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Abstract

Recent laboratory chamber studies indicate a significant role for highly oxidized low volatility organics in new particle formation (NPF) but the actual role of these highly oxidized low volatility organics in atmospheric NPF remains uncertain. Here, particle size distributions (PSDs) measured in nine forest areas in North America are used to characterize the occurrence and intensity of NPF and to evaluate model simulations using an empirical formulation in which formation rate is a function of the concentrations of sulfuric acid and low volatility organics from alpha-pinene oxidation (Nucl-Org), and using an ion-mediated nucleation mechanism (excluding organics; Nucl-IMN). On average, NPF occurred on ~70% of days during March for the four forest sites with springtime PSD measurements, while NPF occurred on only ~10% of days in July for all nine forest sites. Both Nucl-Org and Nucl-IMN schemes capture the observed high frequency of NPF in spring, but the Nucl-Org scheme significantly over-predicts while the Nucl-IMN scheme slightly under-predicts NPF and particle number concentrations in summer. Statistical analyses of observed and simulated ultrafine particle number concentrations and frequency of NPF events indicate that the scheme without organics agrees better overall with observations. The two schemes predict quite different nucleation rates (including their spatial patterns), concentrations of cloud condensation nuclei, and aerosol first indirect radiative forcing in North America, highlighting the need to reduce NPF uncertainties in regional and global earth system models.

1 Introduction

Particle number concentration is an important factor in the health and climate impacts of atmospheric aerosols. High number concentrations of ultrafine particles may lead to adverse health effects (Knibbs et al., 2011). As cloud condensation nuclei (CCN), atmospheric particles modify cloud properties and precipitation and thus affect the hydrological cycle and climate indirectly. Aerosol indirect radiative forcing (IRF) remains

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a major uncertainty in assessing climate change (IPCC, 2013; Carslaw et al., 2013). Secondary particles formed via nucleation dominate the global total particle number abundance (Spracklen et al., 2008; Pierce and Adams, 2009; Yu and Luo, 2009), contributing as high as 80–95 % to total particle (> 3 nm) and 50–80 % to CCN number concentrations in most parts of the lower troposphere (Yu and Luo, 2009). Global simulations also indicate that the aerosol IRF is quite sensitive to nucleation parameterizations (Wang and Penner, 2009; Kazil et al., 2010; Yu et al., 2012). Furthermore, as a result of its dependence on meteorological conditions, emissions, and chemistry, new particle formation (NPF) is an important physical process involved in a number of climate feedback loops. Laboratory experiments indicate that ammonia, amines, ions, certain organic compounds and oxidants can all contribute to NPF. But there are numerous chemical reactions and physical processes involved, so there is no single unified theory that accurately describes NPF and the levels of enhancements due to different species vary widely among various studies (e.g., Zhang et al., 2004; Erupe et al., 2011; Chen et al., 2012; Dawson et al., 2012; Almeida et al., 2013; Berndt et al., 2014; Riccobono et al., 2014; Glasoe et al., 2015).

The indication that nucleation is significantly enhanced by organic species formed via oxidation of biogenic volatile organic compounds (VOCs; e.g., Riccobono et al., 2014) is of interest as it may have important implications for the interactions of anthropogenic and biogenic emissions and the associated climate forcing. Nevertheless, the various roles of organics and their oxidation products in NPF in the real atmosphere remains an active research area. Particle size distributions (PSDs) have been measured in a variety of environments around the globe and many of these measurements have been used to study NPF events (e.g., Kulmala et al., 2004). To evaluate the potential role of oxidation products of biogenic VOCs in NPF, PSDs taken over forested areas are particularly useful. In the present study, we analyze NPF events and non-events based on PSDs measured over nine forest areas in North America (NA) and compare them to model simulations with and without including organics in the nucleation rate calculation. Since biogenic VOC emissions and their oxidation are strongest in the summer, we use

the observed spring and summer contrast in NPF events to study the possible role of organics in NPF in NA and evaluate our current understanding of NPF processes in the atmosphere.

