Long-term hygroscopic properties of ambient aerosol in a boreal environment, as measured by the size-resolved Cloud Condensation Nuclei counter (CCNc)

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Ambient aerosol CCN and hygroscopic properties were measured with a size-segregated CCNc in a boreal environment of Southern Finland at the SMEAR II station. The instrumental setup operated at five levels of supersaturation S covering a range from 0.1% to 1%, and measured particles with a size range of 20–300 nm. The instrument has been operating since February 2009, and a total of 29 non-consecutive months of measurement data are presented.

Critical diameter D_c was calculated for each activation spectrum as a mid-point of the fitted sigmoid curve, i.e. 50% of particles activated. For each pair of D_c and S, hygroscopicity parameter κ was calculated according to the EH1 Köhler model using Eq. A30 in Rose *et al*. (2008). The surface tension of pure water of 0.072 J m^{-2} was assumed. No normalisation of activation spectra took place prior to the curve fitting; therefore, discussion below concerns only the CCN-active fraction of the ambient aerosol.

The median D_c and κ for the whole dataset in this study are reported at 75 nm and 0.22, respectively. The median κ shows that aerosol in the boreal environment of Southern Finland is less hygroscopic than the global continental and European continental averages, as presented by Pringle *et al*. (2010), most likely due to a presence of a large organic fraction within the aerosol. The aerosol is, however, more hygroscopic than ambient aerosol in an Amazon rainforest, the European high alpine site or the mountainous forest (Fig. 1). The Figure also reveals that the rate of change of κ with size at the SMEAR II station is higher than in three other locations, indicating differences in the species of condensing material and the oxidation and aging processes.

A month of CCNc data was analysed in parallel with aerosol mass spectrometer (AMS) measurements in order to determine the effect of atmospheric refractory species on κ. A negative correlation was observed between κ and the organic mass fraction, and a positive correlation was found between κ and sulphate and ammonia species, logically following the behaviour of these species in the atmosphere with respect to their affinity for water.

The biogenic emissions in the boreal environment of Southern Finland make ambient aerosol of >100 nm in diameter less hygroscopic in the spring and summer time, compared to other seasons – the seasonal variation of D_c needed for CCN activation is important to remember when estimating CCN concentrations from aerosol particle number measurements. The participation of biogenic emissions in photochemical reactions is responsible for introducing a diurnal pattern in the behaviour of aerosol hygroscopicity in the spring and summer for particles ~50 nm in diameter. The diminished photochemistry, temperature and biogenic activity in autumn and winter result both in the highest seasonal hygroscopicity of larger ambient aerosol and in the absence of the diurnal trend of D_c and κ for particles of any size.

No distinct effect of atmospheric new particle formation (NPF) on D_c and κ was observed. Ambient aerosol was found to be internally mixed in the summer, and externally mixed during the rest of the year.

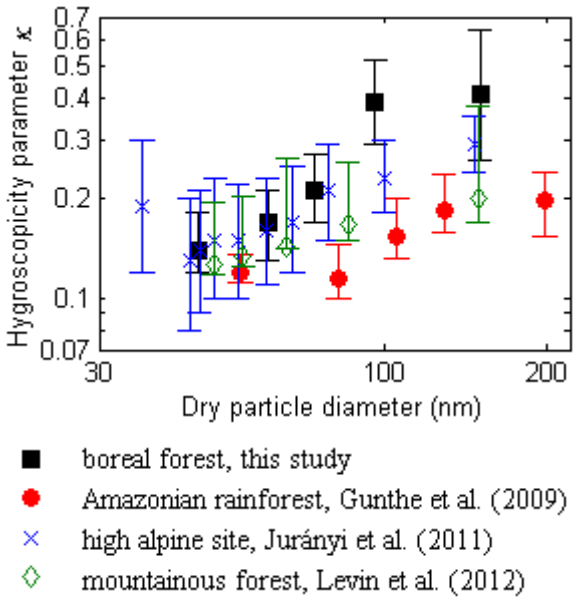

Figure 1. Relationship between particle dry size (taken as D_c) and κ for several studies. Shown are the median values, with error bars being 25th and 75th percentiles (for Gunthe *et al*. (2009) percentiles estimated from the original publication).

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