Enhancement in CCN concentrations during new particle formation events

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Keywords: NPF, CCN, particle growth, organics

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New particle formation (NPF) events are an important source of cloud condensation nuclei (CCN). However, the contribution of atmospheric NPF events to the global CCN budget remains the single largest uncertainty (Kerminen, et al., 2012). Therefore, information on mechanisms and the vapors responsible for NPF and growth would be needed for accurate prediction of CCN in order to realistically capture aerosol radiative effects. In our study, we investigate the effects of particle formation and growth and chemical ageing of pre-existing particles on CCN number concentration during the NPF events based on the observations made at a central Europe research station, Melpitz, Germany.

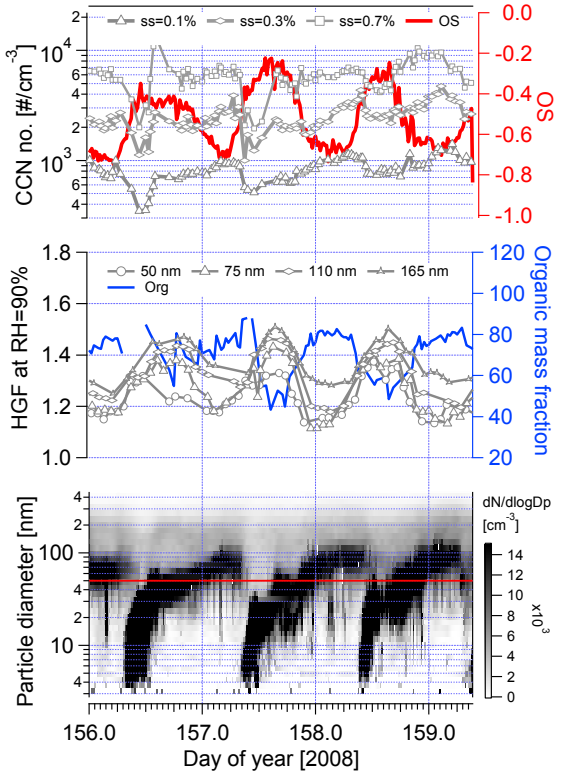

Fig. 1: Time series of different parameters during NPF events. The detail description is given in the text.

Investigations of the contribution of NPF on CCN concentrations are based on the size-resolved chemical composition (AMS), particle hygroscopic growth factor (HGF), particle number size distributions, sulfuric acid [H_2SO_4] and OH radical concentrations at Melpitz in 2008. The NPF bursts and continuous growth were continuously observed (Fig.1.). As a first step, the critical diameter (D_{crit}) at which 50% of the particles were activated was calculated from κ:

$$D_{crit} = \left(\frac{4A^3}{27\kappa \ln^2 S_C}\right)^{1/3}$$ (Petters and Kreidenweis, 2007)

Here, κ is calculated from size-resolved AMS data using the ZSR mixing rule.Secondly, the number concentration of potential CCN is derived by integrating the number size distribution of particles larger than D_{crit}. The oxidation state (OS) was calculated from AMS data according to Kroll et al, (2011) for the whole size range (PM1). The organic mass fraction was taken in the size range 50-200 nm (mobility diameter).

Comparing the increase of CCN number concentrations at the start time of the NPF bursts (B in table 1) and at the end time of NPF bursts (E in the table 1) shows that on average, the CCN number concentrations are enhanced by the factors of two between the beginnings and the end of NPF events. However, the increments are varied with varied super saturation values (SS). (See Table 1)

Table 1: Comparisons in CCN number concentrations (unit: /cm^3) before and after NPF events.

SS	0.1%		0.3%		0.7%	
DOY	B	E	B	E	B	E
157	419	913	1510	2696	2923	5982
158	542	1054	1313	3798	2298	6508
159	800	1277	2592	3651	4941	6794

As shown in Fig. 1, the HGF of 50-165 nm particles increased from 1.2 to 1.5 during NPF event. This is most likely due to that pre-existing particle gets aged quickly in a photochemically active environment associating with NPF. Both condensation of H_2SO_4 and aging of organic fraction (as indicating by increase in OS) could lead to increase in particle hygroscopicity. Correspondingly, their ability to act as CCN enhanced even though the NPF did not grow to CCN size during this timeframe. During nighttime, particles grew quickly due to condensation of organic vapors. At the same, the particle HGF declined significantly. As consequence, the ability of particles acting as CCN activity decreased significantly. Typically H_2SO_4 concentration is close zero during nighttime. Thus, the semi-volatile organic condensation plays a key role in particle growth. More organics condensed onto newly formed CCN-sized particles and higher supersaturations are needed to activate particles with the same size. This process weakened the contribution of NPF to CCN. But, the net effect leads to increase in CCN concentration, as listed in Table 1. More analysis for a longer-data set at Melpitz site, are ongoing and will be discussed in our presentation.

Petters, M.D. and Kreidenweis, S.M. (2007). Atmos.Chem. Phys., 7, 1961-1971.; Kerminen, et al., 2012, Atmos. Chem. Phys., 12, 12037-12059.; Kroll et al., 2011, Nature Chemistry 3,133–139.