A novel method to determine the growth rate of a growing particle mode

T. Anttila[1], J. Leppä[1] and H. Lihavainen[1]

[1]Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland

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Presenting author email: tatu.anttila@fmi.fi

Atmospheric new particle formation (NPF) events are characterized by appearance of particles at the lowest measured diameter range and subsequent emergence of a growing mode which can often be visualized as a "banana" (Fig. 1). These events have atmospheric relevance as they provide a potentially large source of particles that act as seeds for cloud droplets. A key parameter associated with this phenomenon is the growth rate (GR) of a newly-formed aerosol mode as it contains information on the mechanisms underlying atmospheric NPF events and provides constraints for numerical models simulating NPF (Yli-Juuti *et al.*, 2011). Several methods for determining GR have been developed, but obtaining accurate size- and time-resolved data on GR can still be challenging with the current methods.

Here we propose a method for determining the growth that is based on calculating the count mean diameter CMD over a certain size interval [$d_{min}(t)$, $d_{max}(t)$], and using the following relation:

$$\mathrm{GR}(t) = \frac{\mathrm{d}}{\mathrm{d}t}\mathrm{CMD}(t) = \frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\int_{d_{\min}(t)}^{d_{\max}(t)} n(d_p) d_p \mathrm{d}d_p}{\int_{d_{\min}(t)}^{d_{\max}(t)} n(d_p) \mathrm{d}d_p} \right) \quad (1)$$

where d_p is the particle diameter and $n(d_p)$ is the particle number size distribution. In practice, GR is obtained through numerical differentiation and integrals are replaced by sums.

Two main challenges in applying equation (1) are: how to determine parameters d_{min} and d_{max}, and how to eliminate "noise" that arises from numerical differentiation of CMD together with "noise" in the original size distribution data? We approached the former issue by first calculating the diameter corresponding to the peak concentration in the growing mode as a function of time, $d_{peak}(t)$. A log-normal fit was then performed around d_{peak} which yields the geometric standard deviation of the mode as a function of time, $\sigma_g(t)$. Finally, the following relations were used: $d_{min}(t) = d_{peak}(t)/(\sigma_g(t)\times\alpha)$, and $d_{max}(t) = d_{peak}(t)\times(\sigma_g(t)\times\alpha)$. Here α is a free dimensionless parameter that, together with σ_g, determines the size range over which CMD is calculated. Regarding the latter issue, GR was calculated numerically with finite differences, and the resulting time series was then smoothed using an algorithm described by Stickel (2010).

The approach was evaluated against NPF events generated with an aerosol dynamic model (Anttila *et al.*, 2010), where GR was held constant. To imitate atmospheric data, particle concentrations were perturbed by ±10%. Such comparison is illustrated in Figures 1 and 2. As can be seen, after a growing mode has emerged, the smoothed GR tracks the correct GR accurately despite the fact that non-smoothed GR contains significant amount of noise.

Systematic study on the performance of the method will be presented later, and the feasibility of the method in interpreting atmospheric and laboratory data will be investigated.

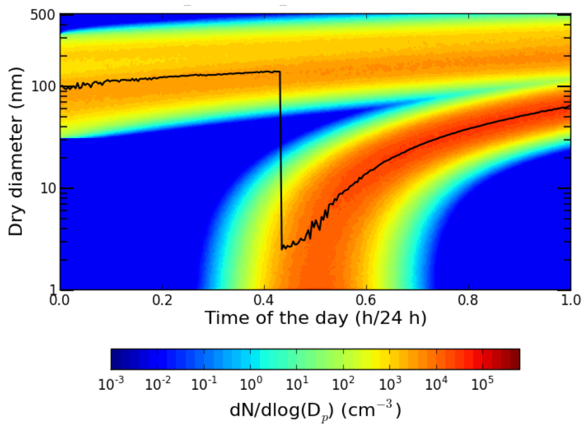

Figure 1. Time evolution of a particle size distribution and CMD (black line).

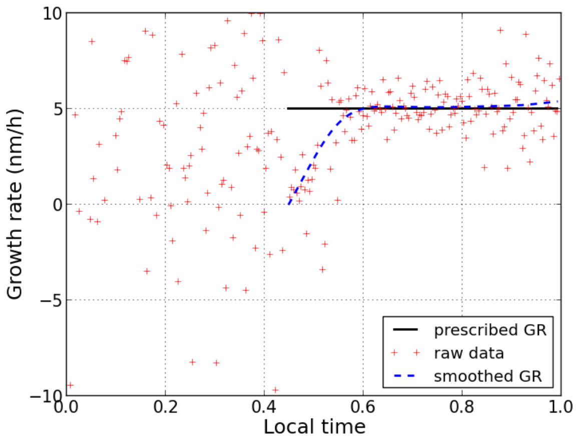

Figure 2. Various growth rates (see legend) during the simulation depicted in Figure1.

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