Manuscript ID: acp-2021-455
TITLE: Long-term trends and drivers of aerosol pH in eastern China

We thank the editor and the reviewers for the comments concerning our manuscript. They are valuable in helping us improve our manuscript. Below please find our point-by-pint responses to reviewers' comments.

Comments to the Author

Major Comments

1. The estimates ALWCo seem unreasonably small (lines 120 - 127)? How was organic aerosol measured? Was it $PM_{2.5}$ as well, or was it PM_1?

Response: The concentration of organic aerosol was estimated by multiplying the measured concentration of organic carbon by a factor of 1.6 (Turpin and Lim, 2001). A Thermal/Optical Carbon Aerosol Analyzer (model RT-4, Sunset laboratory Inc.) equipped with a $PM_{2.5}$ cyclone was used for the organic carbon measurement. The annual concentrations of organic carbon in Shanghai were 5.6–10.6μg/m^3 from 2011 to 2019, and the relative humidity were 69-75%. $ALWC_o$ was calculated by the following equation (Guo et al., 2015).

$$ALWC_o = \frac{m_{org}\rho_w}{\rho_{org}} \frac{k_{org}}{(\frac{1}{RH}-1)} \quad (1)$$

where m_{org} is the mass concentration of organic aerosol, ρ_w is the density of water (ρ_w=1.0g/cm^3), ρ_{org} is the mean density of organics assumed to be 1.4g/cm^3)(Guo et al., 2015), and k_{org} is the hygroscopicity parameter of organic aerosol ($k_{org} = 0.087$)(Li et al., 2016). Adopting these values, we estimate that the annual $ALWC_o$ and $ALWC_i$ from 2011 to 2019 are approximately 1.4–2.5μg/m^3 and 25.8–35.8μg/m^3, respectively. That is $ALWC_o$ accounted for 4.3%–7.5% of the total aerosol liquid water content from 2011 to 2019.

2. I found the convention used in Figures 1b, 3, and 5 very confusing. The pie charts below each figure are useful and seem straightforward to interpret, but the bar charts need substantial editing. For example, in Figure 1b, the effect of NVCs on the pH trends changes signs with time. Ultimately, using Fig. S6, I was able to deduce that the positive value associated with NVCs for 2011-2013 meant that NVCs had gone up, and the negative value associated with NVCs for 2013 – 2015 meant that NVCs had gone down. However, it took far too much time to interpret and is still not easily understandable even after spending much time on it. The convention used by Tao and Murphy (2021) is much clearer – I suggest edits to follow their approach.

Response: Thanks for the comment. To study the driving factors of aerosol pH, different sensitivity analysis methods have been used in previous studies (Ding et al., 2019; Tao and Murphy, 2021; Zheng et al., 2020). The convention used in Tao and Murphy (2021) defined the base scenario as the average condition, aiming at illustrating the contribution of different factors to the deviation from the base scenario. However, the base scenario can change with the analysis time periods. In comparison, our bar plot here aimed at showing the factor contribution of the ΔpH between two adjacent scenarios (i.e., two continuous years or two continuous hour periods), and is not subject to change in the average conditions. That is, our plots emphasized differently with that used in Tao and Murphy (2021). We've clarified this in the revised figure captions. In addition, to provide more viewpoints, we've added the figures with Tao and Murphy's approach in the supplement following the reviewer's suggestion.

Changes in manuscript:

(1) Line 204-206: "Figure 1b shows the contributions of individual factors to the ΔpH from 2011 to 2019. Here the bar plots indicate the factors contributing to the ΔpH between two adjacent scenarios, e.g., 2011 to 2013. See Fig. S9a for the factor contribution to the variation from average conditions."

(2) Line 241-243: "Figure 3 shows the contributions of individual factors to the ΔpH across the four seasons. Here the bar plots indicate the factors contributing to the ΔpH between two adjacent seasons, e.g., spring (MAM) to summer (JJA). See Fig. S9b for the factor contribution to the variation from average conditions."

