BOREAL ENVIRONMENT RESEARCH 10: 337–355 ISSN 1239-6095
Helsinki 24 October 2005

Evaluation of an automatic algorithm for fitting the particle number size distributions

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Hussein, T., Dal Maso, M., Petäjä, T., Koponen, I. K., Paatero, P., Aalto, P. P., Hämeri, K. & Kulmala, M. 2005: Evaluation of an automatic algorithm for fitting the particle number size distributions. *Boreal Env. Res.* 10: 337–355.

The multi log-normal distribution function is widely in use to parameterize the aerosol particle size distributions. The main purpose of such a parameterization is to quantitatively describe size distributions and to allow straightforward comparisons between different aerosol particle data sets. In this study, we developed and evaluated an algorithm to parameterize aerosol particle number size distributions with the multi log-normal distribution function. The current algorithm is automatic and does not need a user decision for the initial input parameters; it requires only the maximum number of possible modes and then it reduces this number, if possible, without affecting the fitting quality. The reduction of the number of modes is based on an overlapping test between adjacent modes. The algorithm was evaluated against a previous algorithm that can be considered as a standard procedure. It was also evaluated against a long-term data set and different types of measured aerosol particle size distributions in the ambient atmosphere. The evaluation of the current algorithm showed the following advantages: (1) it is suitable for different types of aerosol particles observed in different environments and conditions, (2) it showed agreement with the previous standard algorithm in about 90% of long-term data set, (3) it is not time-consuming, particularly when long-term data sets are analyzed, and (4) it is a useful tool in the studies of atmospheric aerosol particle formation and transformation.

Introduction

Atmospheric aerosol particles affect the climate of the Earth both directly by scattering incoming radiation and indirectly by acting as cloud condensation nuclei (e.g. Seinfeld and Pandis 1998, Haywood and Boucher 2000, Lohmann and Feichter 2005). Besides affecting the Earth's climate, aerosol particles in urban areas cause the loss of visibility (e.g. Finlayson-Pitts and Pitts 2000). In addition, atmospheric aerosol particles, especially ultrafine particles (diameter < 100 nm), have harmful health effects even at relatively low particulate mass concentrations; therefore, it has been suggested that the toxicity of inhaled aerosol particles is not only due to their mass but also due to their number and surface area or even due to their chemical composition (e.g. Osunsanya *et al*. 2001).

Typically, the amount of submicron aerosol particles varies spatially in the atmosphere from few tens cm^{-3} in background conditions to several hundred thousands cm^{-3} in a polluted urban environment (e.g. Seinfeld and Pandis 1998). In addition to the spatial distribution found in the number concentration of aerosol particles, their physical characteristics also exhibit both temporal and spatial variability. For example, aerosol particles residing in the continental air mass differ considerably in terms of size and composition from marine aerosols (e.g. Jaenicke 1993). On the other hand, the total number concentration of submicron aerosol particles may exceed 10^6 cm^{-3} during intensive new particle formation bursts, when the number concentration is dominated by newly formed ultrafine particles (e.g. O'Dowd *et al*. 2002, Kulmala *et al*. 2004).

Because of the high variability of the properties of atmospheric aerosol particles, simplifications are needed before the parameterized particle number size distributions can be used as input data in global climate models or regional air quality models. One way to parameterize the aerosol particle number size distributions is the multi log-normal distribution function (e.g. Whitby 1978). In other words, aerosol particle size distributions have been represented by a sum of several log-normal distributions (e.g. Hoppel *et al.* 1994, Mäkelä *et al*. 2000, Birmili *et al*. 2001, Hussein *et al*. 2004). One of the aims of such a parameterization is to describe the aerosol particle number size distribution by using as few parameters as possible. This representation of the distribution also allows straightforward comparison between several data sets of aerosol particles.

In this study, we evaluated an algorithm that parameterizes aerosol particle number size distributions with the multi log-normal distribution function. The current algorithm is completely automatic in the way that it does not need a user control on the initial assumptions of the log-normal parameters. In addition, it applies several tests to automatically decide the required number of log-normal modes that provide the best fit for the particle number size distribution. It is important to mention that the current algorithm is an upgrade of the previous algorithms presented by Hussein *et al*. (2004, 2005). The upgrade was developed to make the current algorithm suitable for different atmospheric conditions without a need to state the initial conditions of the input parameters. Sensitivity analysis as well as comprehensive comparison between results of the current algorithm and a previous manual algorithm by Mäkelä *et al*. (2000) will be demonstrated in the following sections.

The physical size distribution of aerosol particles

Whitby (1978) showed considerable body evidence that most aerosol particle size distributions can be fitted with the multi log-normal distribution function. The multi log-normal distribution function f is expressed mathematically by

$$f\left(D_{\mathrm{p}}, \bar{D}_{\mathrm{pg},i}, N_i, \sigma_{\mathrm{g},i}\right) = \sum_{i=1}^{n} \frac{N_i}{\sqrt{2\pi}\log\left(\sigma_{\mathrm{g},i}\right)} \times \exp\left[-\frac{\left[\log\left(D_{\mathrm{p}}\right) - \log\left(\bar{D}_{\mathrm{pg},i}\right)\right]^2}{2\log^2\left(\sigma_{\mathrm{g},i}\right)}\right], \quad (1)$$

where D_{p} is the diameter of an aerosol particle. Three parameters characterize an individual log-normal mode i: the mode number concentration N_i, geometric variance $\sigma^2_{\mathrm{g},i}$, and geometric mean diameter $D_{\mathrm{pg},i}$. The number of individual log-normal modes that characterize the particle number size distribution is denoted by n.

For the purpose of discussion and interpretation, the basic four-modal model and nomenclature will be used (Fig. 1). The rationale behind this model is based on the previous observations of particle number size distributions found in the literature (e.g. Whitby *et al*. 1991, Mäkelä *et al*. 1997, 2000, Morawska *et al*. 1998, Birmili *et al*. 2001, Shi *et al*. 2001, Koponen *et al*. 2002, 2003, Wehner *et al*. 2002, Gidhagen *et al*. 2003, Lehmann *et al*. 2003, Hussein *et al*. 2004, 2005).

In the current study we concentrate on the modal structure of aerosol particles within the submicron size range. Therefore, the model presented herein consists of up to three log-normal modes in the fine particle size range. In general, urban and suburban aerosol particle number size distributions in the submicron size range can be characterized by using three log-normal modes (e.g. Hussein *et al*. 2005). However, and under certain conditions,

