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Spatial and temporal scales of new particle formation events in eastern North America

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HIGHLIGHTS

- New particle formation (NPF) over eastern North America is regionally coherent.
- NPF frequently occurs on two sequential days.
- The mean spatial scale of NPF is 120–850 km based on the season and assumptions.
- Local scale NPF variability is linked to variations in boundary-layer dynamics.

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ABSTRACT

New particle formation (NPF) events have been observed in numerous locations. However, questions remain as to the scale of the events and their importance to regional and global particle number concentrations, size distributions and climate forcing. This study presents measured particle size distributions (PSD) at multiple sites across eastern North America and evaluates the degree of coherence on large (hundreds of kilometer) scales and the site-to-site variability across scales of tens of kilometers. Long-term data from sites separated by 1500 km demonstrate frequent and synchronous NPF, that over 80% of event days at both sites are followed by another event day and that event sequences are best described by a Markov Chain of order 1. Estimates of the mean spatial scale of NPF from a site in southern Indiana range from at least 120–850 km depending on the season and the precise assumptions applied. Despite the evidence for regional coherence in NPF, detailed measurements along an 80 km transect in southern Indiana also indicate some important sub-regional variability. While PSD from individual days typically indicate NPF at all three sites or at none of the sites, PSD measured in two urban environments show greater coherence than those from a centrally located site in a forest, and both the number of ultrafine particles and their growth rates are typically (but not uniformly) higher at the forested site. Some of the site-to-site variability appears to be causally linked to planetary boundary layer dynamics and variations in land cover.

1. Introduction and objectives

New particle formation (NPF) events have been observed across a wide array of ground-based stations (Kulmala et al., 2011), in the near-surface planetary boundary layer (PBL) (e.g. Spracklen et al., 2006) and in the free troposphere (e.g. Weber et al., 1999). Questions pertaining to the temporal and spatial scales of NPF events, subsequent growth and removal rates for the resulting ultra-fine particles (UFP) are critical to determining the global and regional importance of NPF in terms of dictating particle size distributions (PSD) and thus their relevance to direct and indirect climate forcing (Spracklen et al., 2006; Riipinen et al., 2011). Observations at regional and sub-regional scales have indicated NPF is observed over tens to hundreds of kilometers, but local variability is manifest at smaller scales. For example, while large spatial scale NPF events are frequently simultaneously observed at five locations over Scandinavia, event characteristics are rarely identical (Hussein et al., 2009). Further, the limited observational data currently available indicate notable variation in nucleation intensity, timing and growth rates. Measurements in northern Germany indicated simultaneous NPF at urban and upwind rural sites, but NPF was uniformly initiated earlier at the rural sites, and there were also cases of horizontally inhomogeneous NPF (Wehner et al., 2007).

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PSD in and near Pittsburgh, Pennsylvania, showed frequent simultaneous NPF on scales of tens of kilometers, but number concentrations at the rural site were 2–3 times lower than urban values (Stanier et al., 2004). Observations at five stations on a 500 km transect in southern Ontario also found clear cases of regional scale NPF events, but documented site-to-site variability that was attributed to local availability of sulfuric acid and variations in the background particle population (Jeong et al., 2010). Modeling studies have also indicated NPF is observed at regional scales, but the intensity of NPF and resulting ultra-fine particle concentrations indicate substantial spatial gradients over the eastern USA (Luo and Yu, 2011). There is also evidence that NPF is not uniform through the PBL, and may be concentrated in the residual layer (RL), with subsequent entrainment into the PBL (Wehner et al., 2010; Pryor et al., 2011; Olofson et al., 2009).

Thus, there is clear evidence for substantial geographic variability in the temporal and spatial scales of NPF, and for subsequent growth. Here we examine the coherence of nucleation events in eastern North America using PSD measurements from sites separated by a distance of hundreds of kilometers, and detailed transects across three different land-cover types coupled with numerical simulations using the Weather Research and Forecasting (WRF) model to examine variability of NPF on smaller spatial scales and links to boundary-layer dynamics. Thus, we present analyses designed to address the following research objectives:

- Analyze the degree to which PSD observed over distances of hundreds of kilometers indicate simultaneous NPF and therefore potentially regional events.
- Analyze seasonal and spatial differences in nucleation duration and spatial scales of NPF, and investigate possible causes of the observed variability.
- Investigate local variability in NPF and the characteristics there-of and specifically examine the degree to which site-to-site variability is attributable to variations in PBL dynamics.

