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Secondary aerosol formation alters CCN activity in the North China Plain

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The formation of secondary aerosols (SA, including secondary organic and inorganic aerosols, SOA and SIA) were the dominant sources of aerosol particles in the North China Plain and can result in significant variations of particle size distribution (PNSD) and hygroscopicity. Earlier studies have shown that the mechanism of SA formation can be affected by relative humidity (RH), and thus has different influences on the aerosol hygroscopicity and PNSD under different RH conditions. Based on the measurements of size-resolved particle activation ratio (SPAR), hygroscopicity distribution (GF-PDF), $PM_{2.5}$ chemical composition, PNSD, meteorology and gaseous pollutants in a recent field campaign McFAN (Multiphase chemistry experiment in Fogs and Aerosols in the North China Plain) conducted at Gucheng site from November 16th to December 16th in 2018, the influences of SA formation on CCN activity and CCN number concentration (N_{CCN}) calculation at super-saturation of 0.05% under different RH conditions were studied. Measurements showed that during daytime, SA formation could lead to a significant increase in N_{CCN} and a strong diurnal variation in CCN activity. During periods with daytime minimum RH exceeding 50% (high RH conditions), SA formation significantly contributed to the particle mass/size changes in wide particle size range of 150 nm to 1000 nm, and led to an increase of N_{CCN} in particle size range of 200 nm to 300 nm, while increases in particle mass concentration mainly occurred within particle sizes larger than 300nm. During periods with daytime minimum RH below 30% in (low RH conditions), SA formation mainly contributed to the particle mass/size and N_{CCN} changes in particle sizes smaller than 300 nm. As a result, under the same amount SA formation induced mass increase, the increase of N_{CCN} was weaker under high RH conditions, while stronger under low RH conditions. Moreover, the diurnal variations of aerosol mixing state (inferred from CCN measurements) due to SA formation was different under different RH conditions. If the variations of the aerosol mixing state were not

considered, estimations of N_{CCN} would bear significant deviations. By applying aerosol mixing state estimated by number fraction of hygroscopic particles from measurements of particle hygroscopicity or mass fraction of SA from measurements of particle chemical compositions, N_{CCN} calculation can be largely improved with relative deviation within 30%. This study improves the understanding of the impact of SA formation on CCN activity and N_{CCN} calculation, which is of great significance for improving parameterization of SA formation in aerosol models and CCN calculation in climate models.