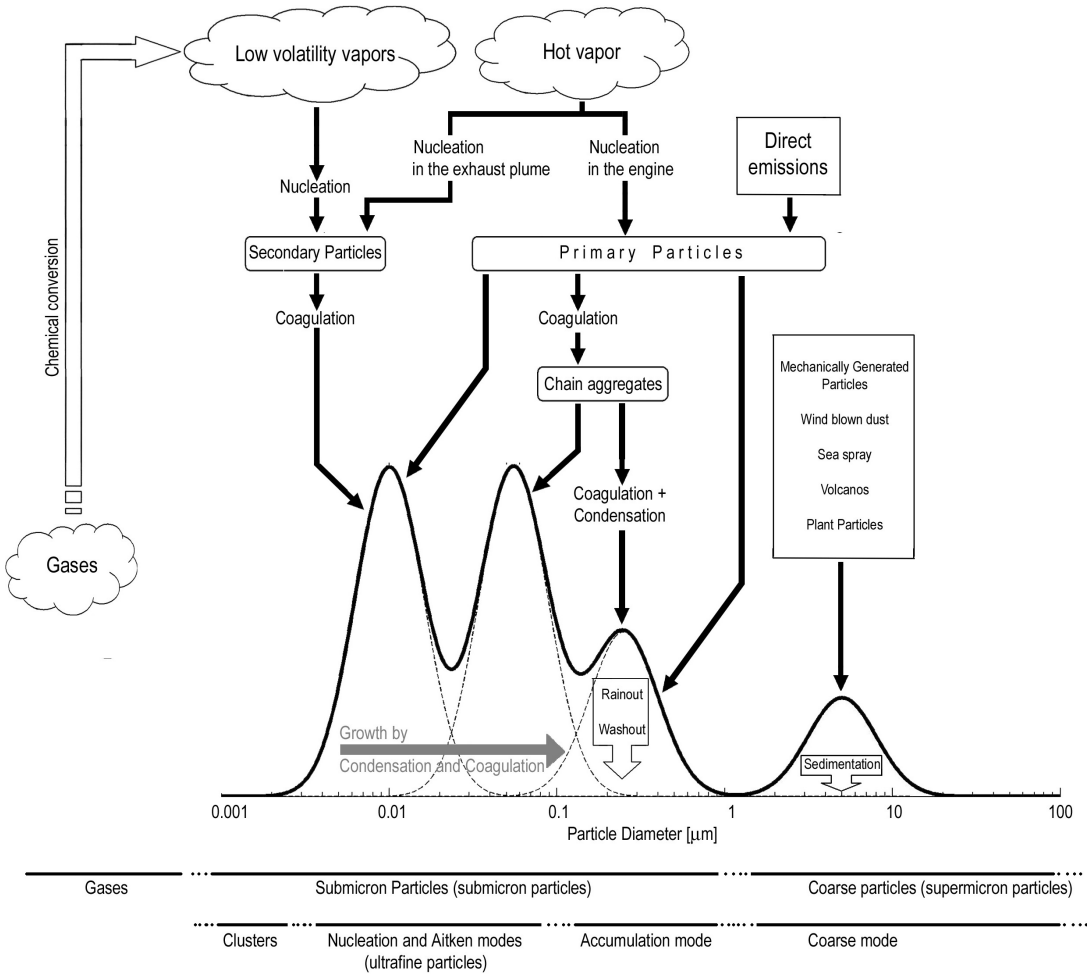

Fig. 1. Idealized schematic of an atmospheric particle number size distribution (Hussein 2005). Principle modes, sources, and particle formation and removal mechanisms are indicated. The figure is a modified version of that presented by Whitby (1978).

it is also possible to characterize the urban and suburban aerosol particle number size distributions by assuming two or more than three log-normal modes (e.g. Birmili *et al*. 2001). Aerosol particle number size distributions in other background environments (such as rural, polar, etc.) may also consist of two log-normal modes or even a single log-normal mode (e.g. Whitby *et al*. 1991, Mäkelä *et al*. 2000, Koponen *et al*. 2002, 2003).

Development of a multi log-normal fitting algorithm

The main objective of the fitting is to obtain the log-normal parameters (N_i, $\sigma^2_{g,i}$, and $D_{pg,i}$) in Eq. 1 that best fit the measured particle number size distribution and thus describe the properties of the atmospheric aerosol particle population. Different research groups have had their own numerical algorithms to fit the aerosol particle number size distributions with the multi log-normal distribution function (e.g. Whitby 1978, Whitby *et al*. 1991, Mäkelä *et al*. 2000, Birmili *et al*. 2001). There have also been several commercial algorithms widely in use such as DistFit™ (Chimera Technologies, USA). As a measure of the fitting quality, some of the algorithms are based on the least square (LSQ) method and others are based on the normalized chi-square method with which

the measured particle number size distribution and its fitting are compared. Typically, the total number, surface area, or volume concentration of either the mode being fitted or the whole particle size distribution can be used as a normalization parameter.

The user interface varies considerably between different algorithms. For example, in their algorithm, Mäkelä *et al*. (2000) introduced a user control on the initial assumptions of the number of individual log-normal modes and their parameters. The user control is not usually preferred especially when long-term data sets are involved. The recent version of DistFit™ provides the possibility to fit either single or several sequential particle number size distributions stored in several data files and the user can pre-define the maximum number of log-normal modes to be used in the fitting.

In general, the LSQ value between the measured particle number size distribution and its fitting can be used to control the fitting quality. However, the best-fit curve can be represented by several combinations of the fitting parameters that lead to the same LSQ value. These combinations of the fitting parameters can be mathematically acceptable, but not all of them are physically acceptable. For that reason, Hussein *et al*. (2004) introduced an algorithm that forces the accumulation mode to have large geometric mean diameters rather than small values keeping in account that the fitting quality is not affected. Similarly, the nucleation mode was also forced to have small geometric mean diameters. These two constraints captured the nucleation and accumulation modes more accurately in comparison with unconstrained fitting algorithms. The constraints introduced by Hussein *et al*. (2004) were suitable to urban aerosols only. An advantage of the algorithm by Hussein *et al*. (2004) is that it did not need a user control on the initial fitting parameters to be defined beforehand. Instead, the algorithm was able to obtain these initial parameters. Later on, the algorithm by Hussein *et al*. (2004) was modified to be also suitable for suburban aerosol particle number size distributions as well as urban aerosols (Hussein *et al*. 2005). One disadvantage of the algorithms by Hussein *et al*. (2004, 2005) is that they required the user to define the number of log-normal modes needed in the fitting. Another disadvantage is that these two algorithms were only suitable for the urban and suburban atmospheric aerosol particles.

In this study, we present the recent development of the fitting algorithm based on the principles by Hussein *et al*. (2004, 2005). The current algorithm was developed to be suitable for any type of aerosol particle number size distributions regardless of their environment type. Another feature in the current algorithm is that it starts fitting by assuming a maximum number of log-normal modes and it automatically reduces the number of modes, if possible, without affecting the fitting quality. The maximum number of modes is defined by the user. Hereafter, we describe the algorithm by assuming three log-normal modes, and the algorithm can adjust itself automatically to reduce the number of modes if necessary. It is also possible to start the fitting by assuming a higher number of modes if necessary; the code can be easily modified.

Fitting procedure

In the current algorithm, the fitting procedure consists of several steps. In the first step, the algorithm assumes that the particle number size distribution consists of three log-normal modes. During this step the algorithm obtains the initial values of the log-normal parameters by iteration: $\sigma_{g,i}$ is iterated between 1.2 and 2.1 for all modes, and $D_{pg,i}$ and N_i are iterated as illustrated in Fig. 2.

In the second step, the algorithm adjusts the log-normal parameters to converge the LSQ value between the measured particle number size distribution and its fitting. This step consists of three sequentially repeated steps: (1) the algorithm iterates $D_{pg,i}$ values within moving particle diameter ranges constrained around the best values of $D_{pg,i}$ obtained from the previous iteration step, (2) the algorithm then iterates $\sigma_{g,i}$ values in the same manner as described for $D_{pg,i}$, and (3) then the algorithm calculates N_i based on an optimization procedure that will be described later in this section. The algorithm repeats these three steps sequentially until the change of the LSQ value is smaller than a certain tolerance (e.g. smaller than 5%). After that, the

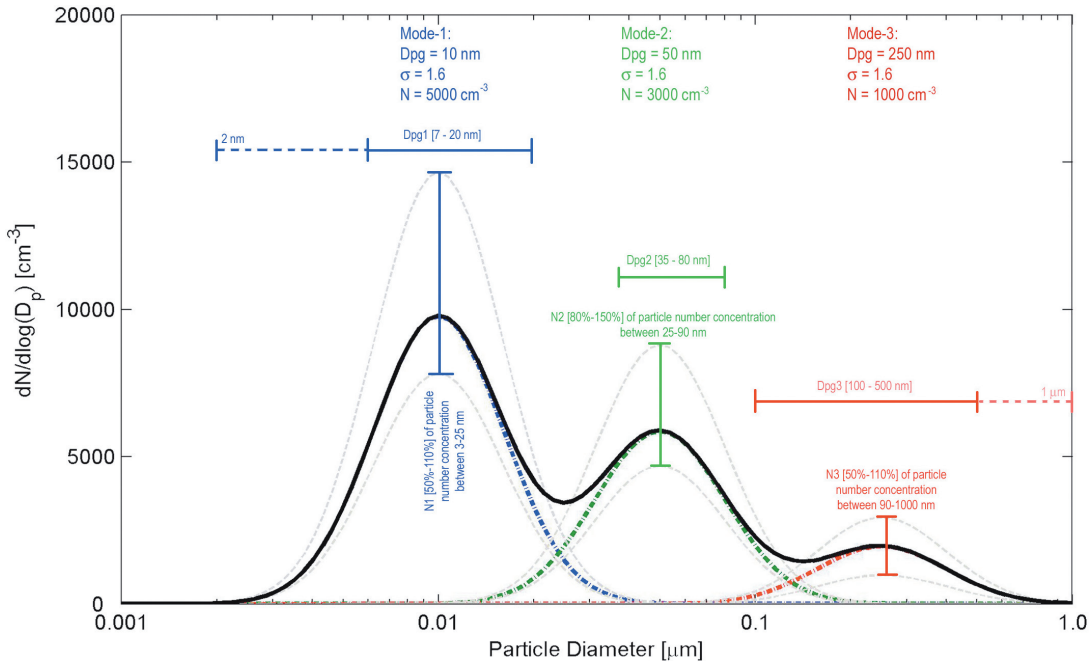

Fig. 2. A sketch showing the iteration ranges for $D_{pg,i}$ and N_i in the first step of the fitting procedure. The solid black line illustrates a measured particle number size distribution being fitted, the dashed lines illustrate the lower and upper limits of the iteration of the number concentrations iterations, and the solid dark dashed lines illustrate the individual log-normal modes that best fit the particle number size distribution. The upper limit of the geometric mean diameter of mode-3 can be extended to 1 μm if needed, and the lower limit of the geometric mean diameter for mode-1 can be extended to 2 nm.

algorithm tests the possibility of reducing the number of individual modes based on the overlapping hypothesis between individual modes. This test is based on statistical investigations of the allowed/forbidden separation of two adjacent modes with respect to their number concentrations.

