Response to Anonymous Referee #2

Manuscript: *Significant contrasts in aerosol acidity between China and the United States*

Manuscript number: acp-2020-879

Journal: Atmospheric Chemistry and Physics

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Comment 1

Overall, the authors have done an excellent job addressing referee comments. The updated manuscript is greatly improved and is nearly ready for publication. I do have one substantive comment and several minor technical corrections. All of the comments are straightforward to address.

Response

We would like to thank the reviewer for the time reviewing our manuscript and providing supportive comments. We have listed the point-by-point responses below. We hope that our revision and this new version of the manuscript have addressed all the reviewer's concerns.

Comment 2

In the revised analysis, one of the findings, in particular, stood out to me as quite novel and significant. This analysis (discussed in lines 385 – 405 and presented in Figure S7) found that pH changed when the concentrations of all chemical components were scaled by a common factor, keeping meteorological parameters constant. I agree with the authors that this is a surprising result. Although the authors cite several references in lines 428-429 that discuss the effects of concentration and chemical composition on pH, none of those studies showed this specific result (that pH changed with mass concentration even though all chemical component mole fractions did not change). I think that the authors should consider highlighting this result more prominently in the paper, e.g., in the abstract.

Response

In response to this comment, we highlighted the results of pH change with mass concentrations by rewriting the following sentences in abstract in lines 24-29.

"Our assessment shows that the differences in mass concentrations and chemical composition play equally important roles in driving the aerosol pH difference between China and the US — increasing the aerosol mass concentrations, but keeping the relative component contributions the same, in the US to the level in China (by a factor of 8.4) increases the aerosol pH by ~1.0 unit, and further shifting the chemical composition from US conditions to China's that is richer in ammonia increases the aerosol pH by ~0.9 units."

Technical Correction 1

- Throughout the manuscript, the ‘dot’ in μg m-3 should be removed.

Response
Removed throughout.

Technical Correction 2

- In Figures 1, 4, and 5, change figure labels from “the United States” to “United States”.

Response
Changed in response to this comment.

Technical Correction 3

- Line 302: delete “process”.

Response
Deleted in response to this comment.

Technical Correction 4

- Line 389: comma after “pH”

Response
Added in response to this comment.

Technical Correction 5

- Sentence beginning on line 390 should be edited for grammar.

Response
In response to this comment, the sentence was revised as follows,
“*The second step that changes the chemical composition shows a further increase of 0.76 units in the aerosol pH, which is mainly achieved through the H_{air}^{+}-modifying pathway (0.88 units). The LWC-modifying pathway plays a minor role (-0.11 unit) in this step (Fig. S7 (d), (e), (f)).*”

Technical Correction 6

- Entire paragraph beginning on line 395 should be edited for grammar.

Response
In response to this comment, the paragraph was edited for grammar and revised as follows,
“*It is surprising, in the first step, that pH changed when the concentrations of all chemical components were scaled by a common factor. This means that pH changes with mass concentration of the aerosol (gas+particle) even though all chemical component mole fractions hold. Further investigation shows that, increasing the aerosol mass concentration drives TNO_3 and TNH_3 partitioning toward particle phases—$\varepsilon(NH_4^+)$ and $\varepsilon(NO_3^-)$ increase from 0.4 to 0.6 and from 0.6 to 0.98, respectively. Given the weak acidity of NO_3^-, the particle is ultimately neutralized by the increased NH_4^+. The repartitioning in response to the increase in mass concentration is thus key to the pH shift and can be explained by Henry’s Law, i.e., $[A_{aq}]=H_A \cdot p_A$, where $[A_{aq}]$ is the aqueous-phase concentration of component A in the unit of moles per liter water, p_A is the partial pressure of A in the gas phase, and H_A is Henry’s law coefficient (Seinfeld and Pandis, 2006). $[A_{aq}]$ is proportional to c_A / LWC (c_A denotes the particle-phase concentration of A, note that LWC and c_A are expressed as mass per unit volume of air, and $[A_{aq}]$ is expressed as moles per unit volume of water). Increasing the concentrations of all chemical components by a*

common factor increases p_A (due to the increase in the gas-phase concentration of A) but does not change [A_{aq}] (because both c_A and LWC increases in the same direction by the same magnitude). According to the Henry's Law, more gas-phase A will thus shift toward the particle phase to achieve a thermodynamic equilibrium of the new system."

Technical Correction 7

- Line 407: what is meant by "weak acidic capacity"? suggest revising this sentence.

Response

We have replaced "weak acidic capacity" with "weak acidity" in the revised manuscript.

Technical Correction 8

- Line 411: change "sensitive" to "sensitivity".

Response

Changed in response to this comment.

Technical Correction 9

- Line 417: delete "found".

Response

Deleted in response to this comment.

Technical Correction 10

- Line 418-419: awkward as written – suggest revising this sentence

Response

In response to this comment, this sentence was revised as follows,

"*In populated continental regions, mass fractions of TNH_3 are often high (Bencs et al., 2008; Behera and Sharma, 2010; Zheng et al.,2015; Cheng et al., 2016; Guo et al., 2017b), and an increase in mass concentration thus typically increases the aerosol pH.*"

Technical Correction 11

- Line 427: what is meant by "availability of the corresponding aerosol components"? suggest revising this sentence.

Response

In response to this comment, "availability" was changed to "fractions".

Technical Correction 12

- Line 440: comma after "study".

Response

Added in response to this comment.

Technical Correction 13

- Line 460: comma after "SO4".

Response

Added in response to this comment.

Technical Correction 14

- Line 462: “linking to the pH difference” is awkward - suggest revising this sentence.

Response

In response to this comment, this sentence was revised as “*Further investigation highlights two pathways related to the pH difference—one associated with changes in LWC and the other associated with changes in* H_{air}^{+}.”