(3) Line 273-275: "Figure 5 shows the effects of individual factors to the ΔpH between day and night. Here the bar plots indicate the factors contributing to the ΔpH between two adjacent hour periods, e.g., 0:00 to 6:00. See Fig. S9c for the factor contribution to the variation from average conditions."

We've revised Fig. 1b, 3 and 5 in the manuscript and added more description in the captions. For example, we've revised Fig. 3 into:

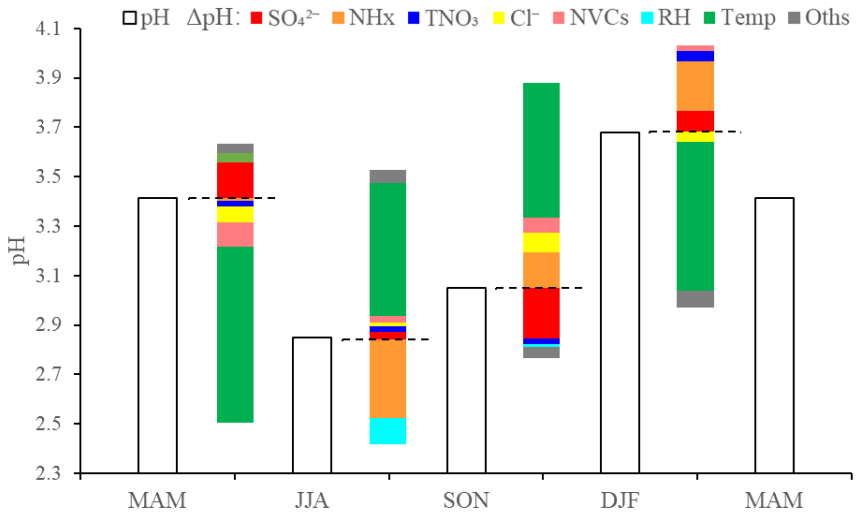

Figure R1 *(revised Fig. 3 in the manuscript)*. **Contributions of individual factors to the ΔpH across the four seasons.** Here the bar plots indicate the factors contributing to the ΔpH between two adjacent seasons, e.g., spring (MAM) to summer (JJA). The meanings of the abbreviations: RH, relative humidity; Temp, temperature; NVCs, non-volatile cations; NH_x, total ammonia; TNO_3, total nitrate.

Changes in* supplement of *manuscript:

Line 111-117: We've added the Figures S9 in the revised supplement as:

