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Mercury speciation and adsorption in cloud water over the North China Plain

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Cloud effects on heterogeneous reactions of atmospheric mercury (Hg) and thus on Hg geochemical cycling are poorly understood due to our knowledge gap in cloudwater Hg chemistry. To investigate Hg species in cloud water and their behaviors in cloud processes, we collected cloud water samples using CASCC sampler in summer 2015 at the summit of Mt. Tai (1545 m) over the North China Plain, where air pollution is severe. Total mercury (THg), dissolved Hg (DHg) and methylmercury (MeHg) were analyzed. Ancillary water-soluble ions, dissolved organic matter (DOM), small carboxylic acids and cloud physical parameters were also determined. The results showed that cloudwater THg concentrations ranged from 10.2 to 773.3 ng L-1 with an average of 70.5 ng L-1, which was the highest in clouds/fogs worldwide until now. Two-thirds of THg was comprised of particulate Hg (PHg), probably contributed by abundant aerosol particles. MeHg, which was almost all dissolved, had moderate concentration level of 0.15 ng L-1 on average. Chemical equilibrium simulations using Visual MINTEQ v3.1 indicated that Hg complexes by DOM dominated the DHg speciation, which was highly pH dependent and competed with chloride. Concentrations and speciation of Hg were irreversibly altered by cloud processes, during which significant positive correlations of PHg and MeHg with cloud droplet number concentration (Nd) were observed. However, unlike direct contribution to PHg from cloud scavenging of aerosol particles, abiotic DHg methylation was likely the source of MeHg. The measured Hg adsorption coefficient Kad (mean of 70 L g-1) was much higher than that employed in transport model (45 L g-1), and exhibited an inverse-power relationship with cloud residues content. SEM-EDS analyses of cloud residue particles indicated that fly ash particles, compared to mineral particles, could enhance physical and chemical adsorption of Hg due to larger specific surface area and more abundant carbon binding sites. Our findings imply that aerosol-cloud interactions over the air polluted northern China may bring substantial Hg into cloud droplets, impacting atmospheric heterogeneous reactions and cycling of Hg.