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Water uptake of subpollen aerosol particles: hygroscopic growth, CCN activation, and liquid-liquid phase separation

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Pollen grains emitted from vegetation can release subpollen particles (SPP) that contribute to the fine fraction of atmospheric aerosols and may act as cloud condensation nuclei (CCN), ice nuclei (IN), or aeroallergens. Here, we investigate and characterize the hygroscopic growth and CCN activation of birch, pine, and rapeseed SPP. A high humidity tandem differential mobility analyzer (HHTDMA) was used to measure particle restructuring and water uptake over a wide range of relative humidity (RH) from 2 % to 99.5 %, and a continuous flow CCN counter was used for size-resolved measurements of CCN activation at supersaturations (S) in the range of 0.2 % to 1.2 %. For both subsaturated and supersaturated conditions, effective hygroscopicity parameters κ , were obtained by Köhler model calculations. Gravimetric and chemical analyses, electron microscopy, and dynamic light scattering measurements were performed to characterize further properties of SPP from aqueous pollen extracts such as chemical composition (starch, proteins, DNA, and inorganic ions) and the hydrodynamic size distribution of water-insoluble material. All investigated SPP samples exhibited a sharp increase of water uptake and k above ~95 % RH, suggesting a liquid-liquid phase separation (LLPS). The HHTDMA measurements at RH> 95% enable closure between the CCN activation at water vapor supersaturation and hygroscopic growth at subsaturated conditions, which is often not achieved when HTDMA measurements are performed at lower RH where the water uptake and effective hygroscopicity may be limited by the effects of LLPS. Such effects may be important not only for closure between hygroscopic growth and CCN activation but also for the chemical reactivity, allergenic potential, and related health effects of SPP.

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