



## Aerosol volatilisation in the urban surface layer

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Vertical flux measurements were made above Beijing during two contrasting periods in Nov/Dec 2016 and June 2017, using the micrometeorological eddy-covariance (EC) technique. For this purpose, a fast response ultrasonic anemometer and two particle counters were operated at a height of 102 m on the 325 m meteorological tower at the Institute for Atmospheric Physics (IAP) of the Chinese Academy of Sciences: a water-based condensation particle counter (CPC3785, TSI Inc.), and an Ultra High Sensitive Aerosol Spectrometer (UHSAS, DMT) providing total and size-segregated particle fluxes over the size range 80 to 1000 nm. Moreover, a sample inlet brought air to a measurement container at the foot of the tower, where an Aerosol Mass Spectrometer (HR-ToF-AMS; Aerodyne Research Inc) provided a fast response measurement of aerosol chemical composition, suitable for flux calculations of non-refractory ammonium, nitrate, sulfate, chloride and organic aerosol in PM<sub>1</sub>. The AMS total organic aerosol flux was further decomposed by application of Positive Matrix Factorisation to virtual eddy-accumulation data, which was derived from the EC data.

As expected, the winter measurements were dominated by emissions of all compounds and size-ranges. The summer measurements showed strong emissions of total particle number as well as primary organic aerosol components associated with fossil fuel combustion and cooking emissions. However, surprisingly, the total submicron mass fluxes as derived independently by UHSAS and AMS were downwards, suggesting that together, the surface and lowest 100 m of the urban surface layer act as a net sink for PM<sub>1</sub>. Analysis of the composition and size-resolved fluxes shows that this net downward flux reflected ammonium, nitrate, chloride and the most volatile organic aerosol component and carried by the accumulation mode ( $> 0.22 \mu\text{m}$ ). The implied deposition rate was too fast for this aerosol fraction to reach the ground. Evidently, these (semi-)volatile aerosol components evaporated very effectively below the measurement height, due to gradients in temperature.

Overall, this process results in an airborne source of ammonia, nitric acid, hydrochloric acid and organic compounds in the gas-phase with implications for atmospheric chemistry. For example, the ammonium volatilisation accounts for on average 15% of the ammonia emission also measured on the tower, using an Aerodyne quantum cascade laser absorption spectrometer. It further limits our ability to reproduce measurements with atmospheric chemistry and transport models with a vertical resolution that is insufficient to resolve the shift in thermodynamic equilibria.