusing on the Great Lakes region.

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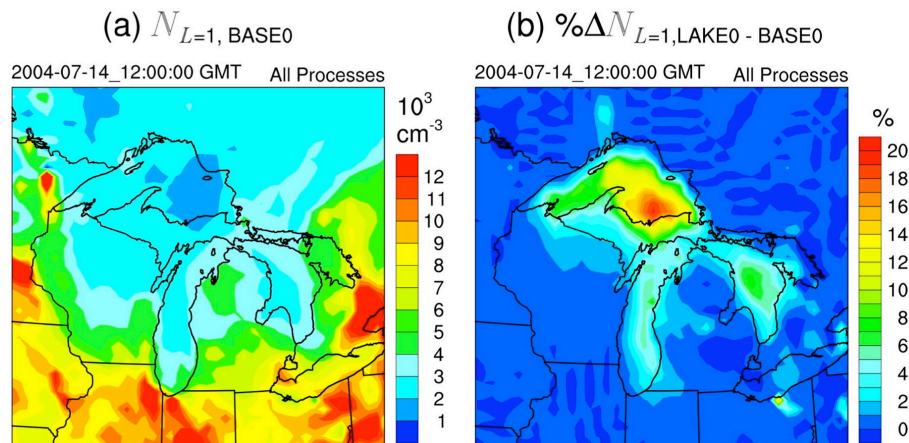


**Fig. 2.** Results for 12:00:00 GMT (08:00 a.m. Eastern Daylight Time) on 14 July 2004: **(a)** 22-m wind speed ( $\text{m s}^{-1}$ ), and **(b)** wind-generated aerosol number flux ( $10^6 \text{ m}^{-2} \text{ s}^{-1}$ ) over the lakes using the parameterization of Geever et al. (2005) (see Sect. 2.3), with wind vector overlay.

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**Fig. 3.** Simulation results for 12:00:00 GMT (08:00 a.m. Eastern Daylight Time) on 14 July 2004 with all aerosol dynamical processes included: **(a)** surface-layer aerosol number concentrations ( $10^3 \text{ cm}^{-3}$ ) for the BASE0 case and **(b)** percent difference between LAKE0 and BASE0 cases.

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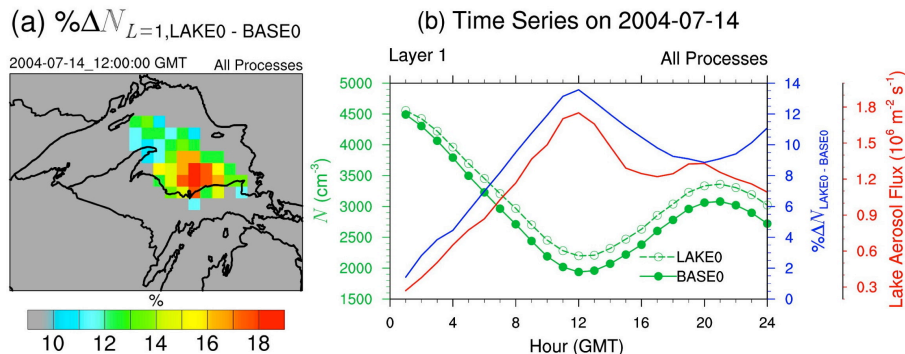
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**Fig. 4.** (a) Same as Fig. 3b but focusing on the Lake Superior region and highlighting grid cells in which the percent difference is greater than 10%; (b) hourly results for 4 July 2004 for the region highlighted in (a). Results shown include the total surface-layer aerosol number concentration  $N$  for BASE0 and LAKE0, the percent difference between them when all aerosol dynamical processes are included, and wind-generated lake surface aerosol number flux.

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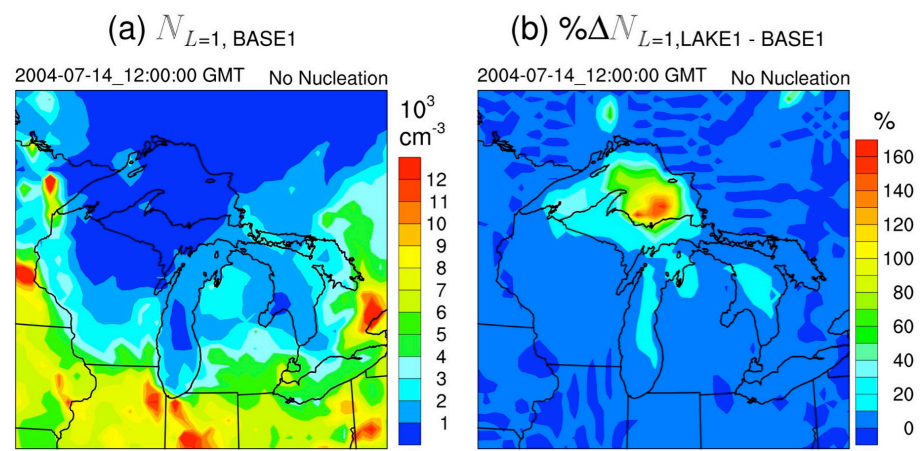
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**Fig. 5.** Same as Fig. 3 except results are based on simulations with nucleation turned off in the model.