In the third step, if any two adjacent modes justify an overlapping condition that they can be replaced by a single mode, the algorithm repeats the second step by assuming two log-normal modes only. In this step, a decision is made whether both modes are located in the ultrafine particle (UFP diameter < 100 nm) size range, or whether one mode lies in the UFP size range and the other does not. In the case of two UFP modes, the iterations of $D_{pg,i}$ are the same as described in Fig. 2 for mode-1 and mode-2. In the case of only one UFP mode, the value of $D_{pg,i}$ of the UFP mode (mode-1) is iterated between 3 and 200 nm, and that of the second mode (mode-2) is iterated between 100 and 1000 nm.

In the fourth step, if the two modes obtained in the third step can be replaced by a single mode, the algorithm proceeds with this step to eliminate one mode. In a single log-normal mode fitting, the algorithm iterates σ_g as described in the first step, and it iterates $D_{pg,i}$ between 3 and 1000 nm.

It should be mentioned that the algorithm does the fitting individually for each particle number size distribution. Therefore, the fitting parameters of individual particle number size distributions do not influence each other.

As mentioned earlier, the fitting quality is controlled by the LSQ value between the measured particle number size distribution and its fitting. The LSQ value is calculated as follows:

$$\mathrm{LSQ} = \frac{1}{m}\sqrt{\sum_{D_p}\left[B\left(D_p\right) - f\left(D_p, \bar{D}_{pg,i}, N_i, \sigma_{g,i}\right)\right]^2}, \quad (2)$$

where m is the number of measured size sections in the particle number size distribution and $B(D_p)$ is the measured particle number size distribution to be fitted.

Calculation of the mode number concentrations

As described in the previous versions of the algorithm (Hussein 2004, 2005), it is possible to calculate the mode number concentrations (N_i) based on the optimization method, which is computationally more efficient than obtaining the mode number concentrations by iteration.

We can re-write Eq. 1 by using the values of the parameters $D_{pg,i}$ and $\sigma_{p,i}$ that provided the best fit for the measured particle number size distribution in the form

$$B\left(D_p\right) \rightarrow f\left(D_p, \bar{D}_{pg,i}, N_i, \sigma_{g,i}\right) = \sum_{i=1}^{n} N_i A_i\left(D_p, \bar{D}_{pg,i}\sigma_{g,i}\right) \quad (3)$$

where

$$A_i\left(D_p, \bar{D}_{pg,i}\sigma_{g,i}\right) = \frac{1}{\sqrt{2\pi}\log\left(\sigma_{g,i}\right)} \times \exp\left[-\frac{\left[\log\left(D_p\right) - \log\left(\bar{D}_{pg,i}\right)\right]^2}{2\log^2\left(\sigma_{g,i}\right)}\right] . \quad (4)$$

Optimization of Eq. 3 with respect to N_i as follows:

$$\frac{\partial}{\partial N_i}\sum_{1}^{j}\left[B\left(D_p\right) - \sum_{i=1}^{n} N_i A_i\left(D_p, \bar{D}_{pg,i}\sigma_{g,i}\right)\right]^2 = 0 \quad (5)$$

generates n-linear algebraic equations that can be represented in the matrix form

$$\sum_{D_p} A_k\left(D_p, \bar{D}_{pg,i}\sigma_{g,i}\right) A_i\left(D_p, \bar{D}_{pg,i}\sigma_{g,i}\right) N_i = \sum_{D_p} A_i\left(D_p, \bar{D}_{pg,i}\sigma_{g,i}\right) B\left(D_p\right) , \quad (6)$$

where k and i have the values 1, 2, …, n. We can solve the set of n-linear equations by using simple matrix algebra. The mathematical solution provides exact estimations of the mode number concentrations N_i when the best-fit values of the parameters $D_{pg,i}$ and $\sigma_{g,i}$ are well known.

The solutions of N_i might result in particle number concentrations below zero for some modes. In that case, the algorithm repeats the fitting procedure to adjust the best-fit values for the parameters $D_{pg,i}$ and $\sigma_{g,i}$ until the negative number concentrations disappear. It is also possible that the mode number concentration of any mode is zero. In that case, the algorithm ignores those modes.

Overlapping conditions between adjacent modes

As mentioned before, the main objective of the fitting is to parameterize the particle number size distributions with the multi log-normal distribution function with the fewest number of modes, i.e. as few parameters (N_i, $\sigma^2_{g,i}$, and $D_{pg,i}$) as possible. This fact requires the elimination of some modes without affecting the fitting quality. For that reason, we visually investigated the so-called overlapping conditions, which mean that any two adjacent modes can be replaced by a single mode.

We investigated the overlapping conditions by testing the relative separation between any two adjacent modes ($D_{pg,i}$) and the ratio between their variance ($\sigma_{g,i}$); then a minimum limit of number concentrations is allowed for the secondary mode. We then obtained a map of the allowed and forbidden locations of adjacent modes with respect to each other. However, the translation of such conditions into logical statements in the algorithm would make the computation time consuming and rather complicated. Therefore, we included only such logical statements that are needed for the common aerosol particle number size distributions. In the following we summarize the logical statements that we see the most important ones to be included in the algorithm.

If any of the following conditions is true after fitting the size distribution by using three modes, the fitting is performed by using one UFP mode and a mode with a geometric mean diameter > 100 nm:

1. The logarithmic difference between the geometric mean diameters of mode-1 and mode-2 is < 0.24 and the geometric mean diameter of mode-3 is > 100 nm.
2. The logarithmic difference between the geometric mean diameter of any two adjacent modes is < 0.33 and the geometric mean diameter of mode-3 is > 100 nm.
3. The logarithmic difference between the geometric mean diameters of mode-1 and mode-2 is < 0.33 and the number concentration of

mode-1 or mode-2 is smaller than 25% of the total particle number concentration.
4. The logarithmic difference between the geometric mean diameters of any two adjacent modes is < 0.5 and the number concentration of mode-2 is smaller than 25% of the total particle number concentration.

If any of the following conditions is true after fitting the size distribution by using three modes, the fitting is performed by using two UFP modes:

1. The geometric mean diameter of mode-3 is < 90 nm.
2. The number concentration of mode-3 is smaller than 3% of the total particle number concentration and the geometric mean diameter of the mode is < 100 nm.
3. The logarithmic difference between the geometric mean diameters of any two adjacent modes is < 0.33 and the geometric mean diameter of mode-3 is < 100 nm.
4. The logarithmic difference between the geometric mean diameters of mode-3 and mode-2 is < 0.24.

Evaluation against a standard algorithm

Pre-defined particle number size distributions

We tested the current algorithm by fitting pre-defined particle number size distributions (Table 1). For comparison, we also fitted the pre-defined particle number size distributions using the previous algorithm by Mäkelä *et al*. (2000). As mentioned earlier, the previous algorithm by Mäkelä *et al*. (2000) requires a user decision on the initial assumptions of the number of modes and their parameters. The previous algorithm by Mäkelä *et al*. (2000) is based on the MATLAB built-in functions "leastsq.m" (Quasi-Newton method) and "fmins.m" (Nelder-Mead simplex method). The user was not told about the number of pre-defined modes in any of the cases. We compared the performance of the current algorithm with the previous algorithm by Mäkelä *et al*. (2000) only because the latter one can be considered a standard procedure because it utilizes the built functions of the MATLAB compiler. Another reason to compare the current algorithm to this previous one is the availability of parameterized data set by that algorithm.