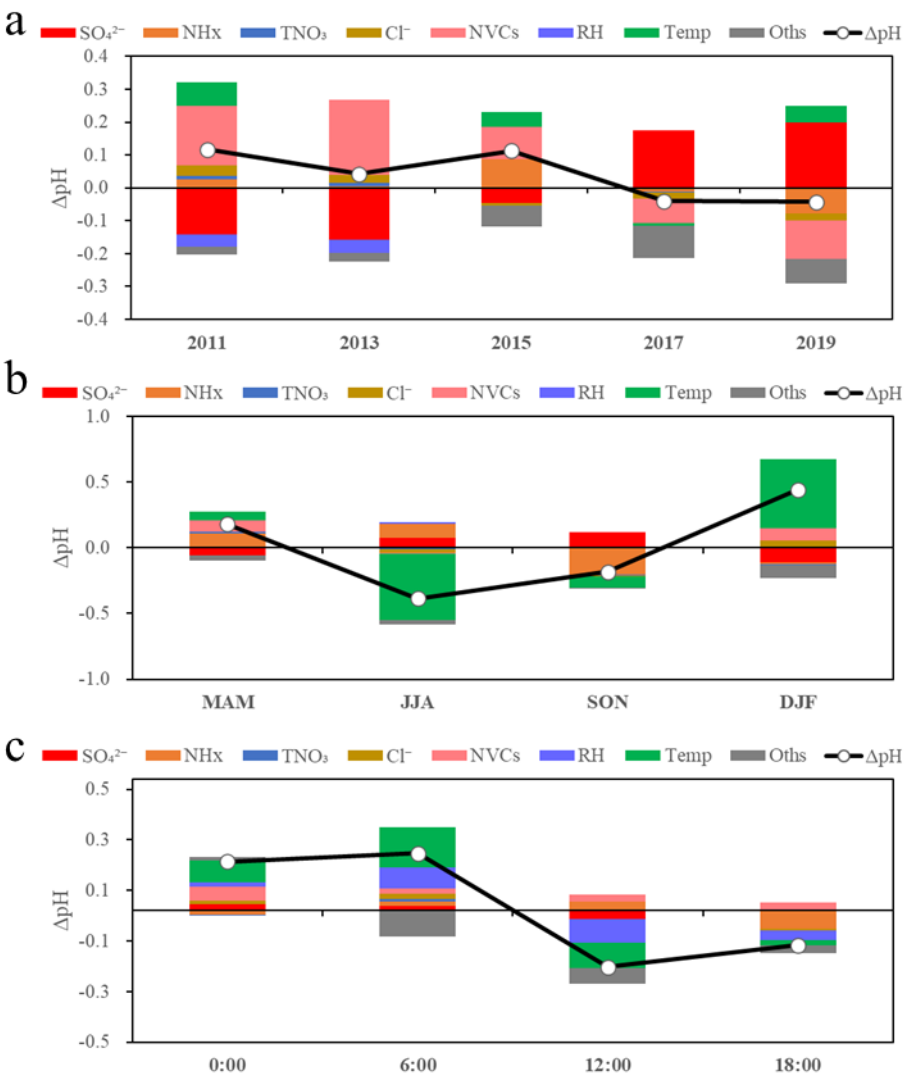

Figure R2 ***(added as Fig. S9 in the revised supplement)*****. Fractional contribution of individual factors to the variations in aerosol pH from average conditions (i.e., averages of all observational data) during 2011–2019.** (a) Annual variation; (b) Seasonal variation, and (c) diurnal variation. The meanings of the abbreviations: RH, relative humidity; Temp, temperature; NVCs, non-volatile cations; NH_x, total ammonia; TNO_3, total nitrate.

3. Discussion about the limited effects of future emissions control measures on haze pollution (e.g., line 35-36, 298-299) is just wrong. Although the partitioning of NH_3 and HNO_3 may shift towards the particulate phase in the future, it does not mean their total PM concentration has increased. If the total concentration (i.e., NH_3 + NH_4^+) decreased enough, then a shift in partitioning towards the particle phase could still occur with a decrease in the aerosol NH_4^+. This discussion would be much better with associated predictions of the $PM_{2.5}$, NH_4^+, SO_4^{2-}, and NO3- aerosol concentrations.

Response: Thanks for the comment. We agree that the precursor decrease will finally lead to a PM decrease. Here we are discussing about the efficiency of PM reduction concentrations against the precursor reduction concentrations. To further clarify our points, we've revised the corresponding manuscript and figures with more detailed explanations. In addition, we've added the prediction of the changes in major chemical components (NH_4^+, SO_4^{2-}, NO_3^- and Cl^-) as Fig. 6g-i following the reviewer's suggestion. See detailed modifications below.

Changes in manuscript:

(1) Line 34-38: We've revised the statement into: "The corresponding aerosol pH in eastern China is estimated to increase by ~0.9, and the reduction in particle phase NO_3^- and NH_4^+ is less than the reduced amount of total HNO_3 and total NH_3. This suggests a reduced benefit of NH_3 and NO_x emission control in mitigating haze pollution in eastern China."