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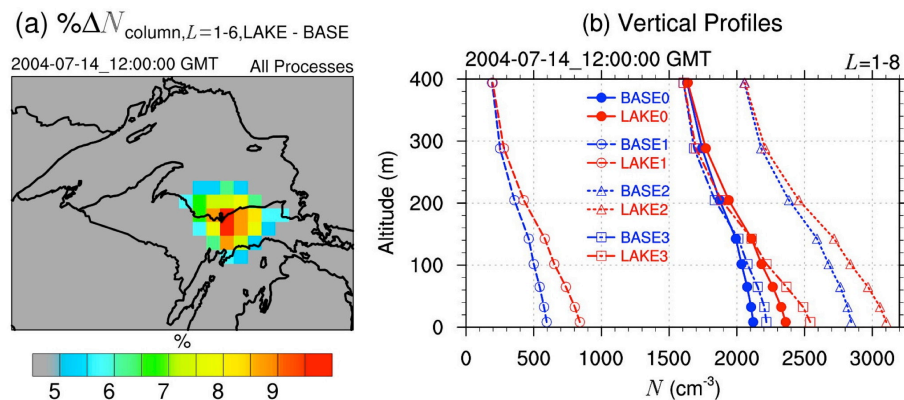
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**Fig. 6.** Results at 12:00:00 GMT on 14 July 2004: **(a)** percent difference in column aerosol number burden summed over the lowest six model layers between the LAKE0 and BASE0 cases when all aerosol dynamical processes are included in the model; **(b)** vertical profiles of aerosol number for the lowest eight model layers and for the grid cells highlighted in **(a)**.

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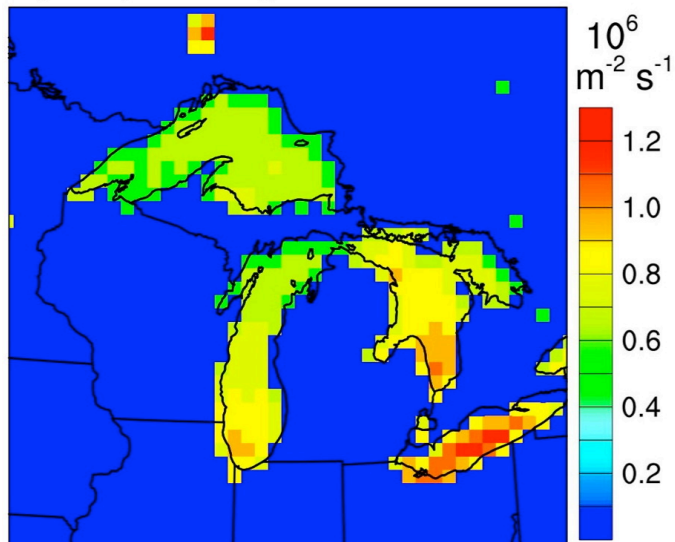


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**Lake Aerosol Number Flux**

