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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol acidity and liquid water content regulate the dry deposition of inorganic reactive nitrogen" *by* Athanasios Nenes et al.

Anonymous Referee #1

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This is a nice study that develops an analytical framework for understanding how particle acidity and liquid water content indirectly modulate the rate of reactive nitrogen deposition through their influence on gas-particle partitioning of total nitrate and total ammonium. The work is presented in a very clear, simple way and makes use of valuable observational datasets from sites around the world to illustrate the application of the conceptual framework. Overall, the study improves how we think about the connections between aerosol thermodynamics and deposition and suggests how the framework might be used to better evaluate and characterize performance of the photochemical models widely used in decision making. The paper is a nice companion to the authors' 2020 ACP paper on how aerosol acidity and liquid water content determine

the sensitivity of particulate matter to the availability of ammonia and nitrate. I have a few comments below for the authors to consider.

General comments:

1. The authors define chemical regimes for deposition velocity that transition from "slow" to "fast" when the particle-phase fraction of the total amounts transition from greater than 90% to less than 90%. One might expect the regimes to vary from "slow" to "moderate" to "fast" as a function of the gas-particle partitioning and that fast deposition (associated with gas-phase deposition rates) would not occur until much more than 10% of the total is in the gas phase. It would be worthwhile for the authors to add a sentence or two describing the slow-fast transition and choice of alpha, beta=0.9. Is the idea that deposition will approach the gas-phase rate once appreciable amounts are in the gas phase because rapid gas-phase deposition would cause evaporation from particles to maintain equilibrium and ultimately rapid removal of the total?

2. The authors use a value of k=10 to represent the ratio of the deposition velocity of the gas phase to the deposition velocity of the particle phase. This is reasonable for the order-of-magnitude considerations presented, but ACP readers would likely be interested in some additional information and references on the deposition rates of gas and particle phase species. Can the authors provide some references for the values that led to the choice of k=10? It would also be interesting know ranges of values for typical variations in ambient conditions either from model predictions or the literature.

3. On p. 4, the authors mention their acidity/water framework for understanding the sensitivity of PM to "emissions of ammonia and nitrate". As the authors are aware, very little nitrate is directly emitted, and nitrate forms largely from oxidation of NOx emissions. A challenge in using the thermodynamic framework is that the conversion of NOx to HNO3 may be oxidant limited such that decreasing NOx emissions could potentially increase HNO3 concentrations. Such nonlinearities in gas-phase oxidation complicate the relationship between the thermodynamic framework and precursor

emissions. In the case of ammonia, changes in ammonia emissions influence cloud pH and therefore in-cloud production of sulfate, and the feedbacks of ammonia on this sulfate production are not accounted for in the framework. Given these complications, I recommend that the authors reword statements on p. 4 to avoid directly connecting the thermodynamic framework to the precursor emissions.

4. Is there any synergy in diurnal profiles of aerosol thermodynamics and deposition? For instance, partitioning to the particle phase tends to be greater during cooler conditions overnight with lower wind speeds, which would presumably influence aerodynamic resistance.

Specific comments:

– P. 1, line 27: clarify that NH3 and HNO3 are intended to represent “total” amounts, not just the gas phase

– P. 1, line 30: Sentence ends in comma, should be period

– P. 1, line 31: Should “reduce the deposition” be “reduce the local deposition” since all of the material will eventually deposit?

– P. 3, line 2: Should “NRC” be “NAS” to match the reference list?

– P. 3, line 11: Recommend changing “significant fraction of Nr” to “significant fraction of Nr deposition”. NOx represents a large amount of oxidized nitrogen but a relatively smaller amount of Nr deposition because its deposition rate is relatively low.

– P. 4, line 1: recommend adding “for deposition” after “regimes”

– P. 4, lines 14-15: “velocity deposition” should be “deposition velocity”

– P. 4, Figure 1: can any intuition be provided on what 10 and 250 ug/m3 of water correspond to in terms of typical PM levels?

– P. 6, line 4: I think pH should be “>” than pH’ in regime 3. Please check.

– P. 6, Figure 2: The larger “fast” region at higher temperature suggests that deposition would be slower and long-range transport more prevalent in winter. Might be worth mentioning the implications of the work for winter/summer differences somewhere.

– P. 7, eqn 5: v is missing the subscript “p”

– P. 8, Figure 4b: a linear (rather than log) scale seems more intuitive to me for the normalized flux plot. Please consider revising.

– P. 9, line 8: might want to indicate “total” in front of nitrate and ammonia for clarity

– P. 10, line 7: the nature of the “positive feedback” is not explicitly stated here and might not be clear to all

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