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Nucleation and growth of sub-3nm particles in the polluted urban atmosphere of a megacity in China

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Outline

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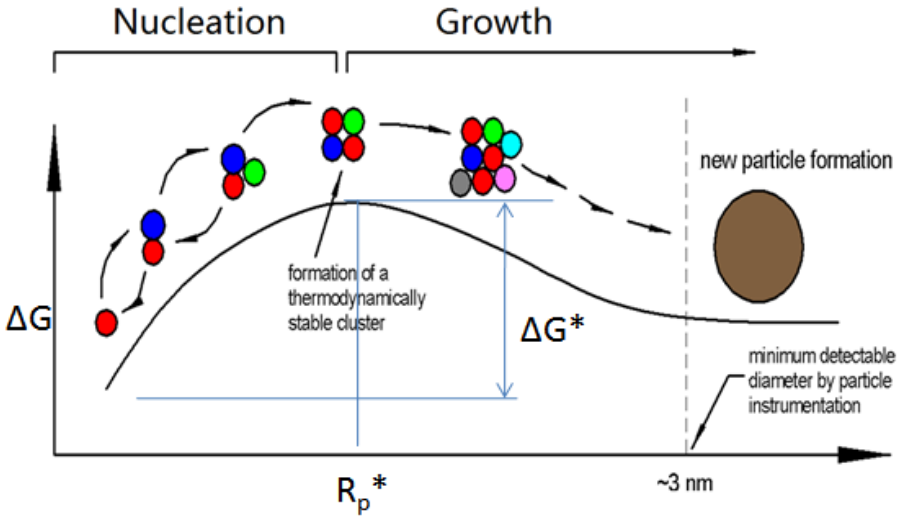
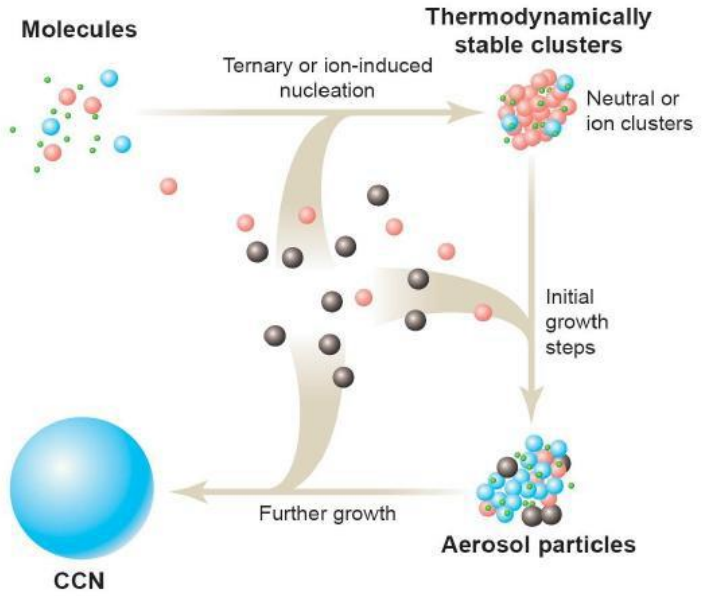
Introduction

- New particle formation (NPF) is an important source of secondary aerosols in the atmosphere.NPF is a two-stage process consisting of the formation of clusters and subsequent growth to detectable sizes.
- Direct measurements of size- and time-dependent nucleation rate and growth rate in sub-3 nm size range are important to constrain the relative contributions from different mechanisms and precursors. Such measurements are also important to evaluate the survival probability of new particle to CCN-active sizes and to reveal the limiting factors in the process.
- Here we reported the nucleation and growth of sub-3 nm particles in the urban atmosphere of Nanjing, China on arbitrarily selected observation days in the spring, summer, and winter of 2014–2015. Our aim was to (1) provide new information about the initial steps of NPF based on size- and time-resolved nucleation rate and growth rate measurements, and (2) find possible limiting factors behind the seasonal and diurnal variations of nucleation events in the polluted urban atmosphere.

