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## 学 位 論 文 概 要 Dissertation Summary

学位請求論文(Dissertation) 題名(Title) 原子間力顕微鏡を用いた個別粒子の物性評価法の開発と実大気エアロゾルへの応用 Direct Measurement of Physical Properties of Individual Particles by Atomic Force Microscopy and its Application to Atmospheric Aerosols

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学位論文概要(Dissertation Summary)

Atmospheric aerosols emitted from various sources vary in chemical composition, size, and physical properties. Their mixing states is also not uniform, with individual aerosol particles having a variety of chemical compositions. Therefore, understanding the chemical composition and physical properties of diverse aerosol particles requires detailed measurements and analysis on an individual particle basis. The physical properties and phase state of aerosol particles are important for (i) global climate, by modifying cloud condensation nucleation activity and optical properties of aerosol particles, and (ii) public health, by facilitating their inhalation into the human body and inducing toxicity. This thesis utilized atomic force microscopy (AFM) to evaluate the physical properties of individual aerosol particles in complex mixing states, with particular emphasis on the the adhesion force, Young's modulus, and viscosity related to the aerosol phase state. This study demonstrated that AFM can be used as an effective tool to quantitatively characterize the physical properties of aerosol particles on an individual basis.

Two methods of force-distance curve mapping were tested for organic (glucose) and inorganic salt (NaCl, MgSO4, artificial sea salt, and ammonium sulfate) particles. The results showed that the difference between the two methods has less influence on the measurement than the variations caused by the probes used, and that the results measured by both methods are reasonably comparable. In addition, two types of AFM instruments and AFM probes were compared by measuring same standard particles. No significant difference was found between instruments. However, there are variations in AFM probes that significantly impact the measurement results. It was found that the adhesion force measured on the particle and on Si wafer substrate scales linearly (Figure 1). Therefore, it is recommended to measure the adhesion force of a standard sample, such as Si wafer, prior to conducting measurements in order to obtain a more precise evaluation of aerosol particle adhesion force.

Highly corrosive sea spray aerosols (SSA) can be deposited on surfaces and cause damage to steel- or concrete-based infrastructure, especially in coastal areas. However, the extent to which the adhesive behavior of SSA is affected by different degrees of organic mixing has not been sufficiently studied. The goal of this study was to evaluate the adhesion forces of both laboratory-generated and ambient SSA with a variety of organic contents on an individual particle basis using atomic force microscopy. Aerosols were also generated using a bulk sea foam sample collected from the coast of Noto Peninsula, Japan to simulate SSA enriched in a complex mixture of marine organics. The adhesion force of the sea foam particles was compared with that of two types of standard monosaccharides (glucose and fucose), artificial inorganic sea salt (ASS), glucose and ASS mixtures at different mass ratios, and ambient sea salt (SS) particles. Pure monosaccharide or its mixtures showed substantially larger adhesion forces than predominantly inorganic particles such as ASS and SS (Figure 2). In addition, it was found that a fraction of sea foam particles showed similarly large adhesion forces, suggesting that organic (e.g., monosaccharides)-enriched SSA can be considerably more adhesive than inorganic salts. Furthermore, a few but surprisingly adhesive particles were also identified among ambient aerosols. Therefore, it is necessary to consider the abundance of organics such as saccharides to correctly evaluate the adhesivity of SSA.

Aerosols exist in various states in the atmosphere, including solid, liquid, semi-solid, and glass. The phase state of aerosols has an impact on global climate and various atmospheric processes. It is known to be influenced by the surrounding environment, such as relative humidity. The viscoelasticity of aerosols, which includes viscosity and elasticity, is closely related to the phase state of the aerosol. Therefore, aerosol viscosity and elasticity measurements are important for understanding impacts on global climate and various atmospheric processes. The study measured the viscoelasticity of aerosol particles using atomic force microscopy with the KV and JKR models. The data indicate that viscosity decreases as tip velocity and indentation rate increase at all humidity levels (Figure 3). To enhance comprehension of the physical properties of aerosol particles, it is recommended to conduct AFM-

based measurements under various velocity conditions that are consistent with the velocity scale of atmospheric aerosol phenomena.

These results suggest that AFM measurements of the physical properties of individual aerosols can lead to a better understanding of the phase state of atmospheric aerosols and their associated properties. The particle adhesion properties determined are expected to provide insight into aerosol adhesion phenomena occurring on the interface of the atmosphere, including infrastructure corrosion and particle re-suspension. Detailed measurements of aerosol viscoelasticity, including Young's modulus and viscosity will provide a better understanding of various atmospheric processes, including new particle formation, particle growth, cloud condensation and ice nucleation activities, and aging of aerosols. Such knowledge will eventually be incorporated into models to better predict the impacts aerosols have on the climate change and public health.



Figure 1 The plot of adhesion forces of Si wafer and aerosol particles. Green and black dashed lines show fitting lines of glucose and inorganic particles, respectively.



Figure 2 Comparison of adhesion force of ASS, ASS/Glu (4:1, 1:1, 4:1), two monosaccharides, SS and the two types of sea foam particles measured in this study. Hollow marks indicate the adhesion force values of individual particles, whereas solid marks and whiskers indicate the mean and standard deviation, respectively.



Figure 3 Log  $(\eta)$  versus indentation rate plot of glucose particles at each RH conditions.