Bessel Beam Optical Trap

Organic aerosols play a major role in the chemistry and physics of the atmosphere, serving as cloud condensation or ice nuclei, sites of interfacial chemistry, and scatterers and absorbers of incident solar radiation. Recent field campaigns have affirmed their prevalence in the troposphere, while aircraft measurements have discovered large quantities of organic material within the nuclei of ice crystals in mid-latitude, mixed-phase clouds.[1] Despite the ubiquity and importance of organic aerosols in the environment, scientists are only beginning to understand aerosol phase behavior and the influence of aerosol phase on various atmospheric processes, such as gas-aerosol partitioning, chemical reactions, and water uptake. Therefore, we proposed a new instrument that would give us insight into these processes at the single particle level.

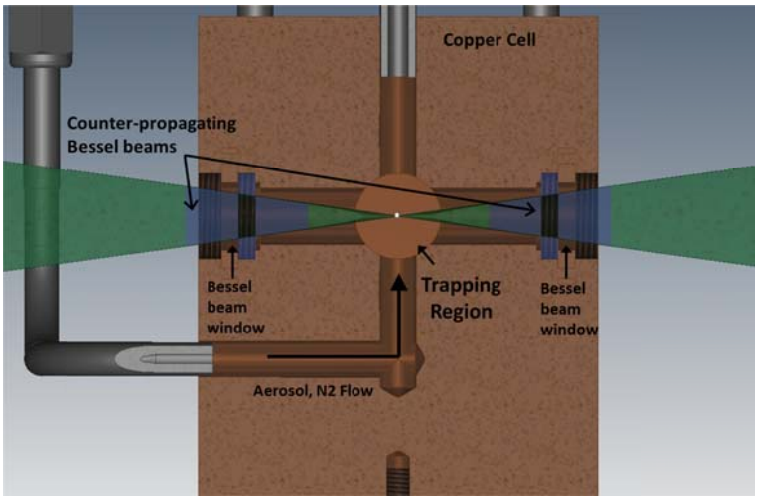

Fig. 1. A schematic of the optical trapping instrument.

The objectives of the project are to develop and construct a single aerosol particle optical trapping instrument equipped with temperature and relative humidity (RH) control, to characterize the phase transitions of organic aerosols across a range of atmospherically relevant conditions. The novelty of the counter-propagating Bessel beams (CPBB) optical trap is in its ability to trap submicron particles at sizes into the accumulation mode, and far from surfaces that interfere with the freezing process. In addition to evaluating the solid-to-liquid phase transitions of different-sized particles, specific interest was placed in studying the glassy behavior of organic aerosols under low RH conditions.

After initial instrument development, the aerosol optical trap was used to examine the glassy behavior of sucrose and raffinose, carbohydrates that model glassy organic particles in the atmosphere. Subsequently, we performed proof-of-concept studies by freezing several supercooled test substances: water and long-chain hydrocarbons (heptadecane, hexadecane, pentadecane, tetradecane). Following our initial publications, we have examined in-depth the freezing behavior of hexadecane, pentadecane, tetradecane, tridecane, dodecane, undecane, decane and 50/50 v/v mixtures of dodecane/tridecane.

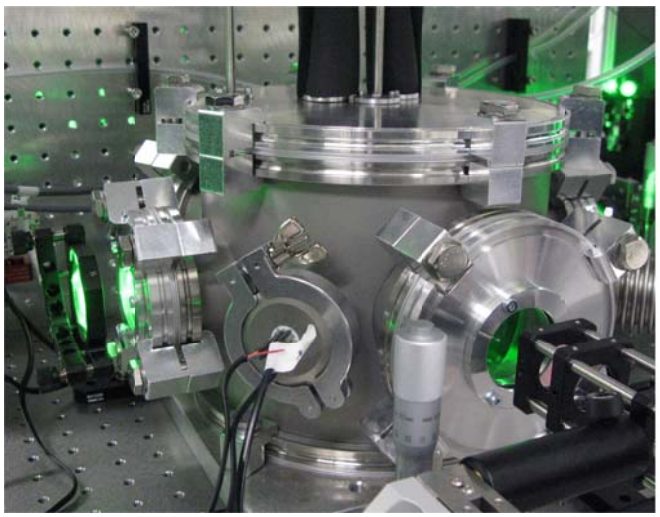

Fig. 2. The cold trapping instrument, with laser on.