2 Methods

2.1 Measurements

PSDs from ~ 3 – 10 nm to $> \sim 100$ nm have been widely used to identify and to study particle nucleation and growth events in the atmosphere (e.g., Kulmala et al., 2004). In this study, we focus on PSDs observed in various years in eight US and one southern Canadian forest sites summarized in Table 1. The locations of these sites are marked on a Google map (<http://maps.google.com>) in Fig. 1. Egbert (EGB) is surrounded by a mixture of forests and farmland which is subject to a strong biogenic influence under certain conditions (Slowik et al., 2010), while all other eight sites are directly located in forest or mountain areas. PSDs have been measured at these nine sites with Scanning Mobility Particle Sizers (SMPSs) during different time periods. Sites 1–4 have at least one year of continuous PSD data while sites 5–9 have PSD data only during the summer (for different years). PSDs for one spring and one summer month for Sites 1–4 and for one summer month for Sites 5–9 are given in Fig. 1. It is clear that nucleation in the spring is much more frequent and stronger (i.e., higher concentrations of nucleation mode particles) when compared to summer months. Detailed analysis of these data and comparisons with simulations are given in the Results section.

2.2 Model and simulations

The modeling work in this study is based on the GEOS–Chem model (e.g., Bey et al., 2001) with an advanced particle microphysics (APM) model incorporated (Yu and Luo, 2009). GEOS–Chem is a state-of-the-art global 3-D model of atmospheric composition driven by assimilated meteorological observations from the Goddard Earth Observing

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System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). The model has been developed and used by many research groups and contains a number of state-of-the-art modules treating various chemical and aerosol processes (e.g., Bey et al., 2001; Chung and Seinfeld, 2002; Martin et al., 2003; Park et al., 2004; Evans and Jacob, 2005; Liao et al., 2007; Henze et al., 2008). The APM module in GEOS-Chem is optimized to simulate the formation of secondary particles and their growth to CCN sizes, using high size resolution (30 bins) for particles in the dry size range of 1.2–120 nm diameter. There are an additional 10 bins for 0.12–12 μm particles (Yu and Luo, 2009). More detailed information about GEOS-Chem and updates can be found at the model website (<http://geos-chem.org/>).

To study the possible role of organics in NPF, we employ the organics-mediated nucleation parameterization (Nucl-Org) derived from a CLOUD chamber study as given in Riccobono et al. (2014),

$$J_{1.7} = k_m \times [\text{H}_2\text{SO}_4]^2 \times [\text{BioOxOrg}] \quad (1)$$

where $J_{1.7}$ is the formation rate ($\# \text{cm}^{-3} \text{s}^{-1}$) of particles of $\sim 1.7 \text{ nm}$, k_m is the fitting pre-factor with a value of $3.27 \times 10^{-21} \text{ cm}^6 \text{ s}^{-1}$ (90% confidence interval: 1.73×10^{-21} to $6.15 \times 10^{-21} \text{ cm}^6 \text{ s}^{-1}$), $[\text{H}_2\text{SO}_4]$ and $[\text{BioOxOrg}]$ are the gas-phase concentrations ($\# \text{cm}^{-3}$) of H_2SO_4 and biogenic oxidized organic (BioOxOrg) vapors, respectively. In the chamber study reported in Riccobono et al. (2014), BioOxOrg molecules are organic compounds of relatively low volatility from the oxidation of pinanediol (a first-generation oxidation product of α -pinene) and represent later-generation oxidation products of biogenic monoterpenes.

In GEOS-Chem v8-03-02 on which this study is based, reactive VOCs are grouped into six categories (VOC_i , $i = 1-6$), with $\text{VOC}_1 = \alpha$ -pinene + β -pinene + sabinene + Δ -3 carene + terpenoid ketones; $\text{VOC}_2 =$ limonene; $\text{VOC}_3 = \alpha$ -terpinene + γ -terpinene + terpinolene; $\text{VOC}_4 =$ myrcene + terpenoid alcohols + ocimene; $\text{VOC}_5 =$ sesquiterpenes; and $\text{VOC}_6 =$ isoprene. Yu (2011) extended the two-product secondary organic

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aerosol (SOA) formation model (Chung and Seinfeld, 2002) to predict concentrations of extremely low volatile secondary organic gas ([LV-SOG]) from successive oxidation aging of the first generation oxidation products of various VOCs, i.e., semivolatile SOG (SV-SOG) and medium-volatile SOG (MV-SOG). LV-SOG has a saturation vapor concentration in the range of $\sim 0.0001\text{--}0.03\ \mu\text{g m}^{-3}$, corresponding to the extremely low volatile SOG reported in Ehn et al. (2014). The production rate of LV-SOG depends on the saturation vapor concentrations of MV-SOG and SV-SOG and temperature (Yu, 2011).

