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COMPARISON OF GROWTH RATE CALCULATION METHODS FOR ATMOSPHERIC ULTRAFINE PARTICLE FORMATION MODELS

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Abstract: Particle growth rate (GR) is a crucial parameter beside the formation rate that largely determines the shape of aerosol particle size distribution evolution in atmospheric new particle formation (NPF) events, the concentration of nanoparticles and their further fate in the atmosphere. The GR can be estimated from measured particle size distribution data by several methods, which can yield different values for GR. Often the known methods are excessively sensitive to various kind of fluctuations within the observed data. We propose a new method for GR automatic estimation. This method considers the entire NPF event profile and is less sensitive to the fluctuations within the measured data that can be due to variation in atmospheric boundary layer mixing, pollutant transport and inhomogeneity of the atmospheric NPF itself. The results of the GR estimation applying conventional maximum concentration method and the proposed method agree within the standard deviation of the corresponding values, but the GR values obtained by the proposed method are less scattered. The conventional method may provide outlier GR values up to at about 25% of the NPF cases, whereas the proposed method yields the outlier values only at about 1% of the cases.

Key words: *Atmospheric new particle formation (NPF), NPF event analysis, particle growth rate (GR) estimation.*

INTRODUCTION

The composition and formation of atmospheric ultrafine particles (the ones up to about hundred nanometer in diameter) is a key factor that determines the cloud properties and, therefore, climate change, as well as affects the human health (Ren et al., 2020; Kwon et al., 2020; Wang et al., 2023). Atmospheric new particle formation (NPF) depends on several parameters, including particle diameter growth rate (GR). The GR is theoretically modelled and/or calculated by several methods from the measured size distributions. The most conventional methods are the maximum concentration method (Hirsikko et al. 2005) and the log-normal distribution function (mode fitting) method (Dal Maso et al. 2005). Recently also cetrain new methods appeared, e.g. Mask Region-based Convolutional Neural Network, abbreviated as mask R-CNN (Su et al. 2022) but recent methods are, as a rule, not easy to implement. We compare the results obtained by the most common maximum concentration method with the results by a new method, proposed and developed by us. The GR comparison results are based on the more than 30-year aerosol data measured at the Tahkuse Observatory, Estonia.

DESCRIPTION OF THE CONVENTIONAL AND THE PROPOSED METHODS

Let particle size distribution data be presented in the form of matrix C(k,j), where an element of the matrix contains particle concentration C within the size fraction k and at the time point j. Conventionally the new particle formation studies just look the events that evolve within one calendar day, therefore all the time points j=1,..,jj belong to a particular day. The time points and the size fractions k=1,...,kk depend on the device employed for the measurements to obtain (record) the data and on the settings of this particlular device. In this study we use the of Tahkuse Air Ion spectrometer (Hõrrak et al., 2000, 2003) and Neutral cluster and Air Ion Spectrometer (https://www.airel.ee/products/nais/) measurements, where jj=288 (data recorded every 5 minutes) and kk=24 (size fractions from about 1.6 nm up to 25 nm).

One aim of NPF studies is to estimate the particle growth rates (GR) according to the observed new particle size distribution evolution shape. Within the conventional maximum concentration method the evolution of the measured particle size distribution during a NPF event is examined starting from the first size fractions where the new particles are detected, and the moment when the concentration of particles reaches maximum value in each size fraction is determined. The GR is obtained as the slope of a line fitted in the data pairs of the determined moment of maximum concentration and geometric mean diameter of the particles of the corresponding size fraction (Hirsikko et al. 2005). Conventionally, the NPF events should start not before sunrise, therefore only the time points $j=j_1,..,jj$ are taken into account, where j_1 is determined by the sunrise time.

