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# 主　論　文　の　要　旨

論文題目　Physical and chemical characteristics of submicron atmospheric aerosol particles observed at a forest site in Wakayama, Japan
（和歌山森林域におけるサブミクロン大気エアロゾル粒子の物理・化学的特徴）

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## 論　文　内　容　の　要　旨

Submicron atmospheric aerosols are of great importance to Earth's climate and weather conditions. They also adversely affect the environment and human health. Terrestrial forest vegetation emits a large amount of biogenic volatile organic compounds (BVOCs) into the atmosphere. Biogenic secondary organic aerosol (BSOA), which is formed from the oxidation of BVOCs, would contribute to secondary formation and growth of aerosol particles, and thus enhance both the number and mass concentrations of atmospheric aerosols. New particle formation (NPF) and growth have been observed in various environments, yet the characteristics of NPF are rarely investigated at the mid-latitude forests. Despite the large contribution of BSOA to organic aerosols on the global scale, their formation pathways, composition, and evolution processes are not well understood. Characterization of BSOA remains an important research topic, especially in the areas where it is rarely studied. BSOA has been detected using traditional off-line techniques at some forest sites in East Asia, but study of BSOA based on real-time measurements in East Asian forest areas is far from sufficient.

This study aims to investigate the physical and chemical characteristics of aerosol particles in the forest area of East Asia, including formation and growth of new particles, influences of air mass and meteorology on NPF, magnitude of the observed NPF, chemical composition of submicron aerosol particles, and formation and evolution of BSOA. In this study, an intensive field measurement of aerosol particles was performed at a forest site in Wakayama, Japan

during 20–30 August 2010. The number size distributions of aerosol particles at 14–710 nm in mobility diameters were measured with a scanning mobility particle sizer (SMPS) system. The chemical composition of non-refractory submicron particulate matters (NR-$PM_1$) was measured with an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). TSP (total suspended particulates) and $PM_{0.95}$ (particle with an aerodynamic diameter $\leq$ 0.95 μm) aerosol samples were collected for chemical analysis of ionic species, organic carbon (OC), and elemental carbon (EC).

Four days (26–30 August 2010) during the observation period experienced NPF, which was characterized by the strong bursts of ultrafine particles and their subsequent growth. The NPF events occurred preferentially under the condition of less pre-existing particles with low vapor condensation sink (CS) and particle coagulation sink. The formation rate of new particles was calculated to be in the range between 0.2 and 1.0 $cm^{-3}$ $s^{-1}$ for the NPF events. The growth rates of the newly formed mode of aerosol particles ranged between 5.0 and 15.7 nm $h^{-1}$. The backward air mass trajectories revealed that the NPF occurred when clean maritime air masses originated from the North Pacific. Relative humidity (RH) was slightly lower during particle nucleation period on NPF event days than that on non-event days, implying its connection to the occurrence of NPF; whereas the effect of RH on particle nucleation was not as obvious as that of air mass origin. The results from the classification analysis of backward air mass trajectories indicate that the maritime air mass conditions are frequent at this forest site and are mainly in summer season. Large increases in the number concentrations of accumulation mode particles (above 90 nm) were followed by the increased precipitation rates in the afternoon hours on NPF event days. Therefore, the newly-formed particles would be involved in the convective cloud formation and precipitation over the studied region.

Total mass concentrations of chemical components in TSP and $PM_{0.95}$ on NPF event days (4.6 and 1.7 μg $m^{-3}$, respectively) were significantly lower than those on non-event days (18.9 and 8.6 μg $m^{-3}$, respectively), indicating that NPF events occurred under relatively low aerosol loadings in this forest atmosphere. Results from an aerosol transport model suggest that aerosols were under the strong influence of anthropogenic pollutants on non-event days, whereas such influence was much smaller on NPF event days. A major fraction of non-

refractory submicron aerosol particles consisted of organics (accounting for, on average, 46% of total mass), sulfate (41%), and ammonium (12%). Positive matrix factorization (PMF) of high-resolution organic aerosol mass spectra identified two oxygenated organic aerosol (OOA) components, i.e., a highly oxidized, low volatility LV-OOA and a less oxidized, semivolatile SV-OOA, which are interpreted mainly as regional continental organic aerosol (OA) and locally-formed biogenic secondary OA (BSOA), respectively. The mass concentrations of SV-OOA increased prominently during the daytime, suggesting a strong photochemical production of BSOA on both non-event and new particle formation (NPF) event days. Increases of $f_{44}$ (fraction of m/z 44 in OA mass spectrum), the fraction of $C_xO_y^+$ fragment, and the O/C after midday (around 1300 LT) suggest that OA became increasingly oxygenated, which can be explained by the aging processing of freshly-formed BSOA. Aqueous oxidation reactions under high relative humidity conditions might be the major pathway for BSOA aging in this forest atmosphere. A substantial increase of the mass concentration of organics in the small size range (below 300 nm in vacuum aerodynamic diameter), without the increase of that of sulfate, suggests that the formation of BSOA made a dominant contribution to the particles in cloud condensation nuclei sizes around the studied area. Further investigation of aerosol particles over forest areas, at which inflow of clean maritime air masses is expected (like the studied forest), is valuable to understand the formation and evolution processes of BSOA as well as its magnitude to the production of CCN and cloud droplets.