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# Theoretical analysis of a method to measure size distributions of solid particles in water by aerosolization

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A B S T R A C T

Reliable measurement of the size-resolved number concentration (size distribution) of solid particles dispersed in water or melted ice is of critical importance in many geoscientific observational studies. Because physical and chemical properties of particles can be measured more unambiguously and accurately in rarefied media (air) than in condensed media (water), *particle measurement after aerosolization using a nebulizer* is a significant method for the observation of solid particles dispersed in water. We propose a mathematical theory for estimating the original size distribution of solid particles in water from the measured size distribution of aerosolized particles. We assume that the size distribution of water droplets produced by a nebulizer is given. The size distribution of solid particles in water can be estimated by solving a system of nonlinear equations. The complexity in solving the equations increases rapidly with the computational resolution of particle size and the assumed maximum number of particles within each droplet. For such a system of equations, we found rigorous error bounds of a true solution using INTLAB, an interval arithmetic package. Our theoretical framework will be useful in many fields in geoscience as a fundamental scheme to quantify solid particles in water. In particular, an application of the proposed theoretical method is shown to be useful for the quantitative observations of the size distribution of black carbon particles in rainwater.



## 1. Introduction

Solid particles dispersed in water, snow, and ice on the earth's surface have many possible environmental and climatic impacts. Marine colloidal particles alter the transfer of solar radiation in seawater through scattering and absorption (Stramski et al., 2004) and may serve as nuclei for larger particles that control the gravitational sedimentation of geochemically important species (Wells & Goldberg, 1992). In groundwater, colloidal particles with size less than several micrometers can be a dominant agent for the transport of hazardous radionuclides and hydrophobic organic molecules across significant horizontal distances underground (McCarthy & Zachara, 1989). Light-absorbing particles (e.g., black carbon (BC)) deposited on snow reduce surface albedo and induce melting (Flanner et al., 2007). These climatic and environmental effects of small particles in water or ice strongly depend on their size, shape, chemical composition, and mixing state. Rainwater and snow contain aerosol particles incorporated through the nucleation of cloud and ice particles and coalescence with precipitating particles. Because the particle size of water-insoluble components does not change even

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**Nomenclature**

| | |
|---|---|
| $d$ | diameter of particle, m |
| $D$ | diameter of droplet, m |
| $l$ | number of size bins for particle |
| $L$ | number of size bins for droplet |
| $n$ | number concentration of particles in water, $m^{-3}$ |
| $\mathbf{n}$ | vector notation for size-resolved number concentration of particles in water, $m^{-3}$ |
| $n'$ | number concentration of particles in air, $m^{-3}$ |
| $\mathbf{n}'$ | vector notation for size-resolved number concentration of particles in air, $m^{-3}$ |
| $N$ | number concentration of droplet in air, $m^{-3}$ |
| $\mathbf{N}$ | vector notation for size-resolved number concentration of droplet in air, $m^{-3}$ |
| $p_{ij}(x_i)$ | probability distribution for number of $i$-th particles $x_i$ in a $j$-th droplet |
| $q_{jk}$ | probability of production of $k$-th particle from $j$-th droplet |
| $v$ | volume of a particle, $m^3$ |
| $V$ | volume of a droplet, $m^3$ |
| $x$ | number of particles in a droplet |
| $X_k$ | a set in $l$-dimensional space of non-negative integers defined by Eq. (4) |
| $\lambda_{ij}$ | mean number of $i$-th particles in a $j$-th droplet |

after incorporation into liquid water, comparison between the size distributions of such particles in air and precipitating water is the basis for discussing the possible size-dependent efficiency of the wet removal of atmospheric aerosols.

The combination of laser-diffraction tomography with fluorescence spectroscopy has been a major scheme for observing the size distribution and chemical characterization of solid particles in water (Kerker, 1983; Miroslaw & Georges, 2011). However, interpreting the microphysical properties of particles solely from these optical signals could sometimes yield ambiguous results, because the angular intensities of scattered light and fluorescence emission depend on the size, shape, chemical composition, and mixing state of individual particles (Wang et al., 1980).

Recently, quantitative measurements of (black carbon) BC particles dispersed in water have become possible using a nebulizer and laser-induced incandescence (LII) technique (Ohata et al., 2011; Schwarz et al., 2013). To date, the LII technique, which requires aerosolization (i.e., extraction of the colloidal particles into air as aerosols) of particles dispersed in water before detection, is the only method available to measure the BC mass present in individual internally mixed multicomponent particles. In general, measurement techniques and their signal interpretations are simpler for particles suspended in rarefied media (vacuum or gas) than those dispersed in condensed media (liquid or solid). Therefore, for a number of geophysical, geochemical, and microbiological research applications, *particle measurements after aerosolization* is a fundamental scheme for the quantitative observations of solid particles in water.

As illustrated in Fig. 1 (see also Fig. 10, Ohata et al., 2011), the aerosolization of solid particles in water using a nebulizer changes their size and number concentration if multiple particles are included within a droplet. The particles are agglomerated to a single larger particle after the evaporation of the droplet. Agglomerations enlarge size and reduce the number of particles observed in air compared to the original particles in water. The occurrence of agglomeration becomes negligible under the limit of dilute particle concentration, wherein individual droplets contain at most one particle.

In this paper, we propose a mathematical theory to determine the original size distribution of solid particles in water by particle measurements after aerosolization using a nebulizer, assuming availability of experimental data of the size distribution of nebulized water droplets. This theoretical method is universally applicable to any observation of solid particles in water using a nebulizer.

## 2. Mathematical theory

A schematic of the proposed theoretical method is shown in Fig. 2. Derivations of important formulae are given in this section. In our formulations, the size of particles and droplets is discretized into $l$ and $L$ bins, respectively. We denote a particle with $i$-th size bin simply as an $i$-th particle and a nebulized water droplet with $j$-th size bin as a $j$-th droplet.

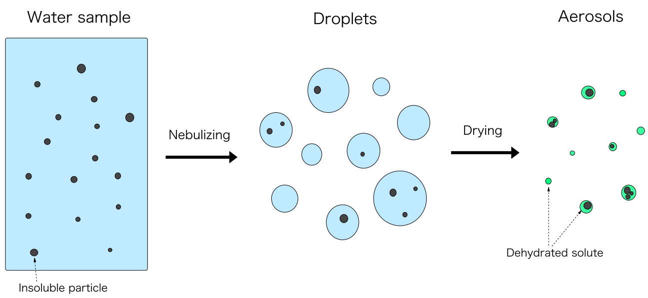


**Fig. 1.** Aerosolization of solid particles dispersed in water using a nebulizer.