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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling reactive ammonia uptake by secondary organic aerosol in CMAQ: application to continental US" *by* Shupeng Zhu et al.

Anonymous Referee #1

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This manuscript presents a model sensitivity analysis to quantify the effects of NH3 uptake by SOA on surface PM2.5 in the US. While this is the first modeling study I know of that simulates the regional effect of this reaction, the effect is parametrized simply as a sink for NH3, not a source for SOA, and thus the air quality impacts resulting from this are all related to changes in the inorganic PM species and aerosol acidity. The aerosol acidity change in turn drives some SOA changes resulting from the acid-catalyzed SOA formation pathways. The paper is generally well-written. I recommend it to publish in ACP after the following weakness/questions are addressed.

Major Comments:

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Discussion paper

1. Pg 4, line 20-29: the discussion of lab studies would indicate the parameterization used in the manuscript is oversimplified and may not fully represent the lab experiments. First, lab experiments show that only about 10% of SOA molecules can react with NH3 to form nitrogen-containing organic compounds. Second, the Liu et al. (2015) study, which the parameterization is largely based on, reported only a few SOAs can uptake NH3. Despite these, the manuscript assumes all SOA uptakes NH3. Although they chose to lower the uptake coefficient to compensate for the fact that not all SOA uptakes ammonia, given the spatiotemporal variability of SOA sources, a uniformly-applied lower uptake coefficient to all the SOA species would not have the same effect as that of applying a higher uptake coefficient selectively to several SOAs. Since the CMAQ model can explicitly simulate SOA species by origin (e.g. isoprene vs. terpene SOA) and by oxidation pathways, would it make more sense to parameterize the uptake only to the few SOA species that lab experiments have shown to have such ability and use the lab derived uptake coefficient directly?

2. Model evaluation: this section presents just general PM evaluation and does not have a clear focus on evaluating the parameterization scheme developed. At a minimum, the sites where the model evaluation is based on should be labeled on the model concentration maps from Figure 1 – 6 with the corresponding model biases. The way they are presently listed in Tables is not illustrative and does not help the readers understand the simulation results in the context of model biases. The concentration maps show large spatial variability of the NH3 uptake effect on different aerosol components (e.g. over Southeast US, Central Great Plains), but they are all based on model simulations without observational backup. If there are some observational sites located within those regions where large model sensitivity is found, more discussions of the model bias or improvement after the parameterization scheme should be given to them.

3. Section 3.2.3: the impact of the NH3 uptake on organic PM is large, which apparently contradicts with the earlier claim (pg 4, line 10-15) that the NH3 uptake does not create SOA mass. Later on the authors explained that it was due to changes in

aerosol acidity, which in turn drives SOA changes from the acid-catalyzed formation pathways. To avoid confusion, I suggest the authors first present the acidity changes (maybe make it a separate sub-section) and then present the acidity-induced SOA changes. In the SOA section, state upfront that the SOA changes are not caused by the parameterization creating more SOA mass by itself.

Technical Comments:

1. Equation 1: is the aerosol surface area (Sj) dry or wet area? I think it should be wet area, i.e. considering hygroscopic growth of aerosols under ambient RH conditions. If wet area, how does the model treat SOA hygroscopic growth?

2. Pg 4, Line 10: “an SOA compounds” should be “an SOA compound”

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