To use Eq. (1) to calculate organics-mediated nucleation, we separate LV-SOG from the oxidation products of VOC_1 (named LV-SOG $_{\alpha\text{-pinene}}$ thereafter) from those associated with VOC_{2-6} . LV-SOG $_{\alpha\text{-pinene}}$, a major component of total LV-SOG, corresponds well to the BioOxOrg reported in the chamber study of Riccobono et al. (2014). For comparison, simulations based on the ion-mediated nucleation of the $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ binary system (Nucl-IMN; Yu, 2010) are also presented. Nucleation rates based on the Nucl-Org scheme depend on $[\text{H}_2\text{SO}_4]$ and $[\text{LV-SOG}_{\alpha\text{-pinene}}]$ only while those based on the Nucl-IMN scheme depend non-linearly on $[\text{H}_2\text{SO}_4]$, temperature (T), relative humidity (RH), ionization rate, and surface area of pre-existing particles (Yu, 2010). Based on the CLOUD chamber study (Almeida et al., 2013; Riccobono et al., 2014), IMN is significant or dominant when $J_{1.7}$ is below $\sim 1\ \text{cm}^{-3}\ \text{s}^{-1}$ but neutral nucleation takes over when $J_{1.7} > \sim 1\ \text{cm}^{-3}\ \text{s}^{-1}$. Nucl-IMN calculates formation rates of particles at wet sizes of $\sim 1.5\ \text{nm}$. The nucleated particles are added into the first bin (wet size $\sim 1.5\ \text{nm}$) of the secondary particles, and the processes to grow them to large sizes considered in GEOS-Chem/APM include kinetic condensation of H_2SO_4 and total LV-SOG, equilibrium uptake of nitrate and ammonium, partitioning uptake of SV-SOG and MV-SOG, and self-coagulation (Yu, 2011).

The horizontal resolution of GEOS-Chem employed for this study is $2^\circ \times 2.5^\circ$ and there are 47 vertical layers (with 14 layers from surface to $\sim 2\ \text{km}$ above the surface). The emission inventories and schemes used can be found at the GEOS-Chem website and have also been summarized in Yu and Hallar (2014). Biogenic VOC emissions from

3 Results

VOC emissions in the summer are known to be much higher than VOC emissions in the spring. The high VOC emissions coupled with strong photochemistry and oxidation lead to significantly higher production rates and concentrations of oxidized SOGs of varied volatilities (medium volatile, semivolatile, and low-volatile) in the summer. Both [LV-SOG] and [LV-SOG _{α -pinene}] in the summer are much higher (by a factor of ~ 4 – 10) than those in the spring while [H₂SO₄] has a similar concentration in the spring as in the summer (not shown). Figure 2 shows horizontal distributions of monthly mean nucleation rates (J) in the boundary layer (0–1 km above the surface) in March and July of 2006 based on two different nucleation schemes (i.e., Nucl-Org and Nucl-IMN). Two nucleation schemes clearly predict significantly different spatial patterns and seasonal variations of nucleation rates. In March, $J_{\text{Nucl-Org}}$ is highest in the southeastern US as a result of high [LV-SOG _{α -pinene}] in the region, reaching 5 – $10 \text{ cm}^{-3} \text{ s}^{-1}$ (Fig. 2a), while J_{IMN} has two separate eastern and western nucleation zones with monthly mean J up to ~ 1 – $2 \text{ cm}^{-3} \text{ s}^{-1}$ (Fig. 2b). In July, boundary layer $J_{\text{Nucl-Org}}$ (Fig. 2c) is two orders of magnitude higher than J_{IMN} (Fig. 2d) although both indicate a high nucleation center in the northeastern US surrounding the Ohio valley with other scattered high nucleation zones in southern and western US along the coast. The low J_{IMN} is due to the high temperature in the summer. The shift of high $J_{\text{Nucl-Org}}$ nucleation zone from southeastern in March to northeastern in July is mainly associated with the change in the spatial distributions of [LV-SOG _{α -pinene}].