For the single mode size distributions described in cases 1–3 (Fig. 3a–c), the previous algorithm provides perfect results while the results of the current algorithm deviated slightly from the ideal values (Tables 2 and 3). This deviation is only because the current algorithm is based on iterative calculations for $D_{pg,i}$ and $\sigma_{g,i}$. If the number of iterations of each parameter increased, we would expect to have better results.

When the particle number size distribution was assumed to consist of two log-normal modes that can be easily distinguished from each other (case 4, Fig. 3d), both algorithms predicted the modes correctly. Cases 5 and 6 look like a single

Table 1. Log-normal parameters of the pre-defined particle number size distributions used for the current algorithm evaluation against the previous algorithm by Mäkelä *et al.* (2000).

	Mode-1			Mode-2			Mode-3		
	$D_{pg,1}$	$\sigma_{g,1}$	N_1	$D_{pg,2}$	$\sigma_{g,2}$	N_2	$D_{pg,3}$	$\sigma_{g,3}$	N_3
Case 1	3	1.6	1000	–	–	–	–	–	–
Case 2	1.5	1.6	1000	–	–	–	–	–	–
Case 3	50	1.6	1	–	–	–	–	–	–
Case 4	25	1.6	1000	75	1.3	1000	–	–	–
Case 5	50	1.5	1000	60	1.9	1000	–	–	–
Case 6	50	1.6	1000	100	1.6	1000	–	–	–
Case 7	7	1.5	1000	50	1.5	1000	250	1.5	1000
Case 8	20	1.5	1000	50	1.5	1000	250	1.5	1000
Case 9	20	1.5	1000	50	1.5	1000	100	1.5	1000

mode and the user of the previous algorithm assumed a single mode. As expected, the current algorithm predicted only one mode.

If three well distinguishable modes were present (cases 7 and 8, Fig. 3g–h), the previous algorithm provided perfect results, whereas the results of the current algorithm deviated slightly from the ideal values. The reason for this deviation was again that the current algorithm is based on iterative calculations of $D_{pg,i}$ and $\sigma_{g,i}$.

In case 9, we assumed that mode-2 and mode-3 are identical and overlap each other so that they are no longer distinguishable (Fig. 3i). As expected, the current algorithm replaced them with a single mode. If the user of the previous algorithm assumed two modes, the results were close to those obtained by the current algorithm. Correct results were obtained if the user assumed three modes.

Algorithm performance based on long-term data analysis

To further evaluate the performance of the current algorithm, we parameterized a long-term data set of the measured particle number size distributions at Hyytiälä that were previously fitted and parameterized by Mäkelä *et al*. (2000) (Table 4). This long-term data set spans one year starting from 1 February 1996. Both methods predicted three modes in about 42.6% of the cases and two modes in about 16.4% of the cases. On average, both parameterizations in these cases were quite similar.

In 31% of the cases the current algorithm predicted three modes and the previous algorithm predicted only two modes. However, the first mode predicted by the current algo-

Table 2. Parameterization of the pre-defined particle number size distributions (Table 1) as obtained by the current algorithm. The maximum number of modes was assumed to be three, and the algorithm was allowed to reduce the number of modes without affecting the least square values between the size distribution and its fitting.

	Mode-1			Mode-2			Mode-3		
	$D_{pg,1}$	$\sigma_{g,1}$	N_1	$D_{pg,2}$	$\sigma_{g,2}$	N_2	$D_{pg,3}$	$\sigma_{g,3}$	N_3
Case 1	2.90	1.60	999	–	–	–	–	–	–
Case 2	1.41	1.67	1066	–	–	–	–	–	–
Case 3	49.19	1.60	1	–	–	–	–	–	–
Case 4	25.83	1.65	1052	75.57	1.28	945	–	–	–
Case 5	52.54	1.64	1947	–	–	–	–	–	–
Case 6	73.02	1.84	2027	–	–	–	–	–	–
Case 7	6.85	1.50	1001	46.36	1.50	992	249.00	1.52	1025
Case 8	22.07	1.67	1105	50.47	1.68	951	251.18	1.47	949
Case 9	25.19	1.77	1517	83.40	1.62	1513	–	–	–

Table 3. Parameterization of the pre-defined particle number size distributions (Table 1) as obtained by the previous algorithm by Mäkelä *et al.* (2000). The user assumed the number of modes without previous knowledge about the ideal cases.

	Mode-1			Mode-2			Mode-3		
	$D_{pg,1}$	$\sigma_{g,1}$	N_1	$D_{pg,2}$	$\sigma_{g,2}$	N_2	$D_{pg,3}$	$\sigma_{g,3}$	N_3
Case 1	3.00	1.60	1000	–	–	–	–	–	–
Case 2	1.50	1.60	1000	–	–	–	–	–	–
Case 3	50.00	1.60	1	–	–	–	–	–	–
Case 4	25.00	1.60	1000	75	1.30	1000	–	–	–
Case 5	58.80	1.84	1558	–	–	–	–	–	–
Case 6	70.70	1.72	2128	–	–	–	–	–	–
Case 7	7.00	1.50	1000	50.00	1.50	1000	250.00	1.50	1000
Case 8	20.00	1.50	1000	50.00	1.50	1000	250.00	1.50	1000
Case 9	22.20	1.54	1277	80.20	1.57	1761	–	–	–

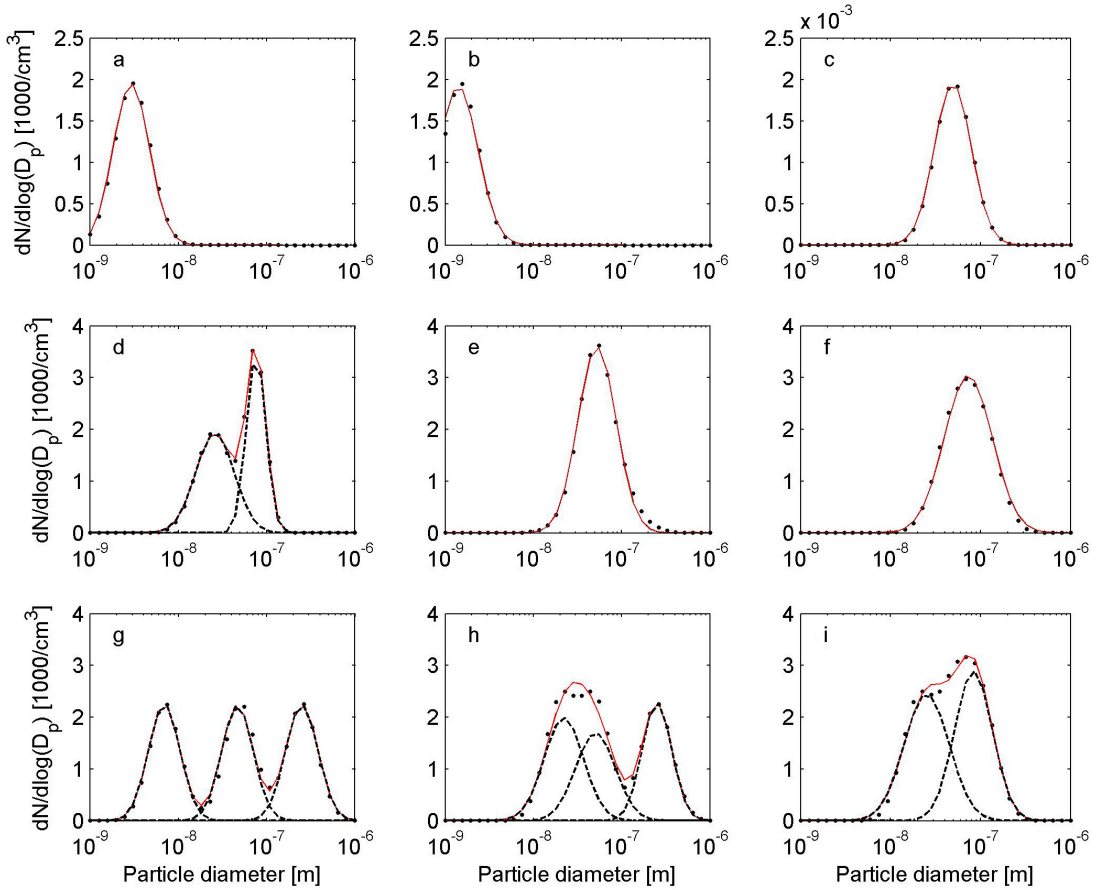

Fig. 3. Evaluation of the current algorithm against pre-defined cases of particle number size distributions (Table 1). The "dots" represent the pre-defined cases and the "solid lines" represent the best-fit curve predicted by the algorithm. The dashed lines represent the individual modes.

rithm was only a minor mode with very low particle number concentration (< 100 cm^{-3}). After neglecting this mode both parameterizations were quite similar. This additional mode was predicted by the current algorithm only to converge the LSQ value, whereas in the

Table 4. Evaluation of the current algorithm against a long-term aerosol data set previously parameterized by Mäkelä *et al.* (2000). The comparison was performed for one year data set of particle number size distributions (dry diameter 3–500 nm) measured at SMEAR II. The total number of cases is 41 692.