Our proposed method is based also on the particle size distribution data, but it handles the data in a different way. For every time series of measured size distribution number concentrations C(k,j) at fraction k, where $j=j_1,...,jj$ correspond to specific day, it computes the 45%, 66% and 98% quantiles $q_0(k)$, $q_1(k)$ and $q_2(k)$, respectively. Next it proceeds just with the concentrations that are between q_1 and q_2, whereas q_0 that is used to calculate the weight of a particular concentration as described below. In this way the method can exclude the near noise level concentration values below q_1 and also the measurement outliers above q_2 but still considers the elevated concentrations between q_1 and q_2 that probably belong to a new particle formation event. Taking into account all the concentrations that are between q_1 and q_2, the method forms two arrays *data_X* and *data_Y*, where the first array will contain the time points j of the concentrations and the second array will contain the diameters of the size fractions, where these concentrations ($q_1(k) < C(k,j) < q_2(k)$) are located. More in detail, looking at every particular concentration C(k,j) the method adds several new points into the both arrays (adds the value j into *data_X* and the value k into *data_Y*), whereas the number of the new points depends on the ratio of the concentration to q_0. Thereby in the arrays *data_X* and *data_Y* the higher concentrations are represented by numerous points and the lower concentrations are represented by less numerous points, therefore the locations (k, j) of the elevated concentrations are more weighty. The array *data_X* contains the times as a usual independent variable but *data_Y* contains the concentrations in an implicit form, the concentrations are given as the number of points determined by particular concentrations at specific size fraction, higher concentrations generate larger number of points. Finally the method takes *data_X* as independent variables and *data_Y* as function values and computes a regression function that connects *data_X* and *data_Y*, the regression will be a second order polynomial curve. From this curve the method computes the GR values for the size ranges 1.6–3 nm, 3–7 nm and 7–25 nm. The method is based on MATLAB software and uses its data processing capabilities.

Figure 1 depicts an example of a new particle formation event and the GR values calculated by conventional method and by our proposed method.

The GR values obtained by two methods for the size fractions of 1.6–3 nm, 3–7 nm and 7–25 nm are comparable but still different. As a reason that leads to such differences, the conventional maximum concentration method is limited by the data within a particular range of particles and ignores the entire duration (shape) of the event. Inhomogeneity in atmospheric NPF in time and space can cause difficulties and uncertainties in new particle GR estimation. In the NPF case depicted in the Figure 1 this applies to the evolution within the first size range (the fractions up to 3 nm) where particle growth is not continuous but includes a gap at about 11–12. As a result, conventional method yields particular GR value equal to 0.33 nm h^{-1}. In contrast, our proposed method takes into account the entire shape of the NPF and is less sensitive to the fluctuations within the measured data or within the underlying NPF and growth processes itself. It can smooth out some local uncertainties, the GR value by this method is equal to 0.81 nm h^{-1} .