Two critical steps in New Particle Formation





Methodology

- Field measurements

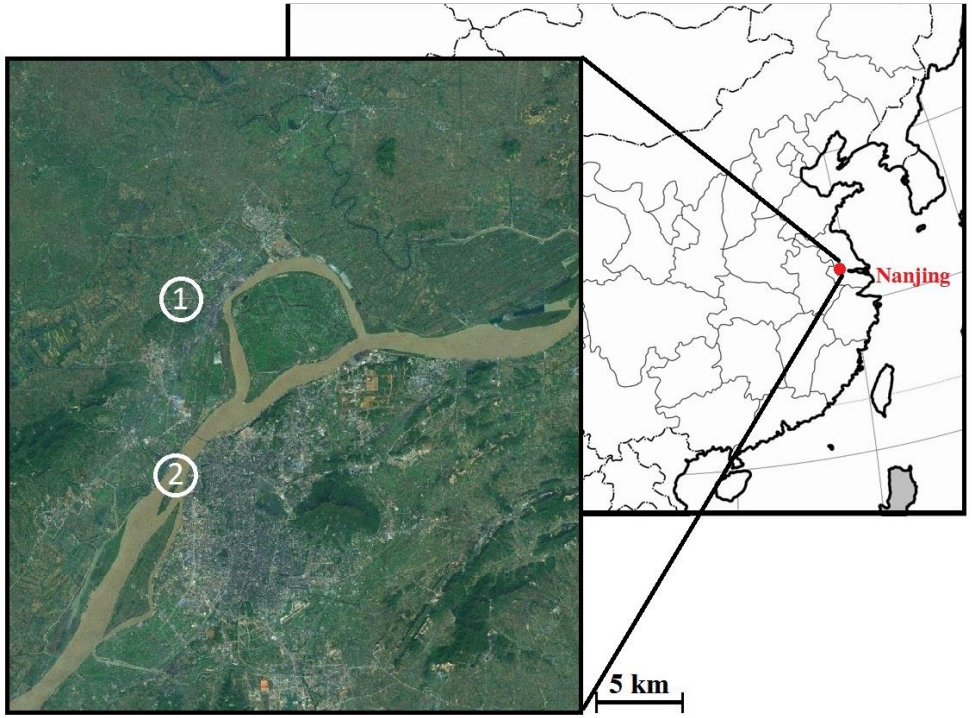
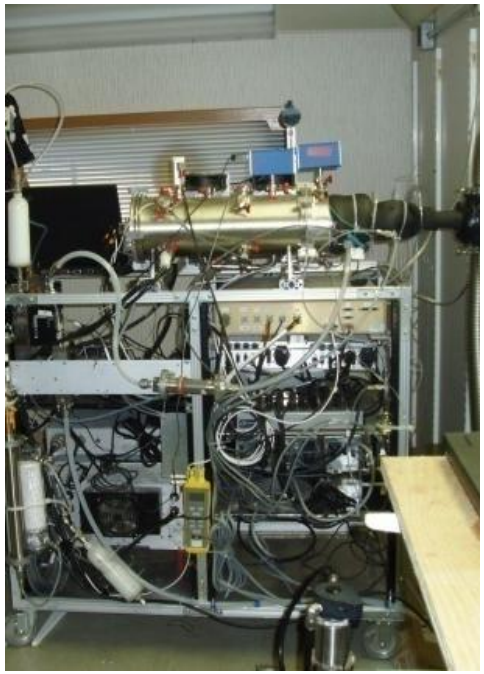


Figure 1. Locations of two urban measurement sites in Nanjing, the second largest megacity in the Yangtze River Delta region, China, (1) are the NUIST site and (2) is the summer measurement site.



PSM	>1 nm particles
Nano-SMPS	3-64 nm particles
Long-SMPS	10 - 478 nm particles
NO_3-CIMS	H_2SO_4
PTR-TOF-MS	VOCs
Gas analyzer	SO_2, O_3, NO_x/NO_y, CO