	Number of cases	Number of modes predicted	Mode-1	Mode-2	Mode-3
Current study	17759	3	14.2	56.9	192.0
Mäkelä *et al.* 2000	17759	3	14.7	56.1	196.0
Current study	6846	2	–	38.8	144.0
Mäkelä *et al.* 2000	6846	2	–	41.8	171.0
Current study	12899	3	15	63.6	196.0
Mäkelä *et al.* 2000	12899	2	–	57.8	190.0
Current study	4188	2	26.7	–	131.0
Mäkelä *et al.* 2000	4188	3	14.9	50.0	188.0

previous method it was omitted based on the knowledge that this mode is due to fluctuations in the measurements at very low number concentrations for particles smaller than 20 nm in diameter.

In 10% of the cases the current algorithm predicted two modes and the previous algorithm predicted three modes. One of the three modes was eliminated by the current algorithm based on the overlapping test. However, as a consequence of this elimination, the remaining two modes should shift toward each other in order to converge the LSQ value.

The agreement between the algorithms can be considered reasonable, since they predicted the same number of modes in about 90% of the cases after eliminating mode-1 in the cases where the current algorithm predicted three modes instead of two as in the previous algorithm. It should be noticed that the user decision significantly influences the parameterization by deciding the number of modes. The significant difference between the parameterizations in 10% of the cases does not imply that the results presented by Mäkelä *et al.* (2000) are not valid. However, we believe that the current algorithm performs better in parameterizing urban and suburban particle number size distributions. Another advantage of using the current algorithm is the fact that it is automatic. In addition, parameterizing different data sets with the same algorithm reduces the user influence, which helps in the comparison process.

Evaluation against case studies of measured particle number size distributions

The performance of the current algorithm is further illustrated by fitting particle number size distribution spectra measured in different environments (boreal forest, remote region, polar, and urban) and under different conditions (background, traffic, long-range transport, local pollution, and new particle formation). The selected examples presented herein are only for illustration and they do not represent the absolute characteristics of particle number size distributions.

Background conditions

Boreal forest

The SMEAR II station is located at Hyytiälä in southern Finland (Vesala *et al.* 1998). Hyytiälä is a background area with only minor local emissions. Particle number size distribution measurements have been performed continuously at SMEAR II since 1996 (Kulmala *et al.* 2001). The mean particle number concentration at SMEAR II is < 3000 cm^{-3} (Laakso *et al.* 2003). In the following we present an evaluation of the algorithm for the particle number size distributions measured at SMEAR II under background conditions, i.e. in the absence of new particle formation events and regional/local pollution episodes. Later on, we will present the evaluation of the algorithm during a new particle formation event at SMEAR II.

During background conditions, the algorithm predicted two major modes (Fig. 4a): Mode-1 had a geometric mean diameter ranging between 10 and 100 nm. Mode-2 showed less variability than mode-1 and had geometric mean diameters between 100 and 200 nm. The total particle number concentration was obtained by integrating the measurements over the particle diameter range 3–350 nm (Fig. 4b), after which it was compared with the sum of the number concentrations of individual modes obtained by the algorithm. As can be seen, total particle number concentrations obtained from the fitting were very close to those integrated from the measurements.

Remote region

The SMEAR I station is located at Värriö (approximately 250 km north of the Arctic Circle) in the eastern part of Finnish Lapland. SMEAR I is an example of a remote arctic region (e.g. Hari *et al.* 1994, Laakso *et al.* 2003, Vehkamäki *et al.* 2004). For most of the time, the air at the station is rather clean without local pollution. Occasionally, polluted air masses arrive from the northeastern direction. The mean particle number concentration at SMEAR I is < 1500 cm^{-3} (e.g. Laakso *et al.* 2003).

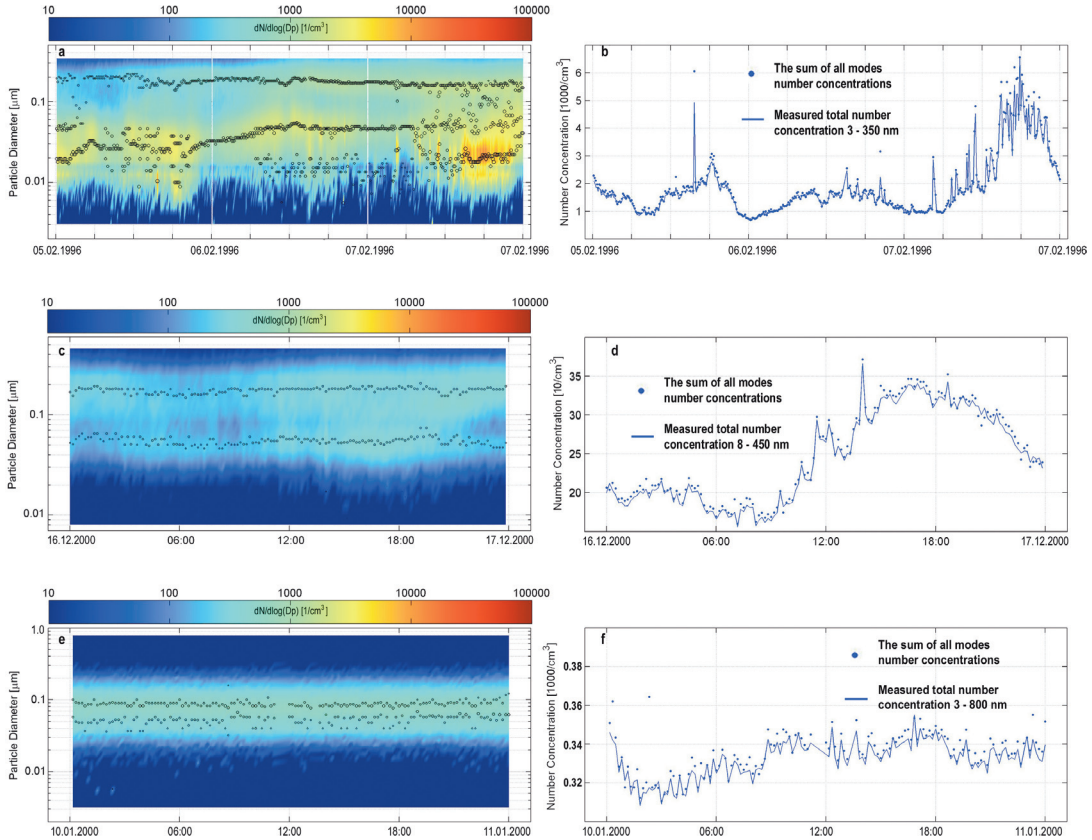

Fig. 4. Selected case studies of particle number size distribution spectra measured at background measurement stations. The left panel shows the particle number size distribution spectra. The circles represent the geometric mean diameters of individual modes and the size of the circle is proportional to the mode number concentration. The right panel shows the corresponding total particle number concentrations. — **a** and **b**: The SMEAR II measurement station in a boreal forest in southern Finland. — **c** and **d**: The SMEAR I measurement station in a remote region in northern Finland. — **e** and **f**: Polar aerosols at Aboa in Antarctica.

