**Institute of Atmospheric physics, National Central University**

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A long term record (2001–2015) of the ratio of nitrate to non-sea-saltsulfate

 in precipitation over East Asia. Implication of the change in anthropogenic emission

**Abstract**

The study identify the long-term record of acidifying species in precipitation over East Asia (nitrate NO3− andnon-sea-salt sulfate nss-SO42−). They compiled groundbased observations of precipitation chemistry over China, Korea, and Japan from 2001 to 2015based on the Acid Deposition Monitoring Network in EastAsia (EANET). Theanthropogenic NOxand SO2 emissions and the NOx/SO2emission ratio were also analyzed. The satellite observationsof the NO2 and SO2 column density to capture the variation in emissions were applied.

The period of analysis was divided into three phases: Phase I (2001–2005), Phase II (2006–2010), and Phase III (2011–2015). The longterm trend in the NO3− concentration in precipitation was not related to the variation in NOx emission and the NO2 column. The nss-SO42− concentration in precipitation over China, Korea, and Japan was partially connected to the changes in SO2 emissions from China, but the trends were not significant. The initial flat trend during Phase I was due to increases in both NOx and SO2 emissions in China, the increase trend during Phase II was triggered by the increase in NOx emissions and decrease in SO2 emissions in China, and return to a flat trend during Phase III was caused by declines in both NOx and SO2 emissions in China. These results suggest that emissions in China had a significant impact not only on China but also on downwind precipitation chemistry during the 15-year period of 2001–2015.

In terms of wet deposition, the NO3− wet deposition over China, Korea, and Japan did not change dramatically. However, the nss-SO42− wet deposition declined over China, Korea, and Japan from Phase II to III due to a strong decrease in the nss-SO42– concentration in precipitation accompanied by a reduction in SO2 emission from China. These findings indicated that the acidity of precipitation shifted from sulfur to nitrogen.

**Key works:** Long term trend, NO3–/nss-SO42– ratio.