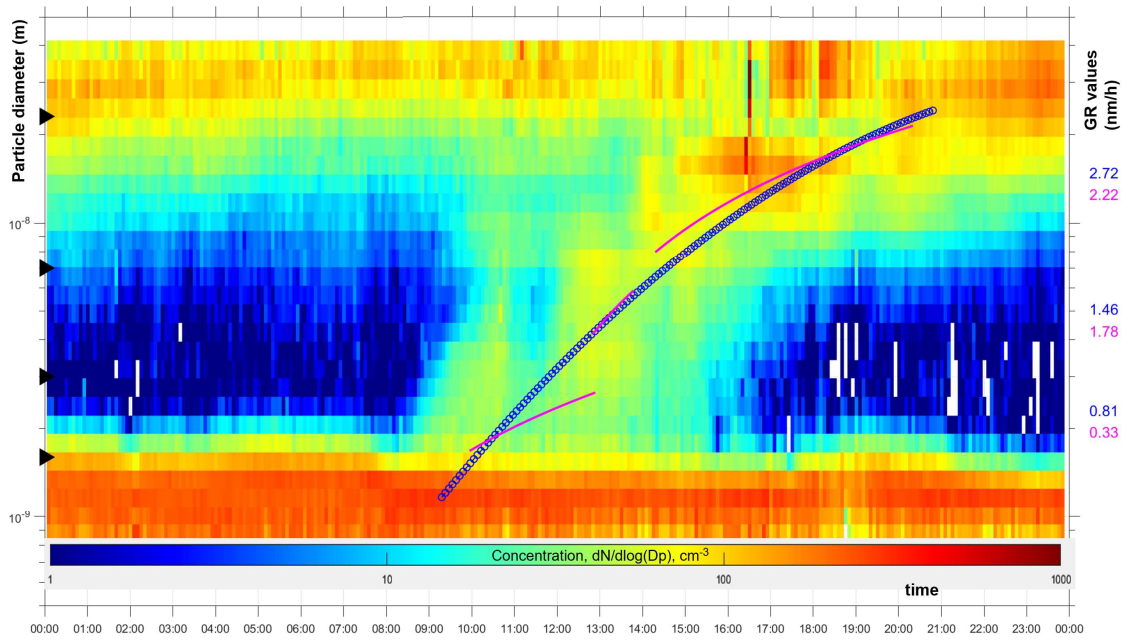

Figure 1. An example of a new particle formation event with the aerosol particle growth curves calculated by different methods. GR values are calculated by conventional maximum concentration method (purple lines and numbers at the right edge of the figure, one for each size range) and by our proposed method (blue line and numbers). The data recorded at Tahkuse Observatory on March 3, 2012 with Neutral cluster and Air Ion Spectrometer (device number NAIS17).

RESULTS AND DISCUSSION

In general, the GR results of both compared methods agree within the standard deviation of the corresponding values, but the proposed method commonly can yield more reasonable GR values (the values that are between 0.1*average and 10*average) even in the case of fluctuating and/or noisy data, and the GR values obtained by the proposed method are less scattered as also seen in the Figure 2. For the most prominent and clear NPF events (so called class 1 events), the conventional method provides reasonable GR values at least at about 75% of the NPF cases (days), depending on certain size class (1.6–3 nm, 3–7 nm and 7–25 nm). The proposed method can yield the GR values at least at 99% of the cases. The conventional method gives average GR values 0.8, 2.7 and 3.9 nm h^{-1} (standard deviations 0.8, 2.1 and 1.9 nm h^{-1}), for the particle size classes 1.6–3 nm, 3–7 nm and 7–25 nm, respectively. The GR values by the proposed method are 1.2, 1.9 and 3.6 nm h^{-1} (standard deviations 0.7, 0.7 and 1.2 nm h^{-1}). The differences in average GR values obtained by these two methods are less than the corresponding standard deviations but the average GR values obtained by these methods still statistically different, according to t-test at 95% significance level.

Figure 2 demonstrates the variations of estimated annual mean GR values within the time period 1989–2022. The GR values calculated by the conventional maximum concentration method are more scattered than the ones by our proposed method but the general trends (elevated or lower annual mean values at certain years) of the GR values during the 33 year time period are similar for the both methods. As mentioned above, the conventional method is limited by the data within a particular range (or size class) of particles and therefore more sensitive to any noise/fluctuations within these particular size regions. Our proposed method takes into account larger size range (considers the entire event profile) and can, therefore, smooth out some local fluctuations, outliers and/or noise in the data.