Comparisons of model predictions with in situ measurements are critical to evaluate our current understanding of atmospheric nucleation processes and to identify areas requiring further research. The PSDs measured at the nine forest sites, as marked in Figs. 1–2, provide a reasonably representative dataset for this purpose. As shown in Fig. 1, all the PSDs have lower cut-off sizes of 10 nm or smaller with some as small as 3–5 nm. Due to wall losses and lower charging and counting efficiencies of small particles, PSDs for mobility diameters smaller than ~ 10 nm may have large uncertainties.

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the duration from two hours to three hours) does not impact the main conclusions of this study.

It is clear from Fig. 3a–b that NPF events observed in Duke Forest are much more frequent and concentrations of nucleation mode particles are much higher in the spring than in the summer. More frequent and stronger observed NPF events in the spring have been reported in many previous field measurement studies (e.g., Dal Maso et al., 2005; Pryor et al., 2010; Kanawade et al., 2011; Pillai et al., 2013; Pierce et al., 2014). Both schemes predict frequent NPF events in March (Fig. 3c, e), which is consistent with observations, although the Nucl-Org scheme generally gives stronger NPF events and higher CN_{10-100} (Fig. 3g). Model simulations based on Nucl-Org scheme (Eq. 1) suggest that frequent (almost daily, Fig. 3d) and stronger NPF (Fig. 3h) should occur in the summer in Duke Forest, which was not observed with measurements (Fig. 3b). The relatively less frequent or lack of events in the summer indicates that, at least, $LV-SOG_{\alpha\text{-pinene}}$ cannot be the dominant nucleation factor as $[LV-SOG_{\alpha\text{-pinene}}]$ should be highest during the summer. However, the Nucl-IMN scheme is consistent with the observed lack of nucleation in the summer in Duke Forest. Note that there is no explicit temperature dependence in Eq. (1), an issue that is discussed later.

Figure 4 presents a statistical analysis and comparison of simulated NPF event days and CN_{10-100} based on two nucleation schemes with observations at four forest sites for the spring month and for all nine sites for the summer month. Table 2 gives the averaged values. For the statistical analysis, we look into the fraction (or frequency) of nucleation event days (F_{nucl} ; Fig. 4a – spring; Fig. 4b – summer), the proportion correct (PC; Fig. 4c–d), Pearson correlation coefficient (r) of the observed and simulated scatterplots of CN_{10-100} (Fig. 4c–d), and monthly mean CN_{10-100} (Fig. 4e–f). For the four sites with PSD data in the spring, NPF events occurred on 81, 65, 87, and 43 % of days for sites DUK, MMSF, SPL, and EGB, respectively (Fig. 4a), with an average value of 69 %. Both nucleation schemes capture most of the NPF events in the spring month (four-site average of 76 % for both Nucl-Org and Nucl-IMN). In July, there are no obvious NPF events for sites 1, 5–9 (DUK, WFM, PSP, UMBS, TNF, OZA) and infrequent

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events ($\sim 20\%$) for sites 2–4 (MMSF, SPL, EGB). Nucl-Org significantly overpredicts the nucleation frequency (by up to ~ 50 – 97%) for sites 1–2 (DUK, MMSF) and 4–6 (EGB, WFM, PSP) while Nucl-IMN slightly underpredicts F_{nucl} (by $\sim 10\%$) for sites 2–3 (MMSF, SPL). On average, Nucl-Org overpredicts F_{nucl} by 50% while F_{nucl} based on Nucl-IMN (7%) is close to the observed value (8%) for the nine sites in the summer month.

F_{nucl} alone is not adequate to assess the performance of the model. For example, for Site 2 (MMSF) in March, F_{nucl} based on Nucl-IMN is closer to the observed values than F_{nucl} based on Nucl-Org but r and PC are actually lower (Fig. 4c). Our analysis indicates that a large fraction of NPF event days predicted by Nucl-IMN for MMSF in March does not match the NPF events observed on specific days. In terms of r and PC, the values based on Nucl-Org and Nucl-IMN are close for the spring month (Fig. 4c and Table 2), but Nucl-IMN overall gives higher r and PC values for July (Fig. 4d and Table 2). The correlation was calculated for a whole month of hourly CN_{10-100} data (~ 700 data points). Due to clear diurnal variations associated with NPF in the spring (Fig. 3a) and the fact that both schemes capture $\sim 70\%$ of event and non-event days, the average r value reaches 0.37 for Nucl-Org and 0.49 for Nucl-IMN. As a result of the lack of obvious diurnal variations and inability of the global model to capture the sub-grid CN plumes (such as the short-duration CN spikes in Fig. 3h), the r value is much lower for July (nine-site average of 0.16 for Nucl-Org and 0.22 for Nucl-IMN) although PC reaches 0.88 for Nucl-IMN. The lower average PC value for Nucl-Org (0.43) is due to the over-prediction of NPF events (i.e., F_{nucl}).