(2) Discussions in section 3.4: We've revised Fig. 6 and the corresponding discussions into (Line 320-366 in the revised section 3.4):

"Under the reference scenario of SSP3-70-BAU with weak control policy (blue lines in Fig. 6 a-f), SO_2 and NOx are predicted to increase, while the NHx is relatively stable. Correspondingly, both SO_4^{2-} and NO_3^- will increase, and NH_4^+ will also increase in response (Fig. 6g). Considering the stable NHx, NH_4^+ partition ratio (NH_4^+ / (NH_4^+ + NH_3)) will increase. In comparison, there is little change in aerosol pH and the predicted NO_3^- partition ratio (NO_3^- / (NO_3^- + HNO_3)).

Under the moderate control policy (SSP2-45-ECP), the emissions of SO_2, NO_x, and NH_3 in 2050 will be reduced by 62.7%, 49.0% and 25.0%, respectively. Correspondingly, SO_4^{2-}, NO_3^- and NH_4^+ will all decrease (Fig. 6h), with a total PM reduction of ~14.4 μg m^{-3}. Moreover, the predicted pH will increase by ~0.5, and the NO_3^- and NH_4^+ partition ratios will decrease by 0.14 and 0.23, respectively (green lines in Fig. 6d-f). That is, more nitrate and ammonium will exist in the gas phase as HNO_3 and NH_3, thus the reduced NH_4^+ and NO_3^- is higher than the reduced NHx and TNO_3, which is a control bonus in terms of reduced PM per reduced emissions for this scenario.

With the strict control policy (SSP1-26-BHE), the emissions of SO_2, NO_x and NH_3 in 2050 will decrease by 86.9%, 74.9% and 41.7%, respectively. Its effect on PM reductions resembles that of the moderate one (SSP2-45-ECP) before 2040. Afterwards, however, the NO_3^- partition ratio increased despite the increasing pH, and reached near 1 in 2050 (Fig. 6 d, e). On second check, we found this pattern is due to the sharp decrease in SO_4^{2-} and constant NVCs. After 2040, there will be a major anion deficit considering the non-volatile species only (sulfate and Ca^{2+}, K^+, Mg^{2+}), and therefore more NO_3^- will be captured by the NVCs to the particle phase. As a result, NO_3^- partition ratio even increased from 0.92 in 2015 to 1.00 in 2050. Although NH_4^+ partition ratio showed a continuous decrease, in 2050 both the reduced NH_4^+ and NO_3^- is smaller than the reduced NHx and TNO_3 (Fig. 6i). That is in contrast with the effect of the moderate one (SSP2-45-ECP). Correspondingly, the total reduced PM is only slightly larger for the strict SSP1-26-BHE policy (~18.6 μg m^{-3}) than the moderate SSP2-45-ECP policy (~14.4 μg m^{-3}) indicating a reduced efficiency in terms of PM controls in responses to the emission controls. This would suggest a reduced benefit of NH_3 and NO_x emission control in mitigating haze pollution in eastern China, especially after 2040."

