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Using ATom observations and models to understand what precursors drive NPF in the remote free troposphere

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Current estimates suggest that globally, about one third of low-level cloud condensation nuclei (CCN) originate from new particle formation (NPF) in the free troposphere. However, the exact mechanisms of how these new particles form and grow to CCN sizes are not yet well quantified. We investigate the formation of new particles and their initial growth in the remote marine atmosphere over the Pacific and Atlantic basins (~80 °N to ~86 °S using (1) gas-phase and size distribution measurements (0.003-4.8 μm) from the airborne-based NASA Atmospheric Tomography global survey (ATom; 2016-2018), (2) back trajectory data, and (3) two aerosol microphysics box models.

In the ATom observations, newly formed particles were ubiquitous at high altitudes throughout broad regions of the tropics and subtropics under low condensation sink conditions and were associated with upwelling in convective clouds. This pattern was observed over four seasons and both ocean basins.

In this study, we explore processes that govern NPF and growth in the tropical and subtropical free troposphere, discuss similarities and differences in NPF over both ocean basins, use box models to examine which nucleation schemes (e.g. binary, ternary, or charged) best explain the observations, and evaluate whether sulfuric acid precursors alone can explain the NPF and the initial particle growth. Comparing aerosol size distribution measurements with box model simulations shows that none of the NPF schemes commonly used in global models are consistent with observations, regardless of precursor concentrations. Newer schemes that incorporate organic compounds as nucleating or growth agents can plausibly replicate the observed size distributions. We conclude that organic precursor species may be particularly important in NPF in the tropical upper troposphere, even above marine regions.