The aerosol particle number size distributions consisted of two modes during background conditions at SMEAR I (Fig. 4c). The geometric mean diameters of the individual modes were similar to those found at SMEAR II, except that the number concentrations of individual modes were two orders of magnitude lower at SMEAR I than at SMEAR II. Both the mode geometric mean diameters and their number concentrations showed less variation at SMEAR I than SMEAR II. Another difference between the modal structure at SMEAR I and SMEAR II was the lower limit of the geometric mean diameter of mode-1 that can be 30 nm at SMEAR I, whereas at SMEAR II it can be as small as 10 nm.

The evaluation of the algorithm for this kind of data set measured at SMEAR I showed rather stable results. This is because of the quality of the measured particle number size distributions at SMEAR I.

Polar aerosol particles

The Finnish Antarctic station at Aboa is an example of a polar region (Koponen *et al*. 2003). Aboa is located on the Nunatak Basen some 130 km from the coastline of Antarctica. On average, the particle number concentrations in the South Pole range between 100 and 200 cm^{-3} during the austral summer and can be < 20 cm^{-3} during the winter (e.g. Shaw 1988, Ito 1993). In coastal Antarctica, the average particle number concentrations are typically almost an order of magni-

tude higher than in the South Pole (e.g. Jaenicke *et al*. 1992, Grass 1993, Koponen *et al*. 2003).

As predicted by the current algorithm (Fig. 4e), the modal structure of the particle number size distribution may consist of two modes during the background conditions. Both modes showed rather stable geometric mean diameters and number concentrations. The geometric mean diameter of mode-1 varied between 40 and 60 nm while that of mode-2 varied between 70 and 120 nm.

Previously, Koponen *et al*. (2003) analyzed the modal structure of the particle number size distributions presented in Fig. 4e by using the algorithm by Mäkelä *et al*. (2000). Their results emphasized the existence of a similar number of modes but with slightly different geometric mean diameters: 30–50 nm for mode-1 and 80–100 nm for mode-2.

Urban aerosols

Helsinki city is an urban region. It is situated on a fairly flat coastal area by the Baltic Sea (e.g. Hussein *et al*. 2005). There have been continuous measurements of aerosol particle number size distributions in Helsinki since 1997. The annual-average total particle number concentration in Helsinki is below 20 000 cm^{-3} but the daily-median concentration can be as high as 50 000 cm^{-3} during the winter season (Hussein *et al*. 2004).

For a particle number size distribution spectrum measured in Helsinki during a typical workday, the algorithm did not reduce the number of modes to fewer than three as expected (Fig. 5). During the traffic rush hours, the geometric mean diameters showed significant temporal variations. The evaluation of the algorithm for this kind of aerosol data set is rather challenging because of the high variability of the particle number size distributions originating from traffic activities. However, the total particle number concentrations obtained from the sum of individual modes were very close to those integrated (between 7 and 320 nm) from the measurements (Fig. 5b).

Even though there was no difference between the measured and parameterized total particle number concentration, clear differences between the modal particle number concentrations and respective concentrations in the particle diameter ranges 7–35 nm, 35–120 nm and 120–320 nm were seen. However, it seems that such an approximation is not necessarily correct when compared with that of the mode number concentrations obtained from a multi log-normal fitting. These particle size ranges were chosen for this example to be representatives of the nucleation, Aitken and accumulation mode, respectively. These ranges vary from one situation to another depending on the type of aerosol particles.

From our point of view, the above fact should be taken into account in global climate models and regional/local air quality models. This difference should also be taken into account when correlating particle number concentrations with other variables such as traffic activity. For example, the number concentration of mode-3 and the integrated number concentration within the size range 120–320 nm showed clear differences during the daytime. In fact, this difference may enhance the correlation between the traffic activity and the number concentration of particles in the accumulation mode (Fig. 5d). In principle, this enhanced correlation can be justified by the fact that a significant portion of the Aitken mode lies in the particle diameter range 120–320 nm (Fig. 5e–h), keeping in mind that the Aitken mode is usually highly correlated with traffic activities as it contains a significant portion of the combustion emissions of motor vehicles.

Pollution episodes

Long-range transport and local pollution episodes of aerosol particles dramatically influence the characteristics of aerosol particles at a fixed monitoring station. Local pollution episodes occur within the scale of a few tens of kilometers while long-range transport pollution episodes transport aerosol particles across hundreds to thousands of kilometers. In the following we present two special cases of such pollution episodes that were observed in Helsinki. These examples are not meant to add new information about these phenomena but to show the evaluation of the current algorithm.

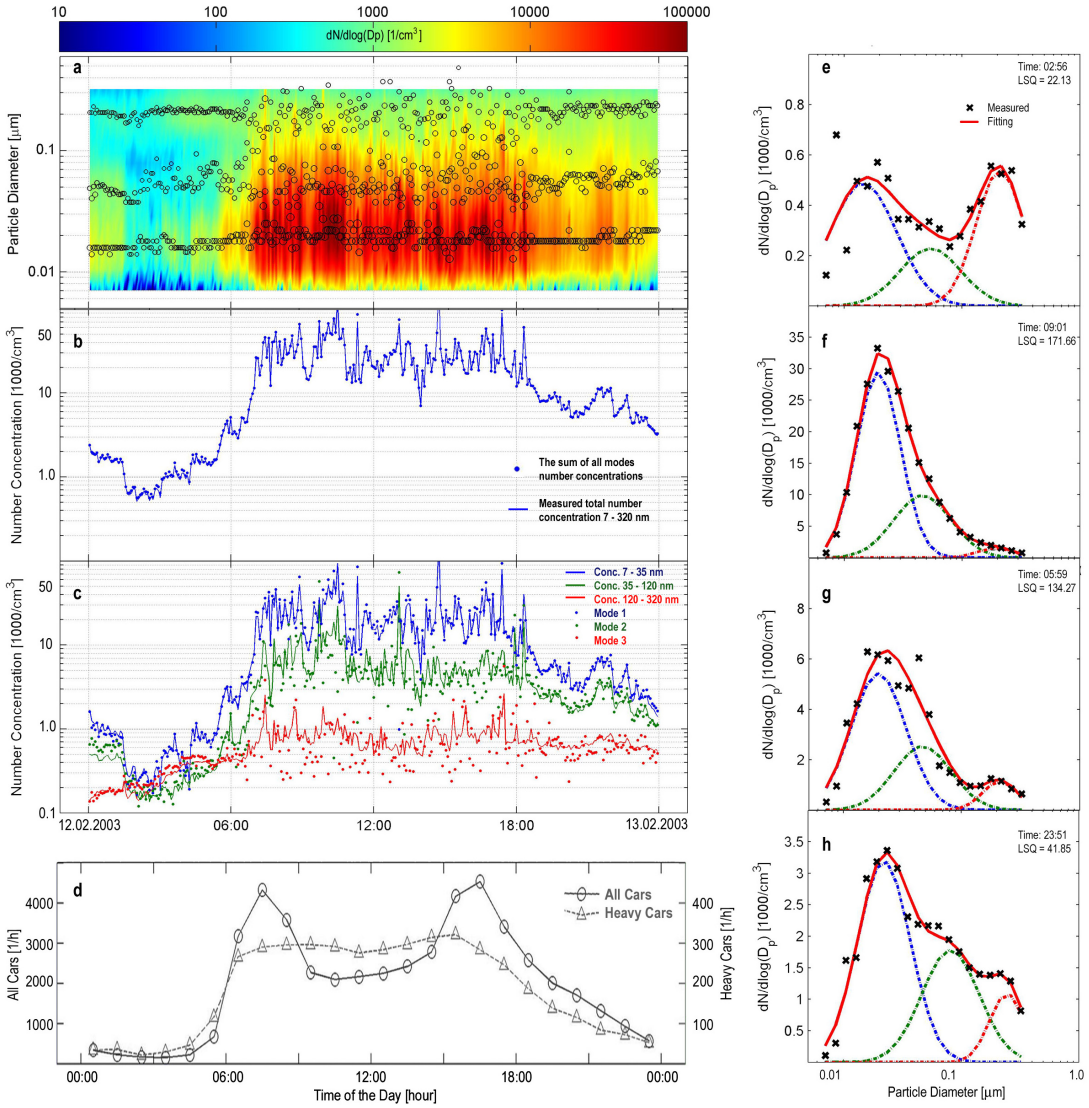

Fig. 5. A case study that represents urban aerosols in Helsinki, Finland. — **a**: Particle number size distribution spectrum during a workday. The circles represent the geometric mean diameters of individual modes and the size of the circle is proportional to the mode number concentration. — **b**: Total particle number concentrations integrated over the measured particle size and also calculated from the sum of the number concentrations of individual modes. — **c**: Measured number concentrations for three size fractions compared with the number concentrations of individual modes. — **d**: Mean traffic volumes during workdays in Helsinki. — **e**–**h**: Selected particle number size distributions and their multi log-normal fitting.

