Response to Reviewer #3 comments:

Ibikunle et al. developed a new algorithm to estimate aerosol pH when gas-phase ammonia measurements are not available. They applied this method to the dataset from KORUS-AQ aircraft measurements to identify effective PM control strategies in South Korea using the framework developed by the same group which separates sensitivity of PM to NH3 and HNO3 availability into different domains. This paper presents an important topic, and will contribute to our understanding of the connections between PM level and aerosol pH. This paper is publishable after considering the following revisions.

Answer: *We thank the reviewer for the enthusiastic response and for feedback that has improved the manuscript. Below, we include the response to comments and questions raised.*

General comments:

Size-dependent aerosol pH could be very important and interesting to explore. In Line 168- Line 172, the authors claim that particles are internally mixed in their study and cite some previous work. However, I couldn't find a size distribution plot for different species in the aircraft AMS in Nault et al. 2018 to support this statement. I suggest that the authors include a P-TOF figure from the aircraft AMS in the Supplementary Information to discuss more.

Answer: *These are interesting points and we will try our best to provide the relevant figure and discussion points in the revised manuscript.*

From the ground-based AMS in Kim et al., they show that organics dominate fine particle mass in the smaller size range, and are major components in larger size range as well. This brings up another issue of how organic-inorganic phase separation affects the pH estimation method used in this work. I think the authors could extend a little bit more on mixing state and the role of organics in the paper to address possible size-dependent pH.

Answer: *Generally speaking, for aerosol containing oxidized organics and humidity above a certain level (e.g., 40%), the single-phase assumption tends to work well. We will address these discussion points in the revised manuscript.*

Minor comments:

1. The term "NVCs" first shows up in Line 127 but is explained later in Line 148. Please explain NVCs in Line 127.

Answer: *Done!*

2. In Figure 2 flow chart, when NH_3 varies much between iterations (the "yes" scenario), shouldn't the algorithm continue to update NH_3total? It seems to me the "yes" and "no" decisions should be reversed here.

Answer: *Indeed so. The typo is now corrected.*

3. In Figure 6, it may be a little misleading to use "NH3 – dominant" because it makes people think that gaseous ammonia dominates total NH3 but in reality ε(NH4+) is high in this region. It can be

labeled as “NH3 sensitive” as shown in Figure 1 in Nenes et al 2020a. Similarly, “HNO3 - dominant” can be “HNO3 sensitive”.

Answer: *These are good points! and will be included in the revised text.*

4. In Figure 6, the caption indicates that the aerosol liquid water content is 10 _g m-3 but in Line 281, the text reads “predicted liquid water content (13.78+/- 10.52 _g m-3).” What is the reason behind the different numbers?

Answer: *We have now clarified the text to clear this apparent discrepancy.*

5. Line 349, “ammonium” in the particle phase?

Answer: *Corrected*

6. Are the PM mass concentrations here PM1? Need to clarify.

Answer: *This is now clear in the text.*

7. What is the difference between “HNO3/NH3 sensitive domain” (in Line 365 and in Line 412) and “NH3-HNO3 sensitivity domain” (Line 381). If they refer to the same domain, better to be consistent. “HNO3/NH3” indicates it is a ratio.

Answer: *We now keep a consistent naming shceme throughout.*

8. Different fonts were used in figure captions in Supplementary Information. Please be consistent.

Answer: *Consistent fonts are now used.*