➢ Nucleation event and growth patterns

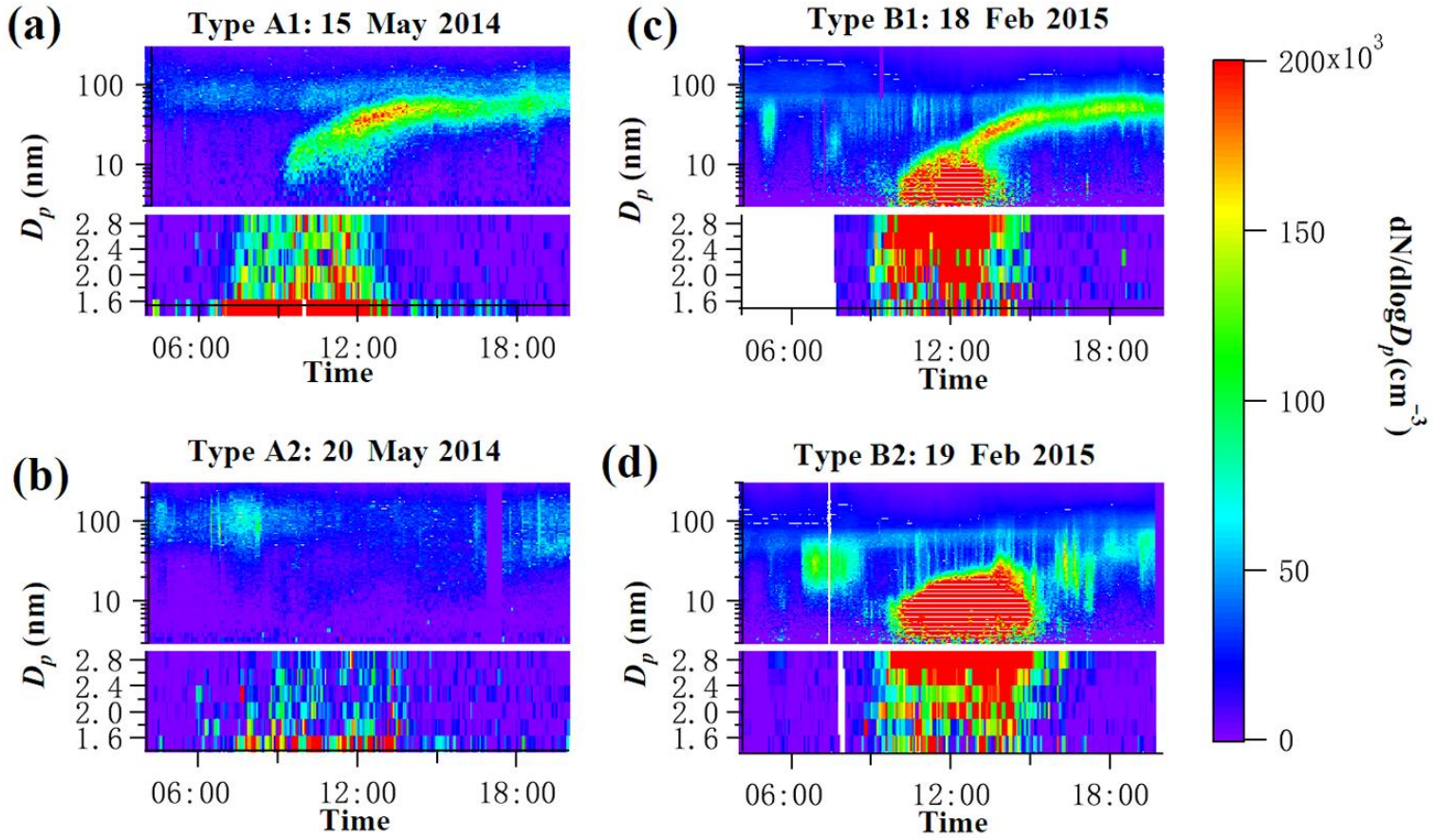


Figure 2. Size spectra of typical (a) Type A1 event on 15 May 2014, (b) Type A2 event on 20 May 2014, (c) Type B1 event on 18 February 2015, and (d) Type B2 event on 19 February 2015, during our measurement period.