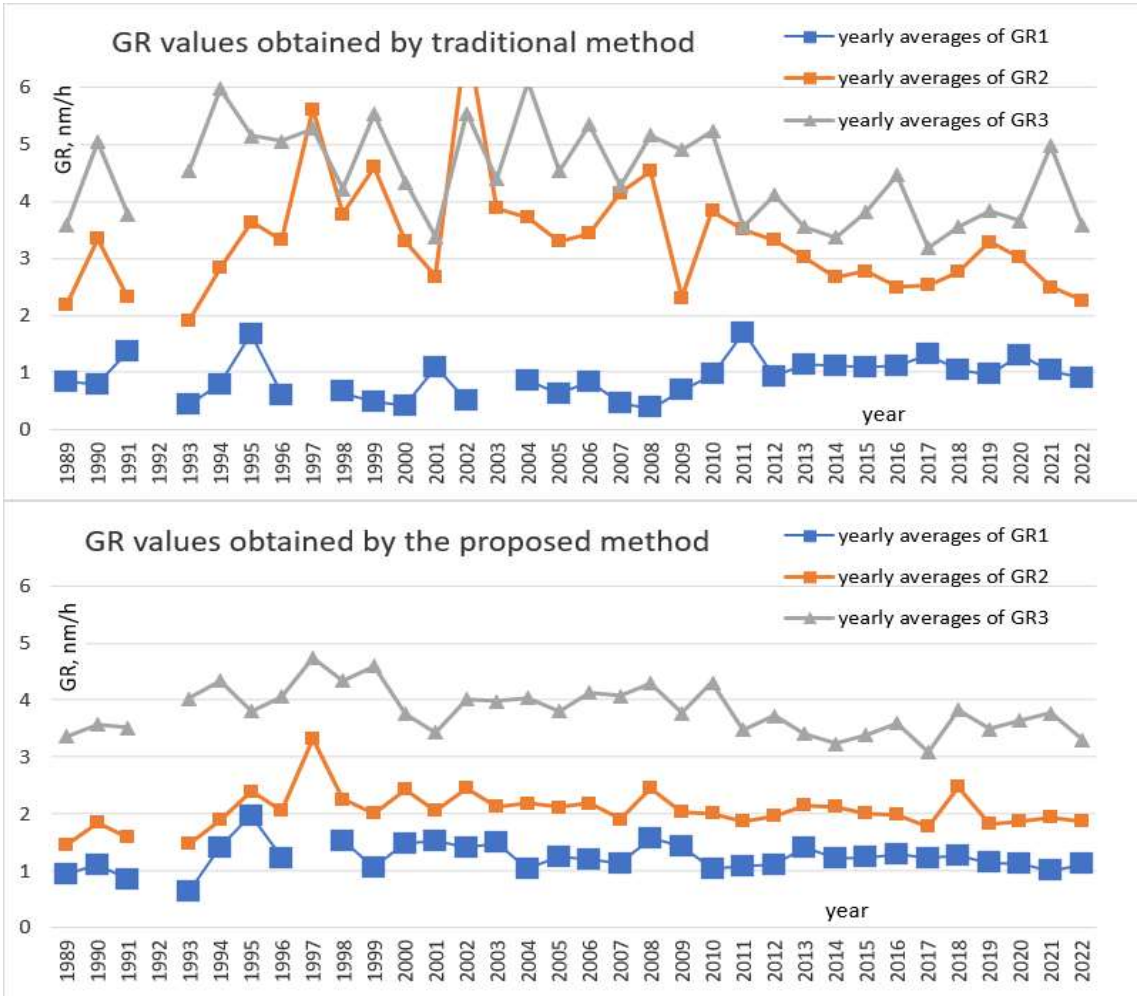

Figure 2. Variations in the yearly average GR values calculated by two different methods. GR1, GR2 and GR3 are the growth rates calculated for the size ranges 1.6–3 nm, 3–7 nm and 7–25 nm, respectively. Data recorded by air ion and aerosol particle spectrometers at Tahkuse Observatory between 1989 and 2022.

The differences in the GR values obtained in this work can be compared against certain former results. The paper by Yli-Juuti et al. (2011) demonstrated that the GR values of nucletion mode size range (3-20 nm) particles obtained with the maximum concentration method were typically larger than the values determined with the mode fitting method. The median of the difference between the GRs calculated with those two methods was 1.1 nm h^{-1}. In our case these differences are 0.4, -0.8 and -0.3 nm h^{-1}, for the size ranges 1.6–3 nm, 3–7 nm and 7–25 nm, respectively. Therefore, our proposed method tends to yield somewhat larger GR values for smaller particles and somewhat smaller GR values for larger particles but the average differences are below 1 nm h^{-1}.

The proposed method yields reliable results. The GR values of the conventional method and the proposed method agree within the standard deviation and the time trends of the GR values during the time period 1989–2022. At the same time, the GR values by the proposed method are less scattered. The proposed method yields outlier GR values only for about 1% of the strong NPF events occurred in the years 1989–2022, therefore it can be recommended especially for the tasks that need automatic GR calculations.

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