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Evolution of Aerosol Particle Number Size Distribution in Statistical Thermodynamic Equilibrium During New Particle Formation and Growth

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Abstract

The aerosol particle number size distribution (PNSD) is pivotal in estimating the corresponding transport, transformation, environmental impacts, and climate effects. This study explores the statistical thermodynamic characteristics of PNSD during new particle formation (NPF) and growth in clean atmospheric environments. Using the maximum entropy principle, we demonstrate that the PNSD follows the Weibull distribution of $n(Dp) = N_0 Dp^{q-1} e^{-\alpha Dp^q}$ (q is the shape parameter). Field observation and theoretical analysis show that q would evolve from above 6 to 3 during different stages of NPF due to the various strengths of condensation, indicating that the aerosol is in the statistical thermodynamic equilibrium state. The findings provide insights into the underlying physical mechanisms governing aerosol behavior and have implications for model simulations of aerosol evolution.

1 Introduction

The particle number size distribution (PNSD) of aerosols plays a pivotal role in atmospheric science and climate research, significantly impacting the optical characteristics of aerosols, their efficacy as cloud condensation nuclei, and the Earth's radiative balance (Che et al., 2024; Gong et al., 2020; Kopanakis et al., 2013; Liu et al., 2014; Schmale et al., 2018; Chen et al., 2023). Aerosol particles, ranging from nanometers to micrometers, exhibit significant heterogeneity, with their PNSD reflecting the different characteristics of atmospheric chemical processes, and physical dynamics (Kopanakis et al., 2013; Tie, 2015; Izhovkina et al., 2018; Zhang et al., 2020). A comprehensive understanding of the PNSD is essential for unraveling its environmental and climatic impacts.

The evolution of PNSD is governed by different physico-chemical processes such as coagulation, sedimentation, diffusion, convection, growth, and deposition (Elgarayhi, 2002). The ratio of aerosol number concentration at different times at a specific size follows the equation:

$$\frac{\partial n(v,t)}{\partial t} = -n(v,t)\int_0^\infty K(v,u)n(u,t)du + \frac{1}{2}\int_0^v K(u,v-u)n(u,t)n(v-u,t)du + \int_0^\infty L(u)n(u,t)du + SC(v,t) - SK(v,t), \quad (1)$$

Where $n(v,t)$ represents the aerosol number concentrations with particle size v; $K(v,u)$ is the coagulation ratio of aerosols at particle size v and u; $L(u)$ is condensation ratio of aerosols increase from u to v; $SC(v,t)$ represents the source emission of aerosol particles, and $SK(v,t)$ represents the sink term for aerosol particles (Riemer et al., 2010; Zaveri et al., 2010). Although equation 1 can capture the temporal variations in aerosol concentration accurately, it fails to provide a comprehensive picture of the PNSD.

Various statistical distributions have been employed to characterize PNSD. The lognormal distribution, defined by its geometric mean diameter and geometric standard deviation, is widely used due to its ability to describe aerosol populations with a single mode and a relatively narrow size range from specific sources (Unfer et al., 2024; Schuster et al., 2006). The Gamma

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distribution, with its additional shape parameter, offers more flexibility and can capture broader size distributions, including those with multiple modes, making it valuable for complex atmospheric conditions(Nyaku et al., 2020; Unfer et al., 2024). The exponential distribution is also used to describe aerosol populations with a constant removal rate, such as those influenced by dry deposition processes (Schuster et al., 2006). The multi-lognormal distribution extends the lognormal model to account for multiple modes, allowing for a more comprehensive representation of aerosol populations with distinct size fractions, such as those influenced by both natural and anthropogenic sources (Ma et al., 2011; Chen et al., 2014; Shang et al., 2018). The Weibull distribution, with the equation:

$$n(Dp) = N_0 Dp^{q-1} e^{-\alpha Dp^q}, \qquad (2)$$

characterized by its shape parameter (q) and scale parameters (α), is particularly useful for modeling size distributions with a skewed shape (Fisenko et al., 2021; Robin et al., 2013; Zhou et al., 2021). The Weibull distribution has garnered significant attention in aerosol and cloud research due to its flexibility and ability to describe a wide range of size distributions (Liu et al., 1995; Liu and Liu, 1994). Each of the above distributions provides unique insights into aerosol behaviors in the atmosphere. Many of the above statistical distributions are based on the measured PNSD shape, and then the corresponding statistical distributions were adopted without truly considering the underlying physical process. However, there is no satisfying theoretical explanation as to why certain distribution forms preferentially occur instead of others.