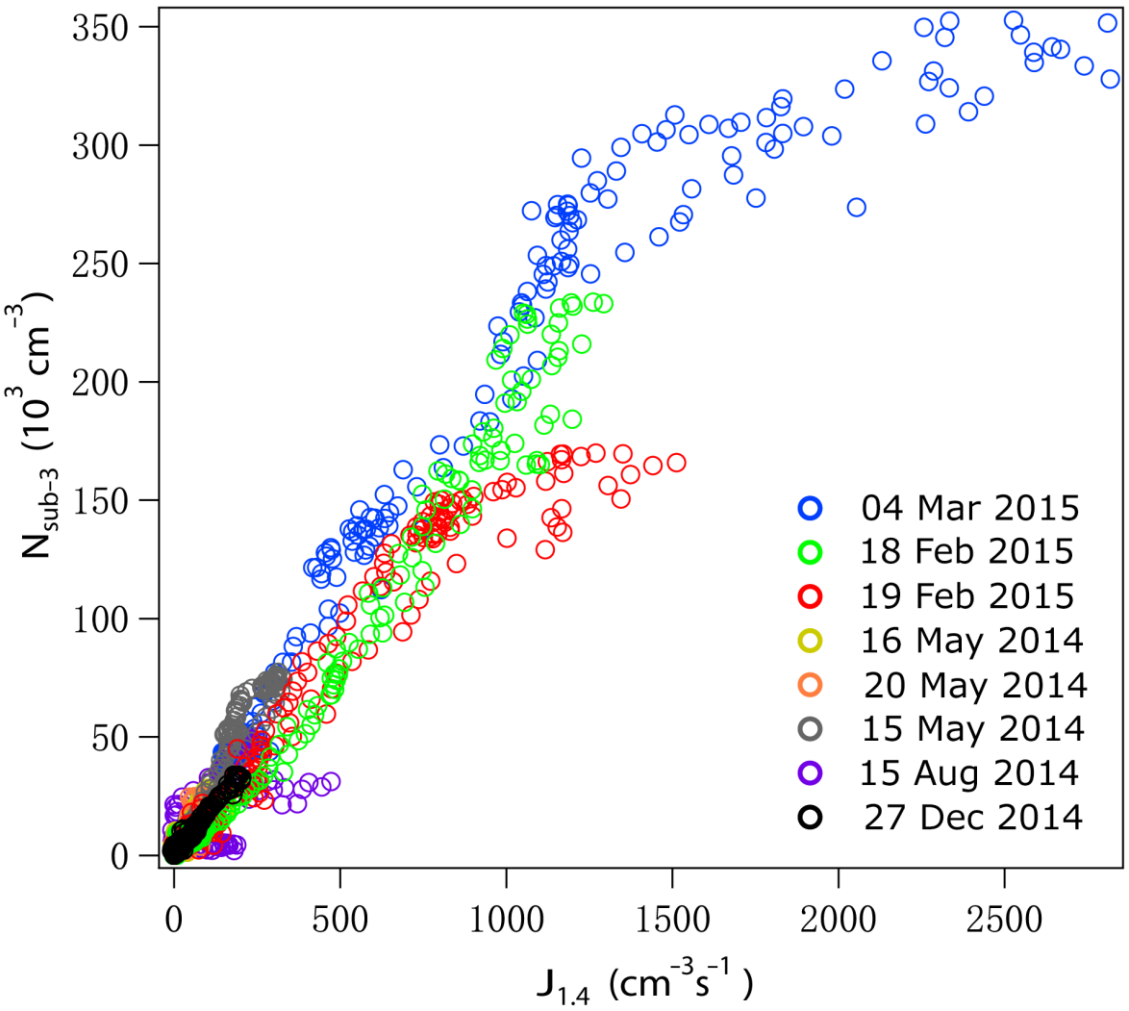


Figure 3. N_{sub-3} vs. $J_{1.4}$ in the eight nucleation events in February, May, December, and August during 2014–2015. The events were indicated by different colors .

- Formation rate and growth rate calculations with a simplified GDE method

$$\frac{\mathrm{d}N\left(D_{\mathrm{p1}},D_{\mathrm{p2}}\right)}{\mathrm{d}t}=J\left(D_{\mathrm{p1}},t\right)-J\left(D_{\mathrm{p2}},t\right)-\mathrm{CoagSnk}\left(D_{\mathrm{p1}},D_{\mathrm{p2}},t\right)+\mathrm{CoagSrc}\left(D_{\mathrm{p1}},D_{\mathrm{p2}},t\right) \quad (1)$$

$$\mathrm{CoagSnk}\left(D_{\mathrm{p1}},D_{\mathrm{p2}},t\right)=N\left(k,t\right)\sum_{i=1}^{98}\left(1-\theta_{\mathrm{k,i,k}}\right)K_{\mathrm{k,i}}N\left(i,t\right) \quad (2)$$

$$\mathrm{CoagSrc}\left(D_{\mathrm{p1}},D_{\mathrm{p2}},t\right)=\frac{1}{2}\sum_{i=1}^{k-1}\sum_{j=1}^{k-1}\theta_{\mathrm{i,j,k}}K_{\mathrm{i,j}}N\left(j,t\right)N\left(i,t\right) \quad (3)$$

$$J\left(30nm,t\right)=0\longrightarrow\ \boldsymbol{J\left(1.4nm,t\right)}$$

$$GR\left(D_{\mathrm{p}},t\right)=J\left(D_{\mathrm{p}},t\right)\Big/n\left(D_{\mathrm{p}},t\right)$$

$$n\left(D_{\mathrm{p}},t\right)=\frac{dN\left(t\right)}{dD_{\mathrm{p}}}$$

Results and Discussion

- Seasonal and diurnal variations of nucleation event
- Size- and time-dependent formation(growth) rates of sub-3nm particles
- Growth rate due to condensing organic vapor on newly formed nuclei in sub-3nm sizes