Particle number concentrations and CN_{10-100} in NA (on a regional scale) are largely dominated by secondary particles formed via nucleation (Yu and Luo, 2009). Our analysis demonstrates that nucleation schemes have a strong impact on the simulated particle number concentrations. Compared to observations, CN_{10-100} averaged at the four sites is 40% higher based on Nucl-Org and 14% higher based on Nucl-IMN for the spring month (Table 2). For the summer month, CN_{10-100} averaged at the nine sites is 161% higher based on Nucl-Org and 27% lower based on Nucl-IMN (Table 2).

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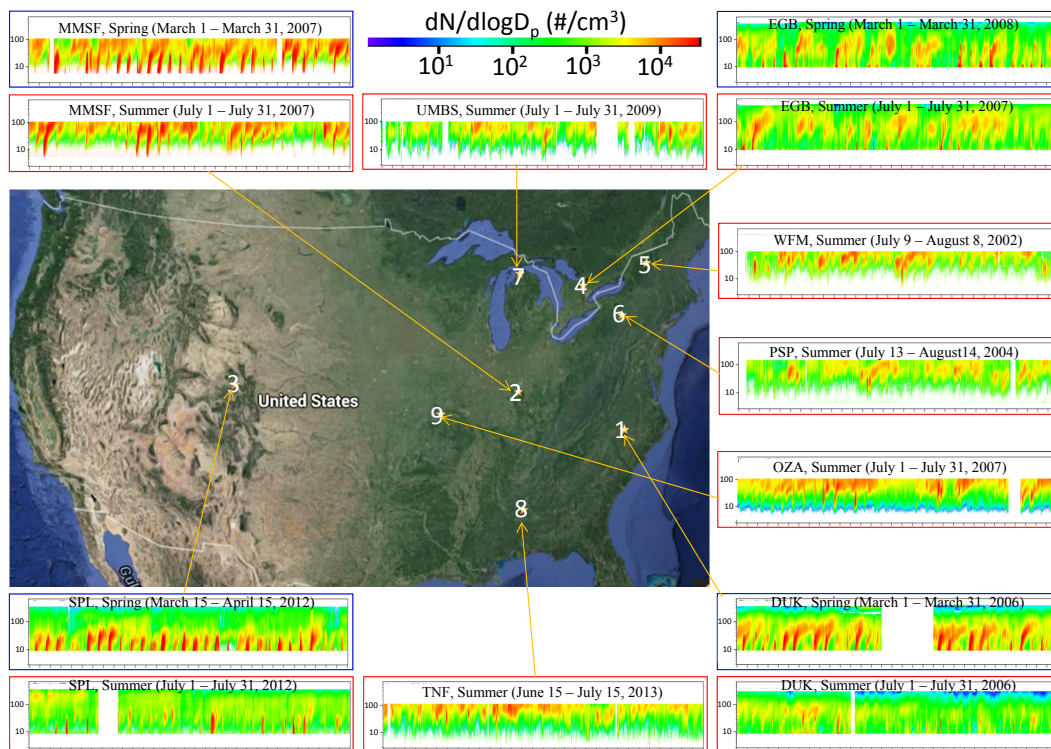


Figure 1. The locations of nine measurement sites in the forest areas in North America (see Table 1 for the names and references of the sites). Also shown are the particle size distributions measured in one spring month at sites 1–4 (with blue frames) and one summer month at sites (with red frames).