Previous studies show that the cloud particle size distribution would follow the Weibull distribution using the principle of maximum entropy when the cloud particles were in statistical thermodynamic equilibrium(Wu and Mcfarquhar, 2018; Liu et al., 1995). Similarly, the aerosol PNSD may also follow a Weibull distribution if aerosols are in statistical thermodynamic equilibrium. However, a critical challenge remains in determining whether the PNSD is in statistical thermodynamic equilibrium in the real atmosphere.

In this study, we would demonstrate that the PNSD in the clean atmosphere during new particle growth procession would follow the Weibull distribution. The consistency between the theoretical analysis and field measurement implies that the aerosol is in statistical thermodynamic equilibrium in the clean atmosphere. The findings provide insights into the underlying physical mechanisms governing aerosol behavior and have implications for improving model simulations. Understanding these elements helps in modeling and predicting the behavior of aerosol particles in a clean atmosphere, shedding light on their impacts on air quality and climate.

2 Theoretical analysis of aerosol PNSD in the clean atmosphere

In the clean atmosphere, the coagulation of aerosols is weak, and thus the impact of coagulation on aerosol PNSD can be neglected. The total aerosol number concentrations should be constant with:

$$N = \sum_{i=1}^{N} N_i = \int_0^\infty n(Dp) \bullet dDp \quad , \qquad (3)$$

Where N_i is the number of aerosols at a small specific diameter range corresponding to the aerosol number size distribution $n(Dp)$. The number of microscopic configurations where the N aerosols can be located over the different diameter is given by

$$W = \frac{N!}{N_1! N_2! \cdots N_n!}, \qquad (4)$$

When the aerosol is in statistical thermodynamic equilibrium, the Shannon's information entropy, which can be defined as:

$$\mathrm{S} = k_B \ln(W), \qquad (5)$$

should reach its maximum number. Another constraint condition is necessary when solving the above equation. We would discuss the corresponding solution when (1) the mass of the aerosol is constant when the condensation is weak, and (2) the inertial moment of aerosols is invariant when the effect of condensation is strong.

2.1 PNSD in a clean atmosphere when the condensation effect is ignorable

When the condensation effect is not strong, the total mass concentration of aerosols is constant and can be expressed as:

$$m = \sum_{i=1}^{N} m_i = \sum_{i=1}^{N} N_i \bullet \frac{1}{6} \bullet \pi \bullet \rho \bullet Dp_i^3 = \int_0^\infty n(Dp) \bullet \frac{1}{6} \bullet \pi \bullet \rho \bullet Dp^3 \bullet dDp, \qquad (6)$$

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Where ρ is the effective density of aerosol. With equations (1), (2), (3), and (4), the maximum number of W can be derived using the Lagrange multiplier with:

$$d\ln(W) - \lambda_0\left(\sum_{i=1}^{n} N_i - N\right) - \lambda_1\left(\sum_{i=1}^{n} N_i m_i - m\right) = 0, \quad (7)$$

Where λ_0 and λ_1 are the Lagrange multipliers. The corresponding solution of equation 3-7 should be:

$$n(Dp) = N_0 Dp^2 e^{-\alpha Dp^3}. \quad (8)$$

Details of deriving equation 8 can be found in the supplementary material.

Based on the above results, the PNSD should follow the Weibull distribution in a clean atmosphere when the condensation effect is not strong, and the corresponding shape factor q is 3.

2.2 PNSD in a clean atmosphere when the condensation effect is not ignorable

When the condensation effect is strong, the total mass concentration of aerosols is not constant. However, the moment of inertial should be invariant when the gas vapor condenses onto the aerosol surface uniformly with:

$$I = \sum_{i=1}^{N} I_i = \sum_{i=1}^{N} M_i R_i^2 = \sum_{i=1}^{N} N_i \bullet (\frac{1}{6} \bullet \pi \bullet \rho \bullet Dp_i^3) \bullet (\frac{1}{4} \bullet Dp_i^2) = \int_0^{\infty} n(Dp) \bullet \frac{1}{24} \bullet \pi \bullet \rho \bullet Dp^5 \bullet dDp, \quad (9)$$

With equations (3), (4), (5), and (9), the maximum number of W can be derived using the Lagrange multiplier with:

$$d\ln(W) - \lambda_0\left(\sum_{i=1}^{n} N_i - N\right) - \lambda_1\left(\sum_{i=1}^{n} N_i I_i - I\right) = 0, \quad (10)$$

The corresponding solution should be:

$$n(Dp) = N_0 Dp^4 e^{-\alpha Dp^5}. \quad (11)$$

Details of deriving equation 11 can be found in the supplementary material.

Based on the above result, the PNSD should follow the Weibull distribution in a clean atmosphere when the condensation effect is strong, and the corresponding shape factor q should be equal to 5.