We selected a long-range transport pollution episode that occurred during a weekend when day-time traffic emissions are intermediate (Fig. 6). A long-range transport episode is usually distinguished at an urban site by looking at the ratio between the accumulation mode particle number concentration and that of the Aitken mode. During a strong episode, this ratio is larger than or equal to unity, whereas during normal conditions it is significantly below 0.3 (Figs. 5 and 6).

Fireworks are interesting examples of local pollution episodes. During a new year celebration at Helsinki we observed a significant increase in the particle number concentrations of particles > 50 nm in diameter (Fig. 7). However,

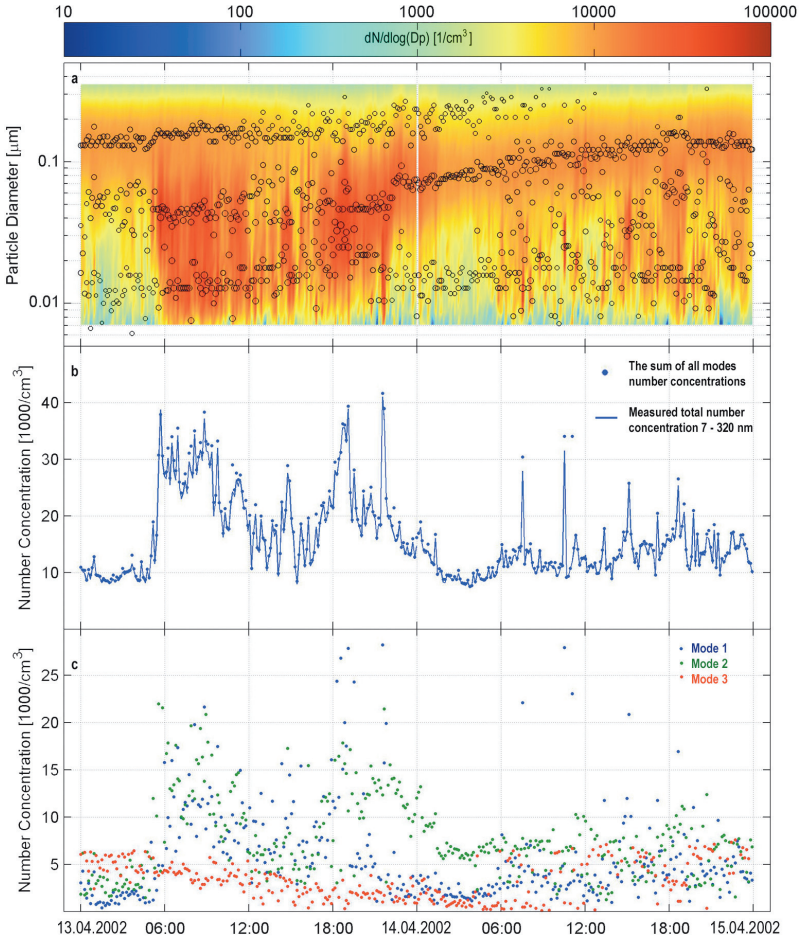

Fig. 6. A case study of long-range transport episode of aerosol particles observed in Helsinki. — **a**: Particle number size distribution spectrum during a workday. The circles represent the geometric mean diameters of individual modes and the size of the circle is proportional to the mode number concentration. — **b**: Total particle number concentrations integrated over the measured particle size and also calculated from the sum of the number concentrations of individual modes. — **c**: Mode number concentrations as predicted by the algorithm.

the geometric mean diameter of the individual modes did not show significant changes during the firework event. The elevated number concentration of mode-3 (accumulation mode) maintained its level for three hours after midnight (the peak of fireworks event). After that, the number concentrations of mode-3 gradually decreased until it maintained its background level at 05:00.

It is also interesting to have a look at the evolution of aerosol particles due to aerosol dynamical processes during the New Year's Day. One possibility is that a new-particle formation event took place around 06:00 when the newly-formed aerosol particles followed a growth pattern and ended up in the Aitken mode regime (Fig. 7a). Another possibility is that a change in meteorological conditions affected the hygroscopic properties of aerosol particles. This second possibility is more probable because the total particle number concentration did not show a significant increase (Fig. 7b–c).

From these two examples, it should be noticed how the modal structure characteristics changed completely during a long-range transport pollution episode. For example, the nucleation and Aitken modes almost disappeared in the afternoon, whereas the accumulation mode dominated the particle number size distribution (Fig. 6). On the other hand, the modal structure was rather unchanged with respect to the geometric mean diameters of individual modes during a local pollution episode in an urban site (Fig. 7).

New-particle formation

Aerosol particles are formed in the atmosphere from gaseous precursors throughout the world

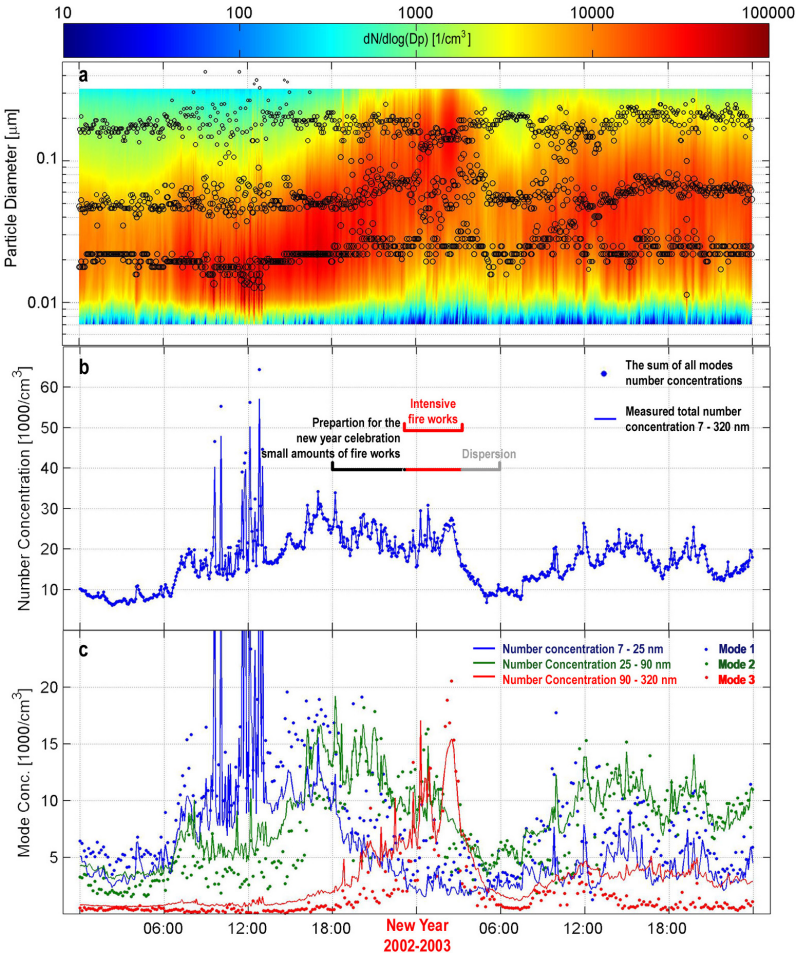

Fig. 7. A case study of local pollution episode due to fireworks during the new year 2002–2003 celebrations in Helsinki. — **a**: Particle number size distribution spectrum during a workday. The circles represent the geometric mean diameters of individual modes and the size of the circle is proportional to the mode number concentration. — **b**: Total particle number concentrations integrated over the measured particle size and also calculated from the sum of the number concentrations of individual modes. — **c**: Measured number concentrations for three size fractions compared with the number concentrations of individual modes.

(Kulmala *et al*. 2004). During a new particle formation event, a new mode of aerosol particles appears with a geometric mean diameter < 10 nm, growing later into the Aitken or accumulation mode regime. The number of newly-formed particles observed in the ambient atmosphere can be as high as 10^6 cm^{-3} (e.g. O'Dowd *et al*. 2002). Both the number concentration and geometric mean diameter of the newly-formed mode are of great interest because the early steps of particle formation and growth hold crucial information about the participating compounds in the formation process as well as about initial conditions. These parameters can be utilized to further understand and estimate the observed new-particle formation rate as well as the growth rate of these newly formed particles.