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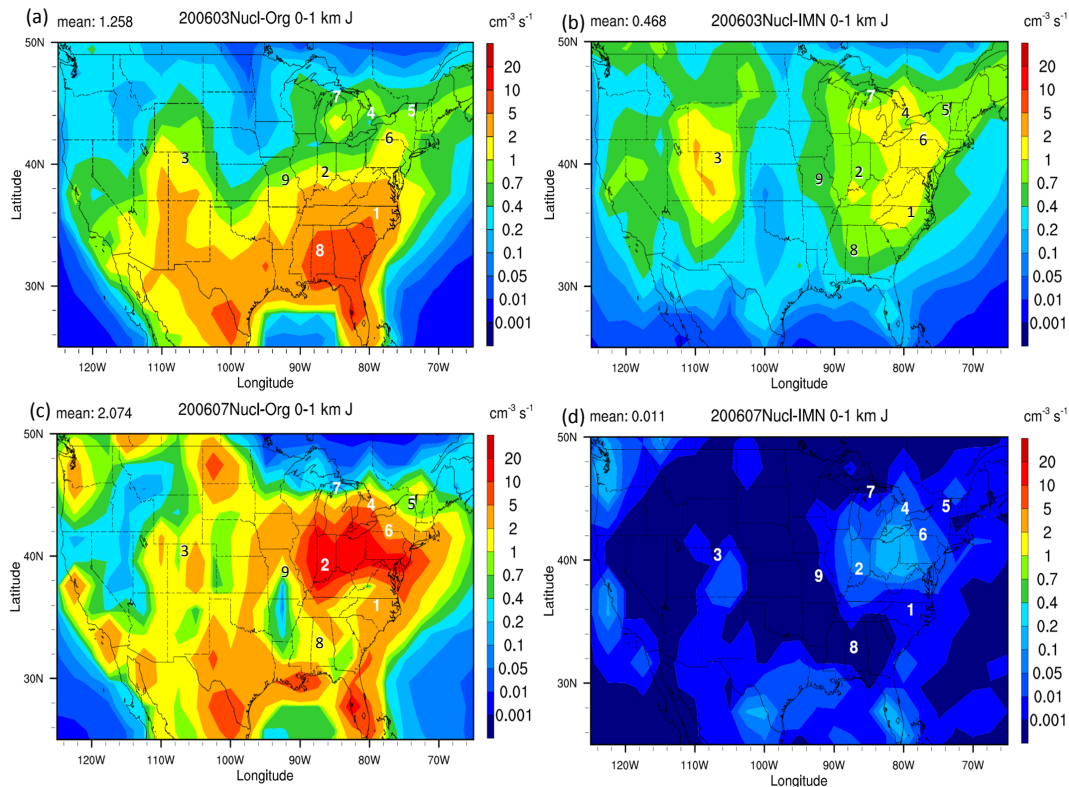


Figure 2. Horizontal distributions of monthly nucleation rates (J) in the boundary layer (0–1 km above surface) in March (a, b) and July (c, d) of 2006 based on two different nucleation schemes, i.e., Nucl-Org (a, c) and Nucl-IMN (b, d) (see text for details). The locations of sites 1–9 (Table 1) are marked.