2.3 PNSD distribution under different stages during new particle formation and growth in a clean atmosphere

Based on sections 2.1 and 2.2, the PNSD in a clean atmosphere follows the Weibull distribution, with the shape parameter q varying across different stages of new particle formation and growth:

(1) The first time stage is NPF. During this stage, the particle number concentrations would increase rapidly. As the number concentration varies, the PNSD fails to meet equation 6, and thus the aerosol PNSD may not follow the Weibull distribution.

(2) The second time stage is shortly after the NPF, and the aerosol particle number concentrations stabilize. However, the condensation effects should be strong due to the high gas precursor concentrations. The aerosol during this state corresponds to rapid aerosol diameter growth. If the aerosol particle is in a statistical thermodynamic equilibrium state, the aerosol PNSD should follow the Weibull distribution, and the corresponding shape factor q should be 5.

(3) The third stage corresponds to those times when the condensation effects are less strong than those in stage 2. However, the condensation effects are still not ignorable as the gas precursors remain at a relatively high concentration.

(4) The fourth stage corresponds to those times when the condensation effects are ignorable. At the same time, the effects of background aerosol on aerosol PNSD are ignorable as the NPF generates a large amount of aerosols. During this state, the aerosol PSND should also follow the Weibull distribution, and the corresponding shape factor q should be 3.

(5) The last stage is that the aerosol mixes with the background aerosols, and the processing of emission, transformation, and coagulation on aerosol PNSD is not ignorable. The PNSD during these stages would not follow the Weibull distribution. In summary, if aerosols are in statistical thermodynamic equilibrium, the shape parameter q decreases from around 5 to 3 as aerosols transition from stage 2 to stage 4. Additionally, q may exceed 5 at the initial stages of new particle formation.

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3 Field measurement of the PNSD in a clean atmosphere

A field measurement was conducted at the University of Tibet, China (29°38′34.65″ N，91°10′56.22″ E; 3650 m a.s.l.) to study the PNSD at a clean atmosphere between 10 October and 13 November, 2024. The measurement site is located on a campus without direct emissions that influence the aerosol PNSD, thus the site can be used as a representative site for the Plateau urban atmospheric environment. During the field measurement, a nanoparticle scanning mobility particle sizer (Nano-SMPS, Model 3087, TSI Inc., USA) and a scanning mobility particle sizer (SMPS, Model 3080, TSI Inc., USA) were used to measure the PNSD over the size range of 3~40 nm, and 20~800 nm, respectively with a time resolution of 5 minutes. During the field measurement, the instruments were placed in a container on the roof of the teaching building, which is about 20 meters above the ground. The temperature was adjusted to be around 25 ºC in the container. The aerosol median diameter, condensation sink (CS), coagulation ratio (Coag), number concentration (N), volume concentration (V), and corresponding shape factor q were calculated and further analyzed. Details of calculating the above parameters can be found in the supplementary materials.

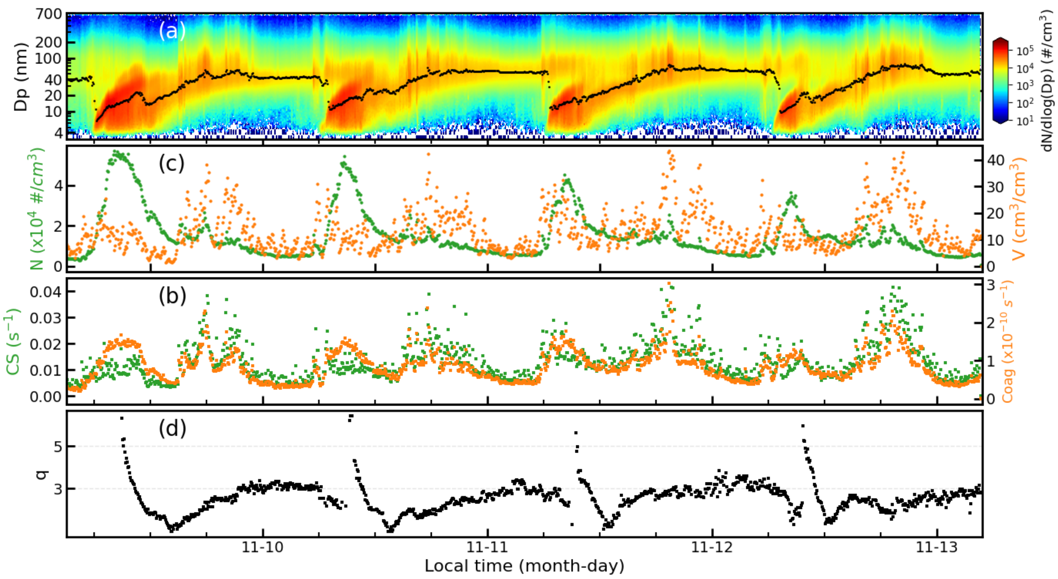

Figure 1. Measured time series of (a) aerosol PNSD, (b) number concentration (green dots), volume concentration (orange dots), (c) condensation sink (green dots), coagulation ratio (orange dots), and (d) the shape factor. The black dots at panel (a) show the corresponding median diameter of PNSD.

