Tracking ambient new particle formation by an expansion-type CPC

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Ambient new particle formation (NPF) is frequently observed in field measurements and is thought to be the dominant source of aerosol particles governing aerosol *number* concentration on global scale (Kulmala *et al.*, 2004). Modeling results show that about half the global cloud condensation nuclei (CCN) originate from nucleation (Merikanto *et al.*, 2009). Despite several hundred individual studies on atmospheric observations of aerosol nucleation, a fundamental understanding is still lacking. This is largely due to experimental shortcomings in the detection and characterization of nanoparticles in the size range between 1 – 2 nm.

Here we report on NPF measurements obtained by the versatile Size Analyzing Nuclei Counter (Pinterich *et al.*, 2013). The vSANC is a newly built expansion-type CPC based on its predecessor SANC (e.g., Wagner et al., 2003). vSANC was designed to allow maximum flexibility for its application in field and laboratory studies. The inlet lines are optimized for minimum particle losses and it can be operated with various working fluids. Concentration measurement is performed by a multi-angle Mie scattering detector delivering five independent concentration measurements simultaneously. Counting efficiency measurements have shown that vSANC detects 10-20% of the particles below 1.5 nm in diameter. Thus vSANC can be used as a nucleation mode particle counter. Moreover, time resolved scattered light measurements enable precision determination of vapor supersaturations. Thereby quantitative heterogeneous nucleation experiments can be performed.

During spring 2014 we participated in the spring campaign at the SMEAR II station in Hyytiälä, Finland. One of our main goals was the detection of the smallest possible clusters at the very beginning of NPF events. To this end, vSANC was operated in a three-stage cycle where we chose three different expansion ratios to achieve size-dependent particle activation at different vapour saturation ratios. Accordingly, concentration measurements in the size bins 2-6 nm, 6-10 nm and >10 nm were performed sequentially. As homogeneous nucleation was observed at the highest supersaturation applied basically all particles and clusters present in the expansion chamber can be considered activated. The time-resolved monitoring of the scattered light signals allows us to separate heterogeneously nucleated droplets from homogeneous ones (Winkler *et al.*, 2008).

For the most part of the campaign we used *n*-propanol as working fluid. Remarkably, NPF was observed only when the nucleation temperature was low enough (<0°C) suggesting that nucleation temperature may play an important role in nanoparticle detection (Kupc *et al.*, 2013). Also it was observed that high relative humidity (RH) led to binary homogeneous nucleation in the vSANC (see Figure 1). At RH close to 100% the scattered light signal reached concentrations of almost 10^7 cm^{-3}, long before the pressure drop reached its final value. No such behaviour was observed for low RH which typically was the case for NPF days.

Diurnal concentration evolution during NPF events showed that particle concentrations as high as 10^4 cm^{-3} were detected in the smallest size bin of vSANC about 2-3 hours before the NPF signal appeared in a differential mobility particle sizer (DMPS). The lower DMPS detection limit of 3 nm suggests that the high vSANC signals can be attributed to the appearance of newly formed sub-3 nm particles. In the next steps we will compare the vSANC results with data acquired with Particle Size Magnifier (Vanhanen *et al.*, 2011) and with air ion spectrometers to explore the existence of the predicted high-concentration pool of nucleating clusters (Kulmala et al., 2013).

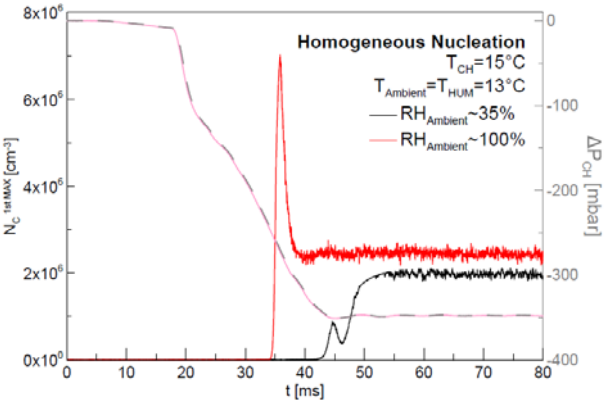

Figure 1. Pressure drop (magenta) and scattered light signals (red and black) at two different RH conditions.

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