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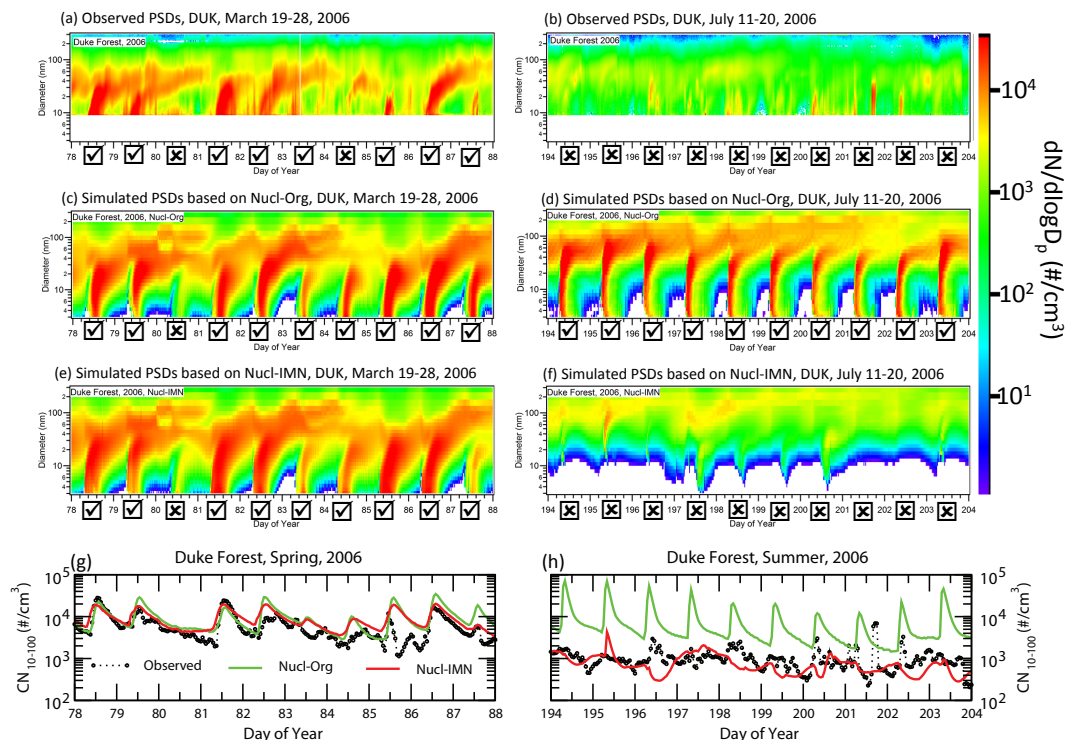


Figure 3. Particle size distributions (PSDs) observed (a, b) and simulated based on Nucl-Org (c, d) and Nucl-IMN (e, f) schemes during two ten-day periods in March (a, c, e) and July (b, d, f) of 2006 in Duke Forest (DUK), along with time series of the concentration of condensation nuclei between 10 and 100 nm (CN_{10-100}) (g, h). The event (☑) and non-event (☒) days determined based on the criterion given in Sect. 2.3 for the observed and simulated PSDs are marked in Fig. 3a–f.

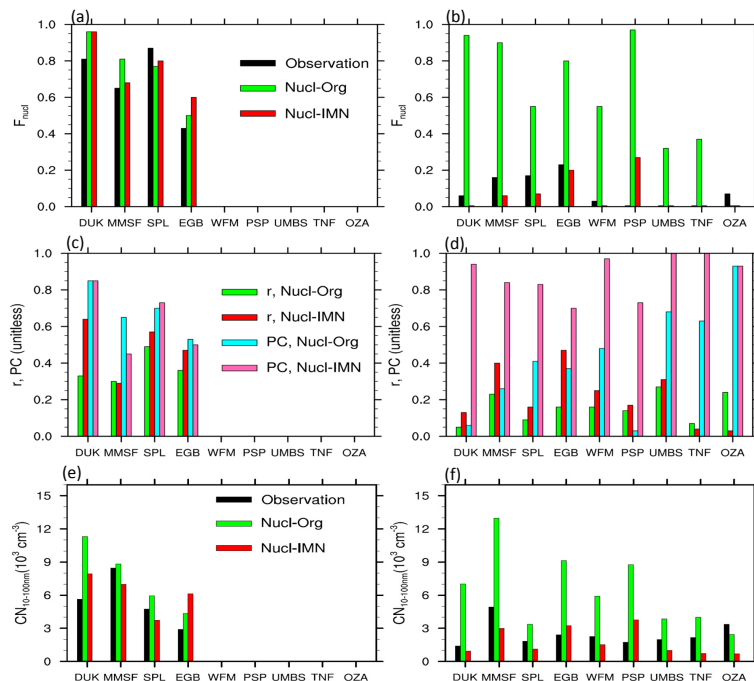


Figure 4. Statistical analysis of simulated NPF events and CN_{10-100} based on two nucleation schemes (Nucl-Org, Nucl-IMN) and comparison with observations at four forest sites for the spring month (**a**, **c**, **e**) and nine sites for the summer month (**b**, **d**, **f**). F_{nuc} is the fraction (or frequency) of nucleation event days (**a**, **b**). The proportion correct (PC) (**c**, **d**) is defined as (# of modeled events that were events + # of modeled non-events that were non-events)/(total # of modeled events + total # of modeled non-events). Monthly mean CN_{10-100} (**e**, **f**) and correlation coefficient (r) (**c**, **d**) of hourly mean CN_{10-100} during the month are also given.