Fig. 1 gives the measured and calculated results of aerosol parameters between 9 November and 13 November, as NPF happens during these days. Thus, we only provide the results of these days to elucidate the main conclusions. From Fig. 1, the aerosol PNSD was mainly influenced by the NPF. The median diameter varies between 6.56 and 76.57 nm. The median diameter would drop suddenly from around 40 nm to around 7 nm due to the large amount of small newly formed aerosol particles. The measured mean aerosol number concentrations (N) and volume concentrations are $1.52\pm1.15\text{x}10^4$#/cm^3 and 13.23 ± 7.76cm^3/cm^3. The corresponding mass concentration is around 19.8 $\mu g/cm^3$ for these aerosols with a diameter lower than 1 μm, indicating that the surroundings are very clean with lower aerosol mass. N would increase significantly by more than five times during the NPF period. Due to its small size and mass, the corresponding mass concentrations didn't experience a remarkable change. During these days, the mean CS is 0.013 ± 0.008s^{-1}, which is larger than those of the Plateau background sites in China with a mean value of 0.0014 (Tang et al., 2023), but smaller than those of the Chinese main city environment in Beijing (Du et al., 2017). The corresponding coagulation ratio is also very small, with a mean value of $8.5\pm4.2\text{x}10^{-11}$s^{-1}.

The PNSD was also fit using the Weibull distribution (examples of fitting using the Weibull distribution are shown in Fig.s S1), and the corresponding shape factor q values are shown fig. 1(d). From Fig. 1(d), q values follow the same variation of diurnal cycles during these days. At first, the q would be around 6 at the initial of new particle formation, this period corresponding to the time stage 1 of 2.3, and the particle number concentration increases significantly. As the aerosol PNSD is not a statistical thermodynamic equilibrium state, there is no brief explanation of why the q is larger than 5. After the N reaches its maximum, the q would decrease to around 5, indicating that the aerosol PNSD was mainly influenced by the condensation sink, which corresponds to stage 2. With the weakening of the condensation sink, the q values continue to decrease to lower than 3, which indicates that those aerosols are mixed up with the ambient aerosols. However, the q values finally reach around 3 as the aerosol would finally be in a statistical thermodynamic equilibrium state, which corresponds to stage 4. However, the shape factor would vary around 3 as the aerosol from other sources may mix with the existing aerosols from NPF. From the above analysis, we can conclude that the measurement results of the variation of q values from field measurement agree well with our theoretical analysis, which, to some extent, validates that the aerosol PNSD would follow the Weibull distribution when the aerosol is in a statistical thermodynamic equilibrium state.

4 Conclusions

Our study reveals that the aerosol PNSD in the clean atmosphere follows the Weibull distribution during the new particle formation and growth process. The shape parameter q of the Weibull distribution decreases from around 6.0 to 3.0 as the new particle formation and growth progress, suggesting that the aerosol tends to reach a statistical thermodynamic equilibrium state. This finding is supported by both theoretical analyses based on the maximum entropy principle and field measurements conducted at a clean atmospheric site in Tibet, China.

The theoretical analysis demonstrated that the PNSD should follow the Weibull distribution in a clean atmosphere, with the shape factor q varying under different stages of new particle formation and growth. The field measurements showed that the q values followed diurnal cycles, starting from around 6 at the initial stage of new particle formation, decreasing to around 5 shortly after the formation, and finally stabilizing around 3 as the aerosol reached a statistical thermodynamic equilibrium state.

These results provide insights into the underlying physical mechanisms governing aerosol behavior in the clean atmosphere and have implications for improving model simulations of aerosol evolution. The findings are important for tracking pollution sources, predicting climate impacts, and enhancing air quality models. Furthermore, the study suggests that the Weibull distribution could potentially simplify the parameterization scheme of aerosol number size distribution in model simulations.

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Author contributions. ZG wrote the manuscript. ZG, TP, YC, and ZT discussed the original ideal and results. LW, GY, SJ, CY, SF conducted the field measurement.

Open Research. The data is available via (Zhao, 2025).
Software. The data is dealt with using Python 3.8.5, and the picture is plotted using Python 3.8.5.
Competing interests. The authors declare that they have no conflict of interest.

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