1. Pratt, K. A.; DeMott, P. J.; French, J. R.; Wang, Z.; Westphal, D. L.; Heymsfield, A. J.; Twohy, C. H.; Prenni, A. J.; Prather, K. A. *Nat. Geosci.* **2009**, *2*, 398

Over the last reporting period, we have been successful in designing, constructing, and implementing the low-temperature CPBB optical trapping instrument, coolable to -50 °C. The apparatus has been used to study the homogeneous and heterogeneous freezing of single submicron- to micron-sized (~450 nm to 5500 nm (radius)) organic aerosols and water droplets in air. Our study presents the first reported observation of the freezing process of levitated single submicrometer-sized droplets in air using optical trapping techniques, including supercooled water droplets at 228 K. Our findings have been reported in a peer-reviewed journal (Lu et al. Review of Scientific Instruments, **85**, 095107 (2014); doi: 10.1063/1.4895118).

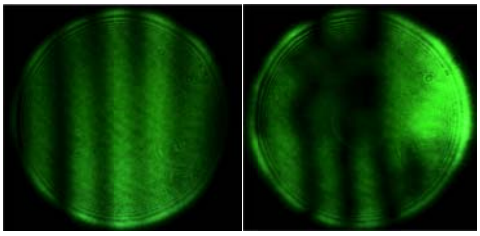

Fig. 3. Liquid dodecane particle (left) and frozen particle (right).

Additionally, we have explored the phase behavior of organic (sucrose) aerosol droplets and the conditions that lead to glass formation of these aerosols at room temperatures (Lu et al. Phys. Chem. Chem. Phys., 2014, **16**, 9819; doi: 10.1039/c3cp54233e). For these submicron- to micron-sized sucrose droplets, the glass-transition RH ranges from approximately 15–40%. These results corroborate well with similar single aerosol studies performed with larger droplets in an optical tweezer and electrodynamic balance.

Our current explorations into both the freezing of long-chain aliphatic hydrocarbons and the response of ultra-viscous organic aerosols to changes in ambient RH have furthered our understanding of the phase behavior of aerosols under atmospherically relevant conditions. In our room temperature studies, we have examined the potential glassy behavior of aerosol droplets composed of raffinose in the CPBB. From this work, the RH range for the glass transition RH was found to be between 45–60% RH, which agrees well with previous measurements performed in an optical tweezer with larger droplets. In addition, we are studying the freezing rates and nucleation mechanisms of long-chain hydrocarbons: decane, undecane, dodecane, tridecane, tetradecane, pentadecane, and hexadecane. Initial results suggest that the nucleation mechanism depends on hydrocarbon chain-length and supercooling temperature.

Our research provides a deeper understanding of the phase transitions of aerosols under well-controlled conditions, particularly the ice nucleating ability of organic aerosols. These topics are an integral part of the continued discourse on the aerosol indirect effect and global climate change. In the 2014 report of the Intergovernmental Panel on Climate Change (IPCC),[2] the aerosol indirect effect on climate continued to be identified as an important topic that "contribute(s) the largest uncertainty to estimates." Our scientific discoveries help to bridge this knowledge gap by supplying data for developing more accurate climate models, thus improving the predictive capabilities of these simulations and informing more appropriate science policy.

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2. Boucher, O.; Randall, D.; Artaxo, P.; Bretherton, C.; Feingold, G.; Forster, P.; Kerminen, V.-M.; Kondo, Y.; Liao, H.; Lohmann, U.; Rasch, P.; Satheesh, S. K.; Sherwood, S.; Stevens, B.; Zhang, X. Y. In *Clouds and Aerosols*; Cambridge University Press: Cambridge, United Kingdom and New York, NY, USA, 2013.