Seasonal and diurnal variations of nucleation event

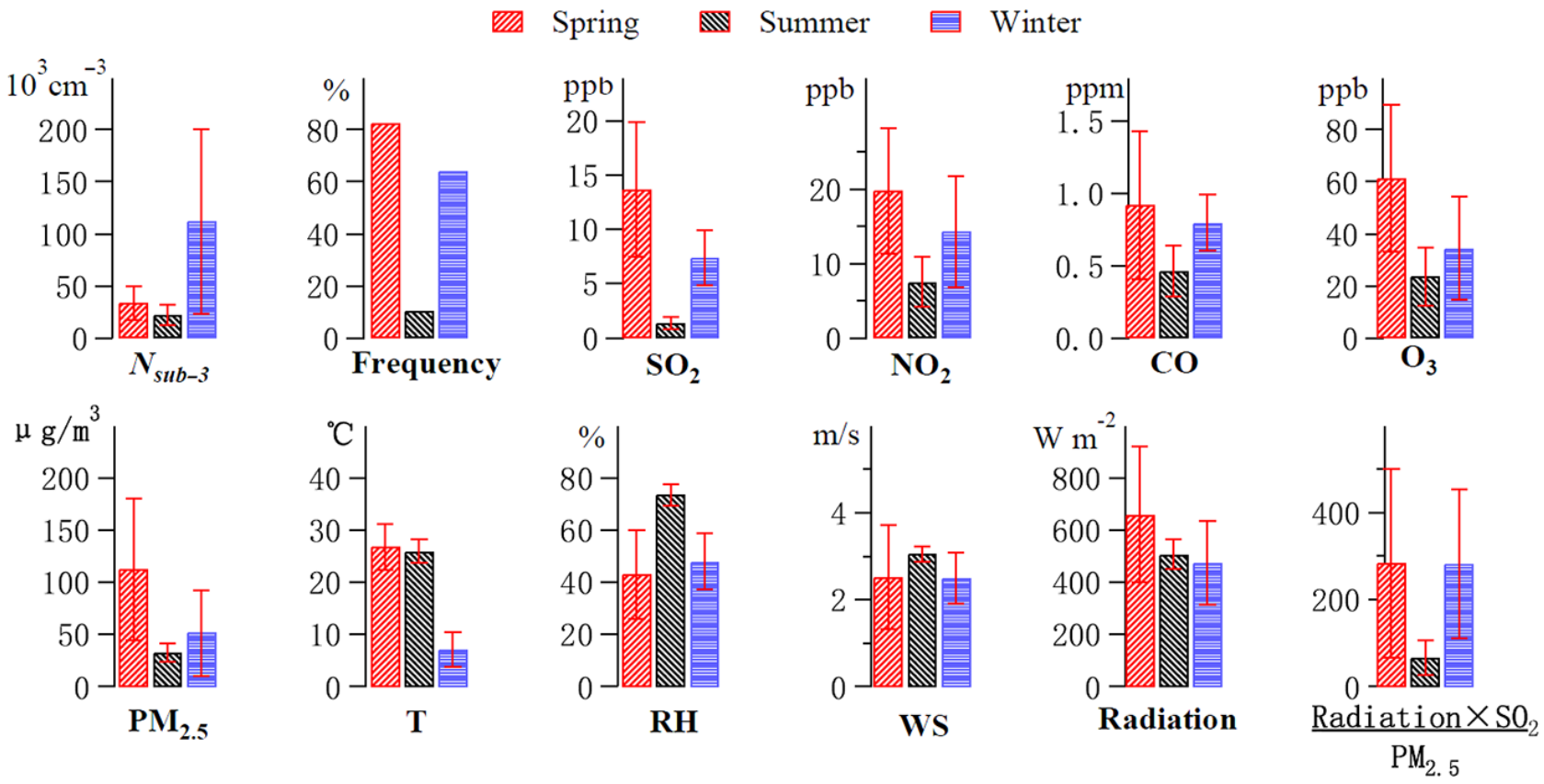


Figure 4. Mean and standard deviation of event-averaged N_{sub-3}, anthropogenic trace gases, $PM_{2.5}$, and meteorological variables for nucleation events in spring, summer and winter.

