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Long-term cloud water chemistry observed at Mt. Bamboo, West Pacific Ocean during northeast monsoon seasons in 1996-2016

Thuy Bui Thu, Wei-Ti Tseng, Stephen Griffith, and Neng-Huei (George) Lin
Department of Atmospheric Sciences, National Central University, Taoyuan City, Taiwan (nhlin@cc.ncu.edu.tw)

This study investigates the long-term chemistry of cloud water at Mt. Bamboo (25.18^{o}N, 121.53^{o}E; 1,100 m MSL) in northern Taiwan under the primary influence of Asian continental outflows during the northeast monsoon seasons in 1996-2016. Cloud water was collected on an hourly basis and measured for pH and inorganic soluble ions. In total, around 180 cloud events were observed and 6433 samples were collected. HYSPLIT backward trajectory analysis was conducted, illustrating the majority of air masses originated from northern and eastern China. To our knowledge, this is the first study in the region that verified the long-term relationship between Asian continental out flow and cloud water in the West Pacific Ocean. The long-term volumetric weighted mean (VWM) pH was 3.97 for cloud samples and 4.21 for mixed samples (cloud and rain together), respectively. Major contributors in cloud water were sea-salt components (Na^+, Cl^-) and nss (non sea-salt)-SO_4^{2-} which might be associated with Asian continental outflow. During the whole sampling period, the nss-SO_4^{2-} ion concentrations VWM was 131.0 μeq l^{-1} (cloud type) and 75.4 μeq l^{-1} (mixed type), while for NO_3^- was 63.4 μeq l^{-1} (cloud type), and 27.3 μeq l^{-1} (mixed type). Average cloud loading during the entire sampling period (averaging cloud and mixed type samples) of nss-SO_4^{2-} was 1.95 μg m^{-3}and that of NO_3^- was 1.02 μg m^{-3}. Mann-Kendall test and Sen's slope were applied to statistically assess the temporal trends of the measured variables. The nss-SO_4^{2-} cloud loading showed a significant decreasing trend over time, which can likely be attributed to SO_2 emission reduction in China, while pH, NO_3^- and NH_4^+ revealed no significant temporal trends. A source (Asian continental outflow) and receptor (West Pacific Ocean cloud water) relationship was verified by using the nss-SO_4^{2-}/NO_3^- cloud loading ratios and China annual emission SO_2/NO_x (CNBS data) ratios from 1996-2014. A significant correlation coefficient was established (R=0.66, p$<$0.05), confirming the strong relationship over a long-term period.