Hygroscopic behavior of individual ambient aerosol particles with X-ray chemical compositional analysis

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The hygroscopic behavior of ambient aerosols collected at Gosan (33°29'N, 126°16'E), Jeju island, Korea were investigated on a single particle level. The changes in size and phase transitions of individual aerosol particles during either humidification or dehydration processes were observed using optical microscopy. The morphology and chemical compositions of the particles were determined by offline SEM/EDX. 61% of the particles studied are reacted (aged) sea salt aerosols (SSAs), supported by the observation of different degrees of Cl^- depletion. Other particles are minerals, Fe-rich, and carbonaceous species, which are not hygroscopic except some reacted $CaCO_3$-containing mineral particles showing very low continuous growth/shrinkage due to presence of amorphous $Ca(NO_3)_2$. However, SSAs showed multiple hygroscopic (deliquescence and efflorescence) phase transitions. Pure salt solutions of NaCl, $MgCl_2$, and $NaNO_3$ were mixed to generate particles with different ratios of Cl^-:NO_3^-. Calibration curves obtained by plotting the mole fractions, i.e., [$X_{(Na, Mg)Cl}$] in these generated particles as a function of their X-ray intensity ratios, i.e., [Cl]/([Na] + 0.5[Mg]), were used to estimate the mole fractions in the SSAs from their respective X-ray intensities. During humidification, the mutual deliquescence transitions in Cl^--rich particles were not clear (apparently at RH = 63.5–65.3 %), whereas clear second transitions at RH= 69.5–73.5 % were observed. During dehydration, these particles showed mostly single-stage efflorescence at RH= 39.6–50.5 %, due to simultaneous crystallization of salts. On the other hand, Cl^--depleted particles showed clear first and second transitions at RH = 63.8 % and 65.4–72.9 %, respectively, during humidification. During dehydration, Cl^--depleted particles showed multiple efflorescence transitions at RH = 36.2–46.0 % and 17.9–33.0 %, respectively. The hygroscopic growth factors seems to be affected by the presence of organic species. The phase diagrams obtained by plotting the deliquescence and efflorescence RHs (DRHs and ERHs) as a function of the mole fractions [$X_{(Na, Mg)Cl}$] show that the hygroscopic behavior of the reacted SSAs are dominated by NaCl and $NaNO_3$ [1], whereas contribution due to SO_4^- seems to be minor.

[1] Gupta et al. (2015) Atmos. Chem. Phys., **15**, 3379–3393.