

Semi-empirical parameterization for sub-20 nm particle growth

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New particle formation occurs frequently in the atmosphere (Kulmala *et al*, 2004). According to global model estimations these newly formed particles contribute significantly to global CCN (cloud condensation nuclei) concentrations after growing large enough (Spracklen *et al*, 2008).

It has been observed that sub-20 nm particle growth is size-dependent (Manninen *et al*, 2010; Yli-Juuti *et al*, 2011) and has in some environments seasonal pattern comparable to that of atmospheric biogenic organics, e.g. monoterpenes (Yli-Juuti *et al*, 2011). These results among others suggest that atmospheric nanoparticles are growing mainly by condensation of low-volatile organic vapors and that different fractions of these condensable organics are condensing onto different sized particles due to Kelvin effect.

The challenge in current atmospheric models is to accurately yet efficiently represent atmospheric nanoparticle growth by biogenic and anthropogenic organics. In this work we introduce a simple semi-empirical parameterization for sub-20 nm particle growth that distributes secondary organics to the nanoparticles according to their size (1.5-3 nm, 3-7 nm, 7-20 nm) and this way captures particle growth rates observed in the atmosphere. The parameterization includes particle condensational growth by sulfuric acid, by secondary organics from monoterpene oxidation (SORG_{MT}) and by additional condensable non-monoterpene organics (“background”) (Eq. 1).

$$I_{tot}(d_p) = I_{SA}(d_p) + k(d_p)I_{SORG,MT}(d_p) + k_{bg}(d_p)I_{SORG,bg}(d_p), \quad (1)$$

where I_{tot} is the total mass flux onto particles with a diameter of d_p , I_{SA} is the mass flux of sulfuric acid, $I_{SORG,MT}$ and $I_{SORG,bg}$ are the mass fluxes of the SORG_{MT} and the background organics, respectively. $k(d_p)$ and $k_{bg}(d_p)$ are factors for distributing the SORG_{MT} and the background organics to particles with diameter of d_p . I_{tot} , I_{SA} and $I_{SORG,MT}$ were obtained from ambient data. $I_{SORG,bg}$, k and k_{bg} values for the three particle sizes were obtained with a least-square method by fitting Eq. 1 to the ambient data. Ambient growth rate data was acquired from particle number size distribution measurements at six continental sites over Europe. The longest time series of 7 years (2003 to 2009) was obtained from a boreal forest site in Hyytiälä, Finland.

The parameterization was able to reproduce observed particle growth rates and their seasonal cycles

with good accuracy (see Figure 1 for Hyytiälä as an example). The weighting factors (k) for distributing the SORG_{MT} were 0 for 1.5-3 nm, 0.7 for 3-7 nm and 1 for 7-20 nm. The concentration of the background organics was comparable to the concentrations of the SORG_{MT} and sulfuric acid, around $6 \cdot 10^7 \text{ cm}^{-3}$. The weighing factors (k_{bg}) for distributing the background organics were 0.3 for 1.5-3 nm, 0.8 for 3-7 nm and 1 for 7-20 nm. Simulations with a global model, GEOS-Chem-TOMAS (Pierce *et al*, 2012), suggest that the “background” can be linked to biogenic organics that are oxidized in the presence of anthropogenic pollution (Spracklen *et al*, 2011).

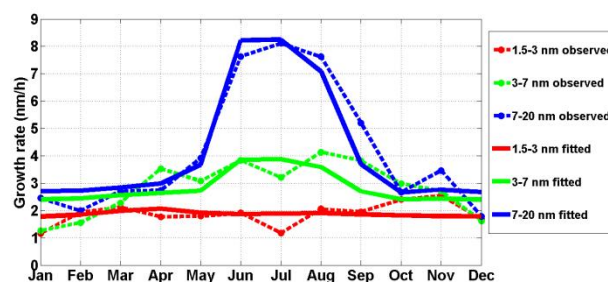


Figure 1. Comparison between particle growth rates (1.5-3, 3-7 and 7-20 nm sizes) obtained from ambient observations and from the parameterization in Hyytiälä.

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