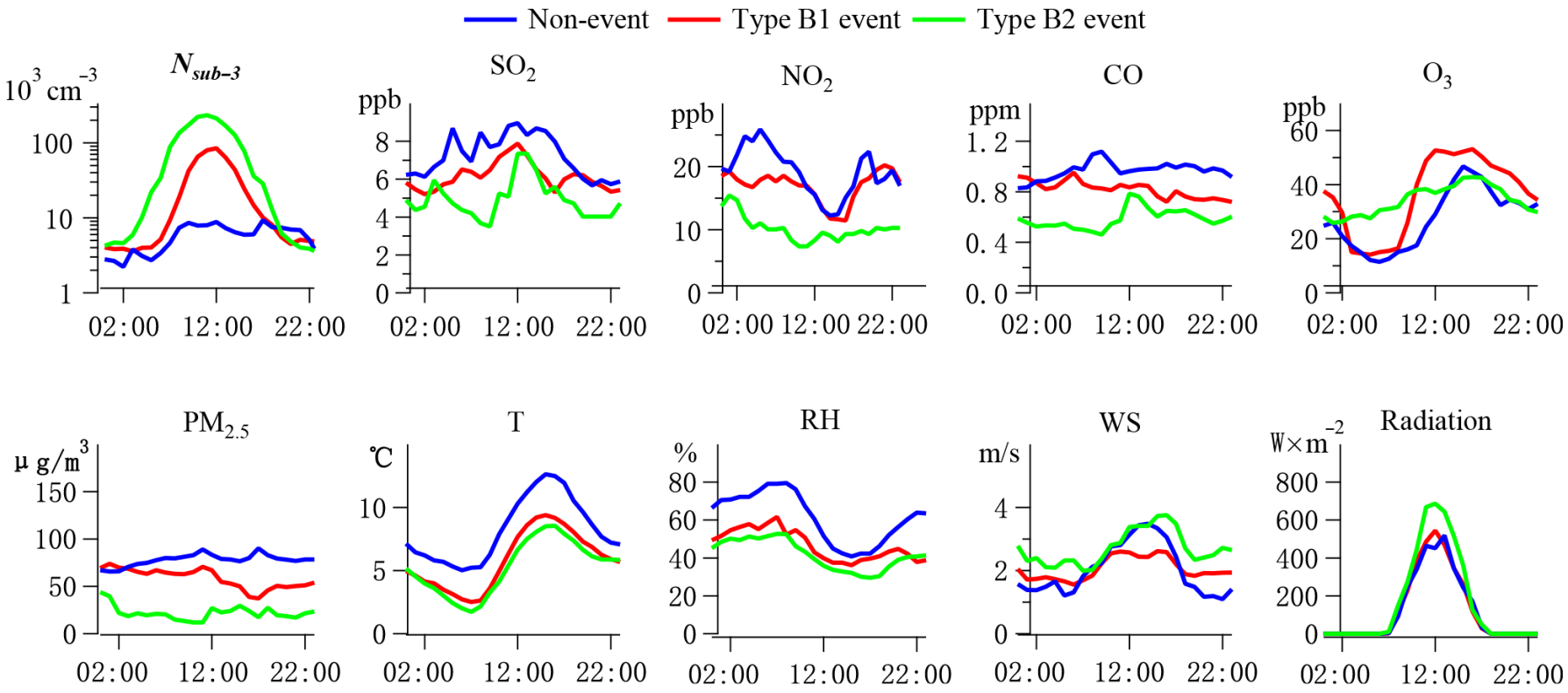


Figure 5. Diurnal variations of mean N_{sub-3}, anthropogenic trace gases, $PM_{2.5}$, and meteorological variables on non-event days and event daysduring winter measurement period.