2. Assessing the regional coherence and scales of NPF

2.1. Methods and approach

Long-term PSD measurements analyzed herein were collected at the Morgan Monroe State Forest (MMSF) in southern Indiana (39.317°N, 86.417°W) during 1/1/2007–07/31/2009, and Egbert in a semi-rural location around 70 km north of Toronto (44.23°N, 79.78°W) during 05/01/2007–05/31/2008. The measurements were conducted using Scanning Mobility Particle Spectrometers (SMPS) with inlets at a height of 46 m at MMSF (above a canopy of 26–28 m) and 5.5 m a.g.l. at Egbert (see Pryor et al. (2010) and Riipinen et al. (2011)). Given this study focuses on investigating the role of background conditions and boundary layer dynamics in initiating NPF and controlling the appearance of freshly nucleated particles in the near-surface layer, the small difference in the sampling heights at the two measurement sites is not expected to affect our conclusions. Data from both sites were used to classify NPF events following the approach of Boy and Kulmala (2002) where class A events exhibit a sudden appearance of a new particle mode with number geometric mean diameter <25 nm and consistent growth for at least 1 h. If this sudden increase in number concentration was not followed by a consistent growth profile a C event was identified. When a sudden increase of ultrafine particles was observed, but not from the smallest measured diameters, the day was classified as B event. A day not conforming to these criteria was defined as non-event.

2.2. Spatial and temporal coherence of NPF occurrence

Nucleation rates are dictated by the availability of nucleation chemical precursors, condensable species, background particle concentrations and meteorological conditions which determine the radiation flux, degree of stagnation and transport of nucleation precursors (Boy and Kulmala, 2002; Pryor et al., 2010). For these reasons the sequence of event and non-event days at a location may not be independent of air mass history (Sogacheva et al., 2007). We analyzed the importance of atmospheric preconditioning in dictating NPF using a Markov-chain approach:

$$X_t|X_{t-1},\ldots X_{t-k} \sim \text{Be}\left(p_{X_{t-1}},\ldots p_{X_{t-k}}\right) \quad (1)$$

where $p_{X_{t-1}}$ = probability of a nucleation event X occurring at time $(t-1)$ and Be = Bernoulli distribution. The zero order log-likelihood L_0 of the model is defined as:

$$L_0 = n_0 \times \ln p_0 + n_1 \times \ln p_1 \quad (2)$$

where n_0 and n_1 = number of non-event and event days and $p_0$and p_1 = respective probabilities. A similar approach can be used to derive the log-likelihood of higher orders:

$$L_1 = n_{01} \times \ln p_{01} + n_{11} \times \ln p_{11} \quad (3)$$

To determine the most parsimonious Markov Chain order we use the Bayesian information criterion (BIC) (Schoof and Pryor, 2008) computed as:

$$\text{BIC}(m) = -2L_m + 2^m[\ln(n)] \quad (4)$$

where L_m = log-likelihood for a model of order m and n = sample size.

Data from MMSF indicate 87% of event days are followed by another NPF event day, while for Egbert p_{11} is 83%. Further, at both sites a first order Markov chain model is the proper model to describe the occurrence of NPF (i.e. BIC is minimized for L_1) (Table 1). This implies that the conditions associated with NPF persist for multiple days. Further, consistent with prior research which has indicated weak association between the condensational sink (CS) and occurrence of NPF (Pryor et al., 2010), the occurrence of consecutive event days is not prevented.

A test of independence applied to event occurrence (i.e. a binary index of the occurrence (1) or not (0)) of NPF indicates that events are not randomly distributed at the two sites, but rather tend to occur simultaneously (Table 2). This suggests regionally coherent NPF, or at least regional coherence in the conditions associated with NPF, and thus regional-scale forcing. A statistically significant dependence is also evident for lag-1 (i.e. when MMSF events are shifted one day back relative to Egbert), but not for the converse (i.e. when MMSF events are shifted one day forward from Egbert) (Table 2). This result may indicate transport of conditions

Table 1
The probability of NPF on any given day (p_1) and two sequential days (p_{11}) during 05/01/2007–05/31/2008 at the MMSF and Egbert sites. The Bayesian information criterion (BIC) for a Markov Chain of up to the third order is also reported.

	MMSF	Egbert
Classifiable days	292	331
p_1	0.47	0.41
p_{11}	0.41	0.34
BIC(0)	453.02	405.06
BIC(1)	232.98	171.64
BIC(2)	414.71	311.30
BIC(3)	417.27	294.73