July 14–27, 2004 Average

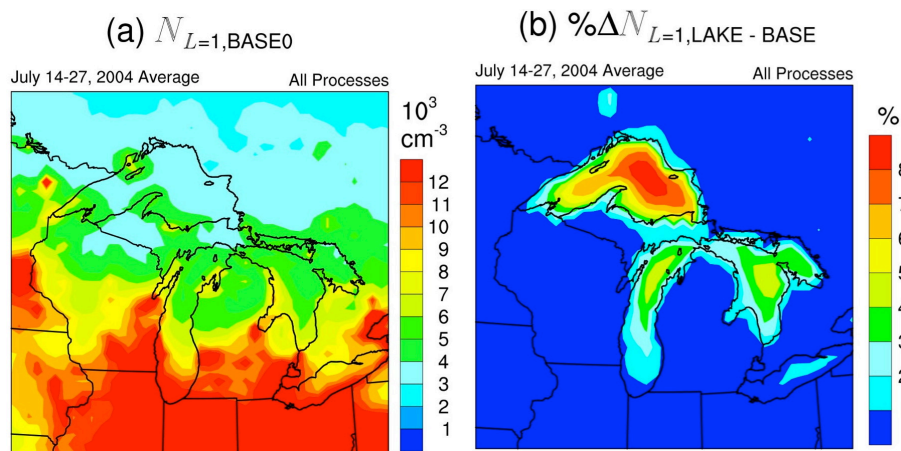


**Fig. 7.** Simulation results averaged over the 14–27 July 2004 two-week period: wind-generated aerosol number flux ( $10^6 \text{ m}^{-2} \text{ s}^{-1}$ ) over the Great Lakes region.

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**Fig. 8.** Simulation results averaged over the 14–27 July 2004 two-week period with all aerosol dynamical processes included: **(a)** surface-layer aerosol number concentration ( $10^3 \text{ cm}^{-3}$ ) for the BASE0 case, and **(b)** percent difference between LAKE0 and BASE0 cases.

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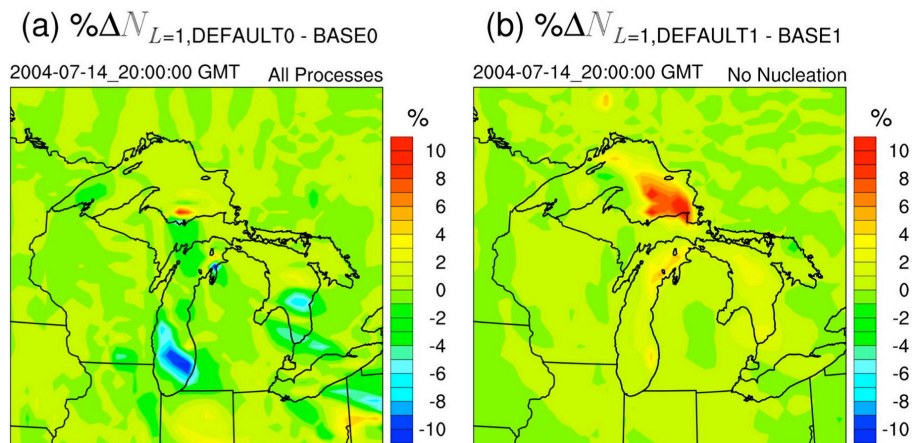
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**Fig. 9.** Percent difference in surface-layer aerosol number concentrations between the DEFAULT and BASE cases for 20:00:00 GMT on 14 July 2004: **(a)** with all aerosol dynamical processes included in the model, and **(b)** with nucleation turned off in the model.

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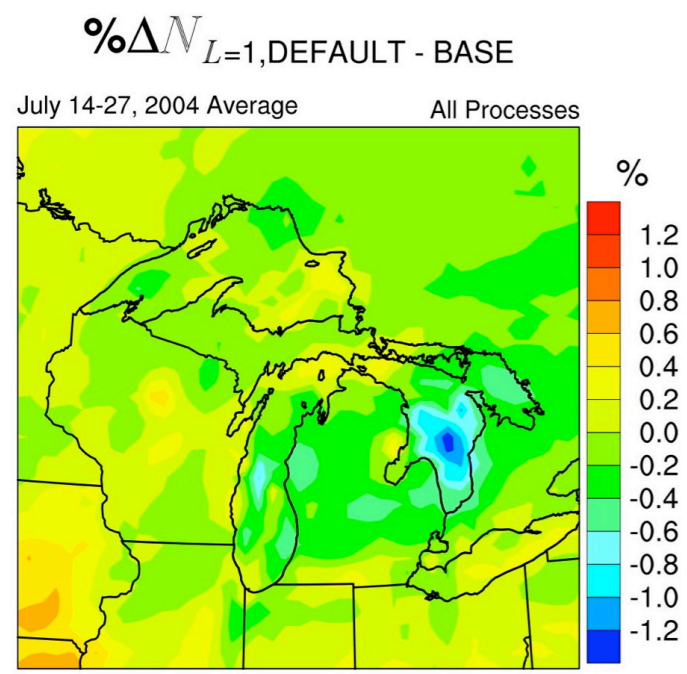
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**Fig. 10.** Percent difference in surface-layer aerosol number concentration between the DE-FAULT0 and the BASE0 cases; results are averaged over the 14–27 July 2004 period.

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