limiting factors for nucleation

a. radiation, temperature, RH, and CS in winter and spring

b. temperature, RH, and available gaseous precursors in summer

Size- and time-dependent formation(growth) rates of sub-3nm particles

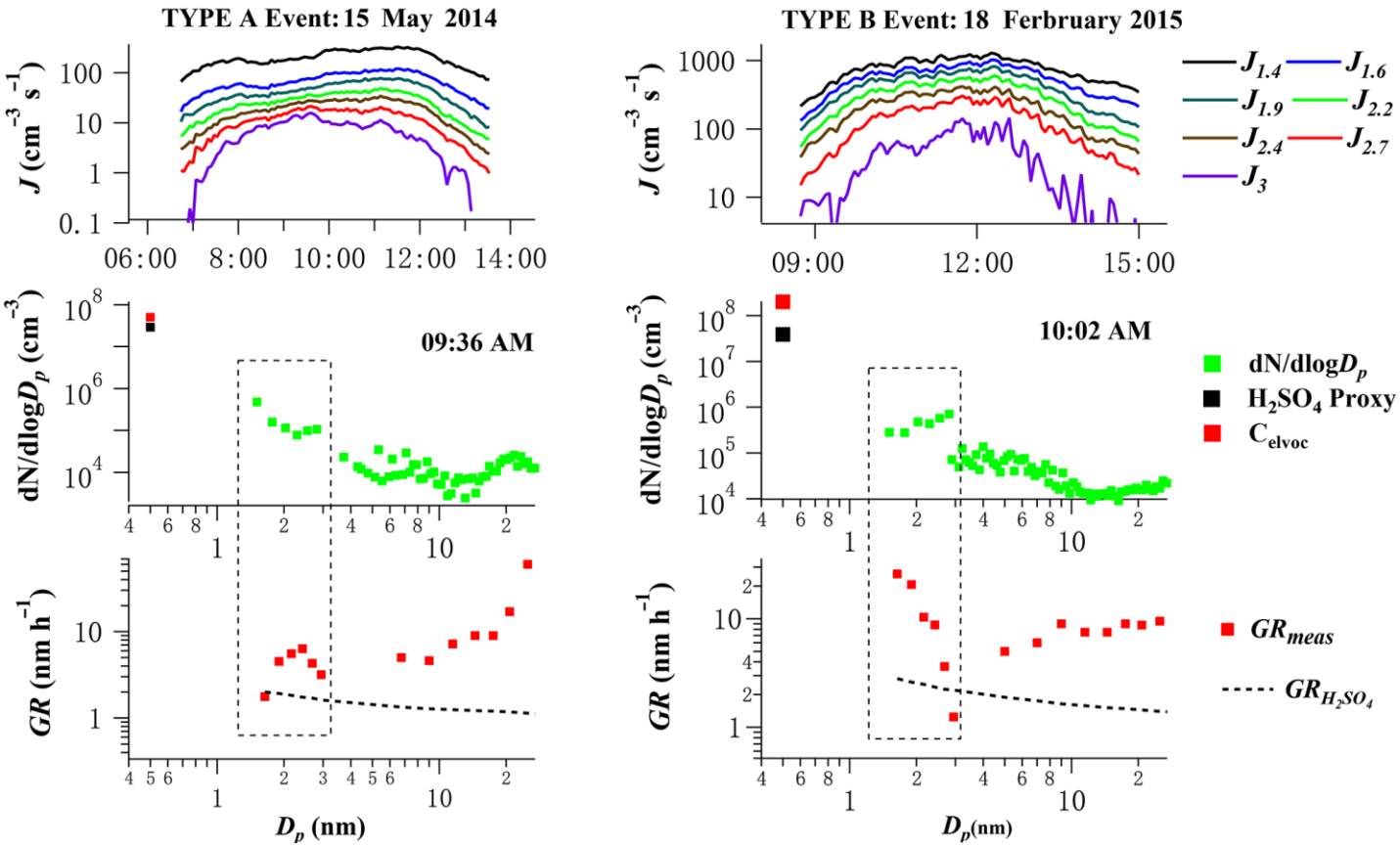
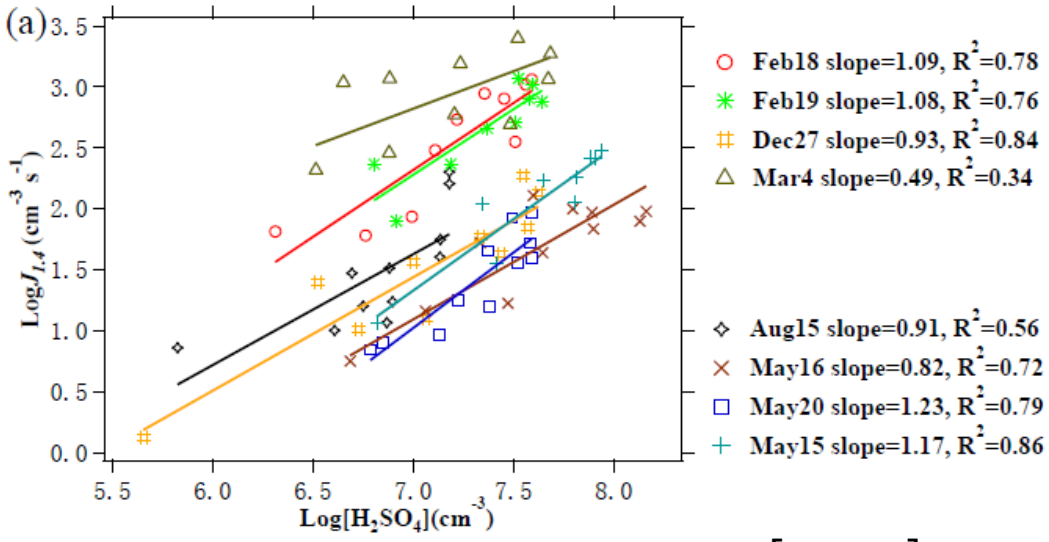


Figure 6. Upper: formation rates of 1.4, 1.6, 1.9, 2.2, 2.4, 2.7, and 3.0 nm cluster/particles on 15 May 2014. Middle: particle size distribution selected during the two events. Lower: particle growth rates measured during the same time periods.



$$[H_2SO_4] = 8.21\times10^{-3} k \cdot \text{Radiation} \cdot [SO_2]^{0.62} \cdot (CS \cdot RH)^{-0.13}$$