The subject of new particle formation is beyond the scope of this study. Here we only present examples of such events to show the performance of the current algorithm. The first example (Fig. 8) was observed in the ambient atmosphere at SMEAR II. The second example (Fig. 9) was observed in the EUPHORE measurement chamber in Valencia, Spain (Becker 1996), and it illustrates the formation of aerosol particles due to the oxidation of toluene in the presence of NO_x under sunlight.

In both examples, the current algorithm does well in predicting both the size and number concentration of the newly-formed mode, even during the early steps of the particle formation burst. As can be seen from these two examples, new-particle formation in the ambient atmosphere is more challenging to parameterize than new-particle formation controlled in a chamber.

Because this algorithm showed reasonable results and is automatic, we utilized it in the

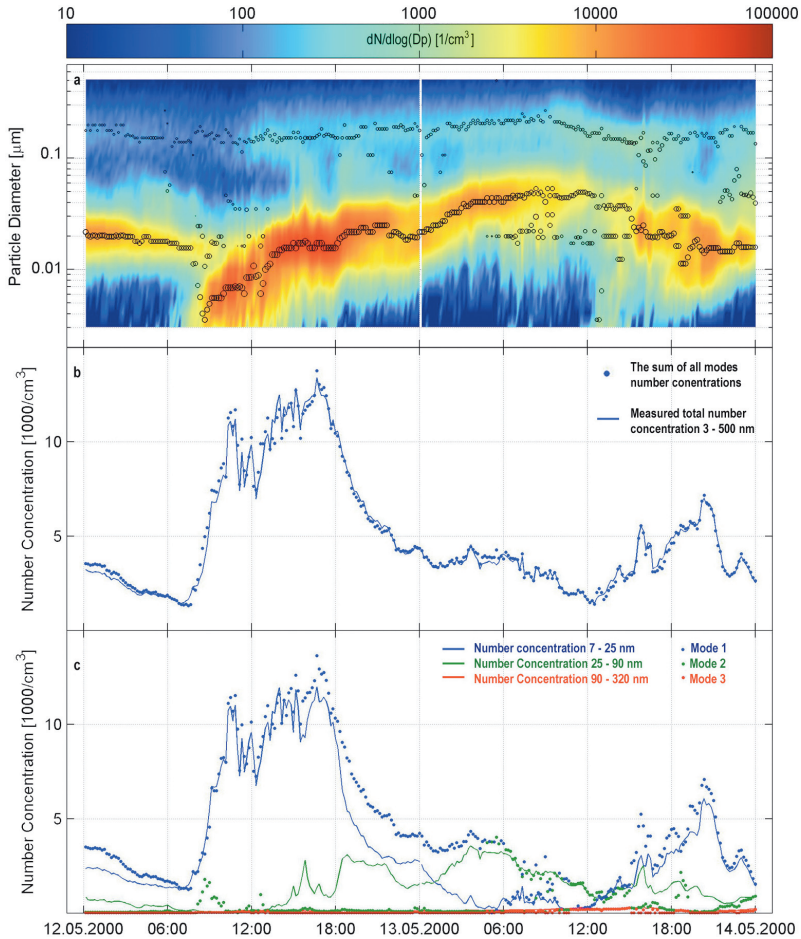

Fig. 8. A case study of new particle formation event in the ambient atmosphere at SMEAR II in the boreal forest in Hyytiälä, Finland. — **a**: Particle number size distribution spectrum during a workday. The circles represent the geometric mean diameters of individual modes and the size of the circle is proportional to the mode number concentration. — **b**: Total particle number concentrations integrated over the measured particle size and also calculated from the sum of the number concentrations of individual modes. — **c**: Measured number concentrations for three size fractions compared with the number concentrations of individual modes.

analysis of new particle formation. The utilization is described in detail in the study by Dal Maso *et al.* (2005).

Summary

In the current study we developed and evaluated an algorithm to parameterize aerosol particle number size distributions with the multi log-normal distribution function. The current algorithm is automatic in the way that it does not need a user control for initial assumptions of the log-normal parameters. The only required input parameter is the maximum number of log-normal modes. We also developed a procedure to test the required number of log-normal modes based on a statistical analysis on the allowed/forbidden separation of two adjacent modes. We utilized this procedure to reduce the number of modes without affecting the fitting quality. The current algorithm is based on the previous algorithms described by Hussein *et al.* (2004, 2005).

The current algorithm was evaluated in comparison with a previous algorithm described by Mäkelä *et al.* (2000) that can be considered a standard procedure. The current algorithm was also evaluated in comparison with a long-term data set and different types of measured aerosol particles in the ambient atmosphere. The evaluation of the current algorithm showed the following advantages:

1. It is suitable for different types of aerosol particles observed in different environments and conditions. This feature generalizes the use of the current algorithm in the future for different data sets of aerosol particles. This

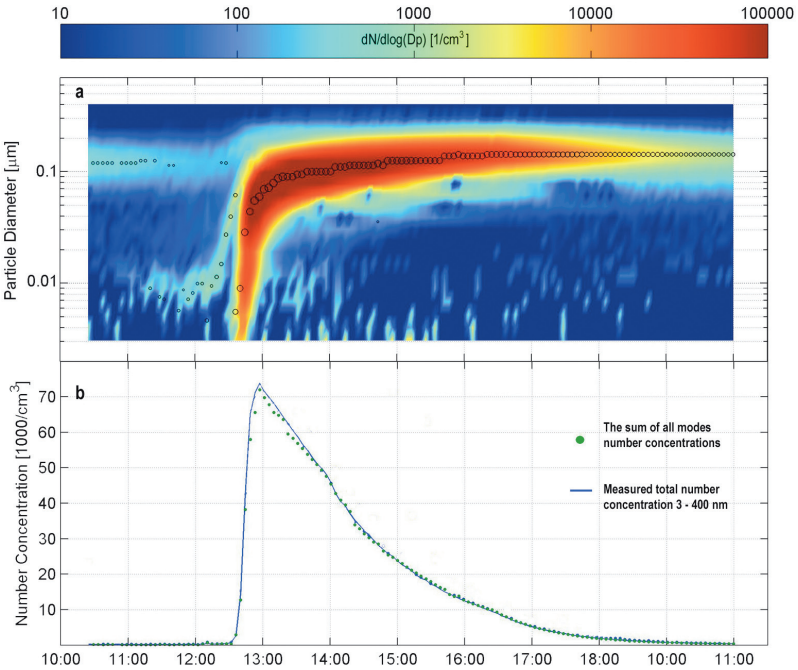

Fig. 9. A case study of new particle formation in the environmental chamber EUPHORE (Valencia, Spain). — **a**: Particle number size distribution spectrum during a workday. The circles represent the geometric mean diameters of individual modes and the size of the circle is proportional to the mode number concentration. — **b**: Total particle number concentrations integrated over the measured particle size and also calculated from the sum of the number concentrations of individual modes.

also validates the inter-comparison between different data sets of aerosol particles by reducing the user influence on the parameterization results because the algorithm is automated.

2. The current algorithm showed agreement with the previous standard algorithm in about 90% of a long-term data set parameterization. The agreement between both algorithms ensures the efficiency of the current algorithm to be used in the future to parameterize aerosol particle number size distributions. Differences between both algorithms are due to the user decision imposed in the current algorithm and those applied in the previous one.
3. Because the current algorithm is automated, it is not time-consuming, particularly when long-term data sets are analyzed. This feature is also very important when comparing the physical characterizations of several long-term aerosol particle data sets.
4. The current algorithm is also a useful tool when investigating atmospheric aerosol particle formation and transformation. For example, it is able to predict both the size and number concentration of a newly-formed aerosol particle mode in the ambient atmosphere as well as in chamber measurements. Parameterization of aerosol particle formation is very challenging due to the high variability of the size characteristics of the new mode at the early stages of the event. This feature of the current algorithm is important in the analysis of aerosol particle formation and growth rate analysis.

Acknowledgements: Financial support from the Academy of Finland is acknowledged.

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Received 25 April 2005, accepted 19 August 2005