

EGU2020-21227

<https://doi.org/10.5194/egusphere-egu2020-21227>

EGU General Assembly 2020

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Aerosol acidity as a driver of aerosol formation and nutrient deposition to ecosystems

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Nitrogen oxides (NO_x) and ammonia (NH₃) from anthropogenic and biogenic emissions are central contributors to particulate matter (PM) concentrations worldwide. Ecosystem productivity can also be strongly modulated by the atmospheric deposition of this inorganic "reactive nitrogen" nutrient. The response of PM and nitrogen deposition to changes in the emissions of both compounds is typically studied on a case-by-case basis, owing in part to the complex thermodynamic interactions of these aerosol precursors with other PM constituents. In the absence of rain, much of the complexity of nitrogen deposition is driven by the large difference in dry deposition velocity when a nitrogen-containing molecule is in the gas or condensed phase.

Here we present a simple but thermodynamically consistent approach that expresses the chemical domains of sensitivity of aerosol particulate matter to NH₃ and HNO₃ availability in terms of aerosol pH and liquid water content. From our analysis, four policy-relevant regimes emerge in terms of sensitivity: i) NH₃-sensitive, ii) HNO₃-sensitive, iii) combined NH₃ and HNO₃ sensitive, and, iv) a domain where neither NH₃ and HNO₃ are important for PM levels (but only nonvolatile precursors such as NVCs and sulfate). When this framework is applied to ambient measurements or predictions of PM and gaseous precursors, the "chemical regime" of PM sensitivity to NH₃ and HNO₃ availability is directly determined.

The same framework is then extended to consider the impact of gas-to-particle partitioning, on the deposition velocity of NH₃ and HNO₃ individually, and combined affects the dry deposition of inorganic reactive nitrogen. Four regimes of deposition velocity emerge: i) HNO₃-fast, NH₃-slow, ii) HNO₃-slow, NH₃-fast, iii) HNO₃-fast, NH₃-fast, and, iv) HNO₃-slow, NH₃-slow. Conditions that favor strong partitioning of species to the aerosol phase strongly delay the deposition of reactive nitrogen species and promotes their accumulation in the boundary layer and potential for long-

range transport.

The use of these regimes allows novel insights and is an important tool to evaluate chemical transport models. Most notably, we find that nitric acid displays considerable variability of dry deposition flux, with maximum deposition rates found in the Eastern US (close to gas-deposition rates) and minimum rates for North Europe and China. Strong reductions in deposition velocity lead to considerable accumulation of nitrate aerosol in the boundary layer –up to 10-fold increases in PM_{2.5} nitrate aerosol, eventually being an important contributor to high PM_{2.5} levels observed during haze episodes. With this new understanding, aerosol pH and associated liquid water content can be understood as control parameters that drive PM formation and dry deposition flux and arguably can catalyze the accumulation of aerosol precursors that cause intense haze events throughout the globe.