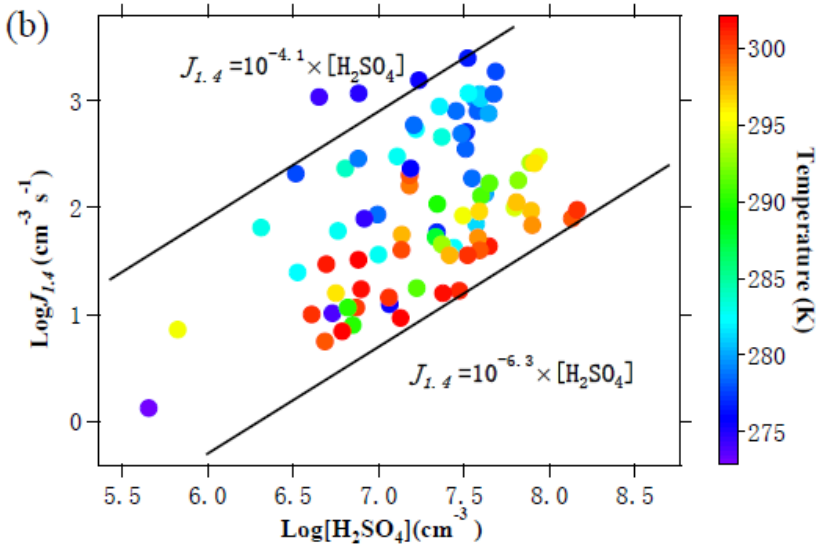


Figure 7. (a) Correlations between log $J_{1.4}$ and log $[H_2SO_4]$ for the eight events.(b) The same data set as panel (a), but with symbol color to indicate ambient temperature.

Growth rate due to condensing organic vapor on newly formed nuclei in sub-3nm sizes

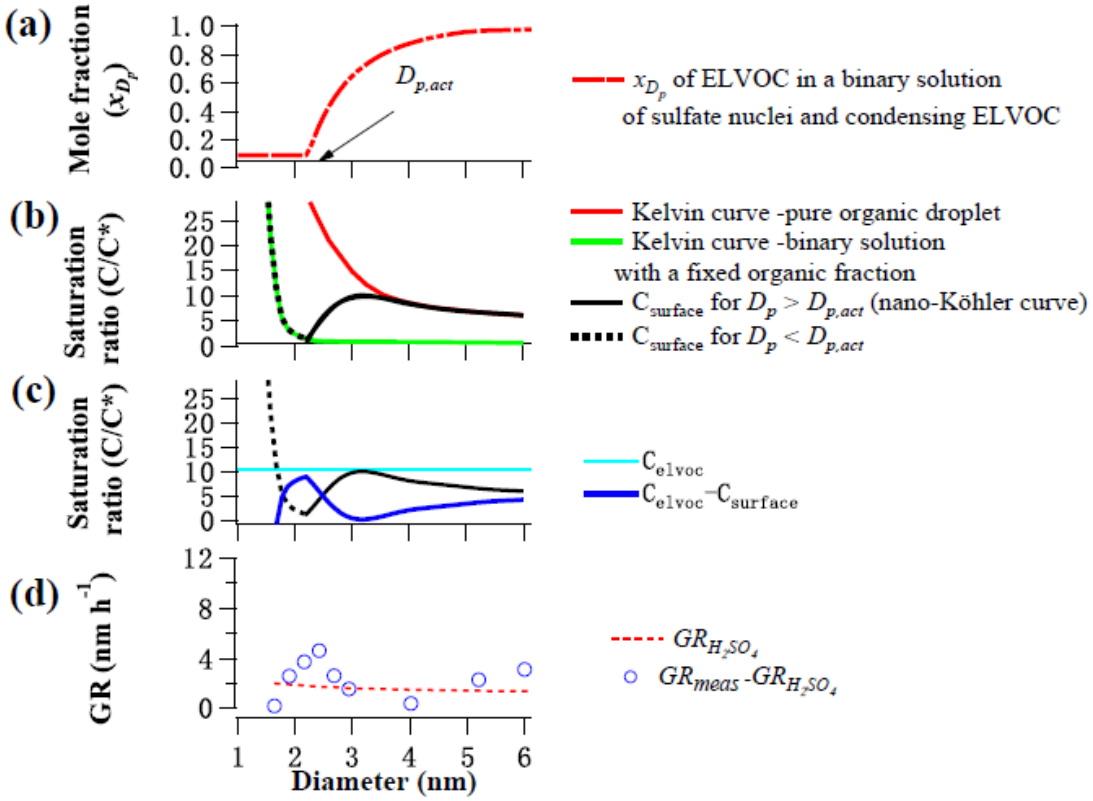


Figure 8. (a) Mole fraction of organics in a binary solution of sulfate nuclei and activating organics (ELVOC) in a new particle. (b) Kelvin equilibrium curves. (c) Gas phase concentration of the organic vapor. (d) Growth rate GR_{H2SO4}.

Summary

- Infrequent nucleation was limited by both low concentrations of gaseous precursors and high temperature and RH in summer.
- In more polluted winter and spring atmosphere, precursor supply was not limiting anymore; nucleation occurred once meteorological conditions were favorable.
- However, for the further growth of sub-3 nm particles to CCN-active sizes, anthropogenic gaseous precursors again became limiting factors.

THANK YOU!