6.161 Observations of particle organic nitrate from airborne/ground platforms: Insights into vertical/geographical distribution, gas/particle partitioning, losses, and contribution to total particle nitrate.

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Abstract:

Organic nitrate formation in the atmosphere represents a sink of NO_X and termination of the HO_X/NO_X- ozone formation cycles, can act as a NO_X reservoir transporting reactive nitrogen, and contributes to secondary organic aerosol formation. However, particle-phase organic nitrates (pRONO₂) are rarely measured and thus poorly understood. We use simultaneous measurements of pRONO₂ and of total (gas+particle) organic nitrate

(totRONO₂), organic aerosols (OA), and ammonium nitrate from the DC3 and SEAC⁴RS aircraft and several ground campaigns to investigate vertical/geographical distributions, gas/particle partitioning, losses, and contributions to total particle nitrate (pTotNO₃) over North America. Apportionment and quantification with aerosol mass spectrometry is evaluated. The fraction of pTotNO3 that is organic increases steeply with decreasing pTotNO₃, approaching 100% at low pTotNO₃, primarily at rural/remote locations. pRONO₂ and totRONO₂ concentrations show strong vertical gradients, with a steep decrease from the top of the boundary layer (BL) up through the residual layer. However, pRONO2 was 10-30% of totRONO2 with little vertical gradient in gas/particle partitioning from the BL to upper troposphere (UT). pRONO₂ contribution to OA shows a moderate increase with decreasing OA in the BL and free troposphere (~2-3% by mass of nitrate group) with higher contributions at the lowest OA (5-8%), mostly observed in the UT. In the BL, RONO2 gas/particle partitioning shows a trend with temperature, with higher particle-phase fraction at lower temperatures, as expected from partitioning theory. However, the temperature trend is much weaker than for single compound partitioning, which may be due to a broad mixture of species. Little to no dependence of pRONO-2/OA on RH or estimated particle water was observed in the BL, suggesting that losses of pRONO₂ due to hydrolysis are too rapid to observe in this dataset and there may be a substantial fraction of pRONO₂ species that are not prone to rapid hydrolysis.