



## **Abrupt increase of nitrate in PM<sub>2.5</sub> during winter haze in Seoul**

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In January 2018, PM<sub>2.5</sub> warnings have been issued for three days across the Korea peninsular, when it was increased up to 106  $\mu\text{g}/\text{m}^3$ . The rapid increase of PM<sub>2.5</sub> mass was associated with increase in nitrate by 10  $\mu\text{g}/\text{m}^3$  and NO<sub>2</sub> by 103 ppbv within 3 hours from 10:00 and 13:00. In Seoul, the maximum NO<sub>2</sub> is usually found around 9 am. While the PM<sub>2.5</sub> mass was best correlated with ammonium ( $r=0.95$ ), the increase in sulfate was insignificant. Therefore, it is critical to understand the rapid conversion mechanism of nitrate and the plausible pathways. The contribution of HNO<sub>3</sub> is very unlikely under overcast conditions in winter (average cloud coverage of almost around 10). In fact, the calculated HNO<sub>3</sub> was not sufficient to explain the observed NO<sub>3</sub> concentration. It might be possible that N<sub>2</sub>O<sub>5</sub> was heterogeneously converted into HNO<sub>3</sub> in liquid droplets during nighttime and mixed with surface air rich in NH<sub>3</sub> during the daytime, leading to NH<sub>4</sub>NO<sub>3</sub> formation. However, it is quite not likely because of the low temperature (minimum surface temperature of -2.5 °C) and low O<sub>3</sub> concentration (maximum O<sub>3</sub> of 40 ppbv). The ceilometer and LIDAR measurements reveal the shallow thin cloud and boundary layer height during the night, which started expanding around 9 10 am. Therefore, we suggest that the vertical mixing played a key role in increase of nitrate, which was enriched in cloud droplets (ice crystal) aloft. Regarding ammonium, it is not clear if it was combined with NO<sub>3</sub> or partitioned into aerosol after being mixed with surface air, which will be further investigated.