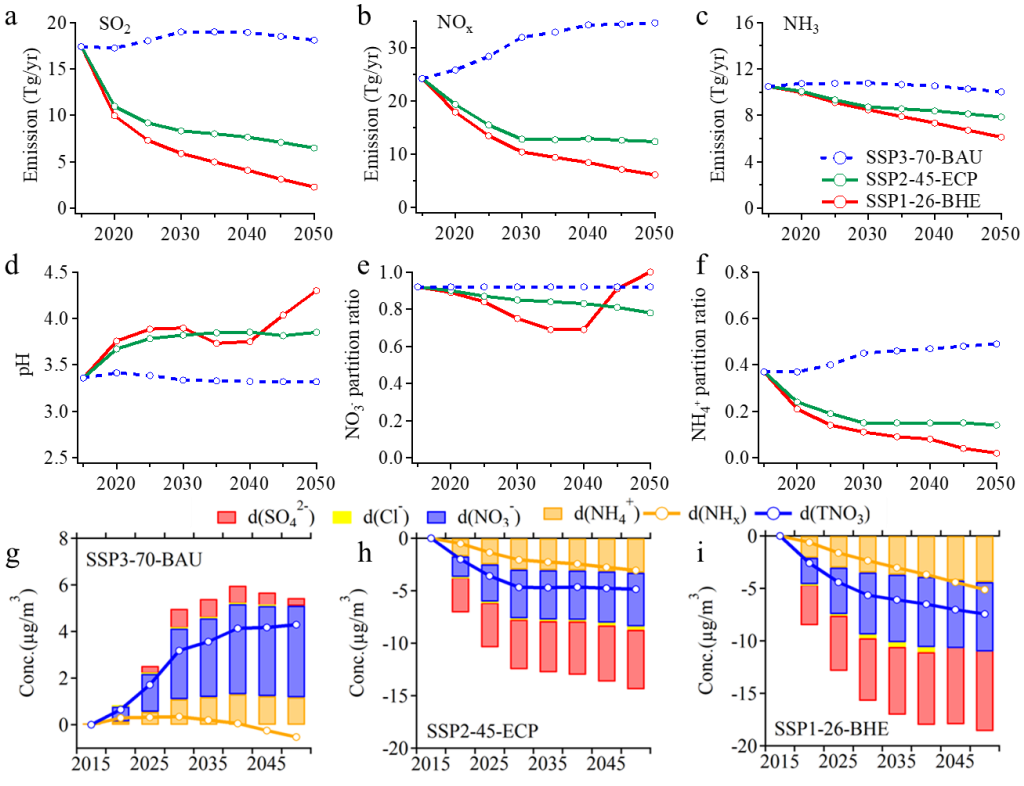

Figure R3 ***(revised Fig. 6 in the manuscript)*****. Emissions of SO_2 (a), NO_x (b), NH_3 (c), predicted pH (d), NO_3^- partition (NO_3^- / (NO_3^- + HNO_3)) (e) and NH_4^+ partition (NH_4^+ / (NH_4^+ + NH_3)) (f) in China from 2015 to 2050 under the three scenarios published in Tong et al.(2020). Predicted the changes in major chemical components (NH_4^+, SO_4^{2-}, NO_3^- and Cl^-) and reductions in TNO_3 and NHx under the three scenarios, including SSP3-70-BAU (g), SSP2-45-ECP (h) and SSP1-26-BHE (i).**

4. The Conclusions section needs substantial revision. A brief recap is ok, but Section 4 is mostly redundant with the prior section. Rather than just reiterating what has already been said, more discussion of the significance of the work is warranted.

Response: Thanks for the comment. We rewrote the conclusions section. Please see the following changes.

Changes in manuscript:

Line 369-415: The specific modifications are as follows:

"The aerosol pH values at an urban site in Shanghai during 2011–2019 were calculated using ISORROPIA II. The trend analysis of aerosol pH in Shanghai during 2011–2019 was reported firstly based on observed gas and aerosol composition. Although significant variations of aerosol compositions were observed from 2011 to 2019 in YRD region, the aerosol pH estimated by model only slightly declined by 0.24 unit. We quantified the contributions from individual factors on the variation of aerosol pH from 2011 to 2019. We revealed that besides the multiphase buffer effect, the opposite effects of SO_4^{2-} and non-volatile cations changes with a contribution of +0.38 and −0.35 unit on aerosol pH, respectively play a key role in determining the moderate pH trend from 2011 to 2019.

Distinct seasonal variations in the aerosol pH were observed, with maximum and minimum aerosol pH of 3.59 ± 0.57 in winter and 2.89 ± 0.49 in summer, respectively. Seasonal variations in aerosol pH

were mainly driven by the temperature, with the max ΔpH of 0.63 existed between fall and winter. The diurnal cycle of particle pH was driven by the combined effects of temperature and relative humidity which could result in ΔpH of -0.22 and +0.10 units, respectively. These results emphasized the importance of meteorological conditions in controlling the seasonal and diurnal variations of aerosol pH.

Finally, to explore the effects of China's future anthropogenic emission control pathways on aerosol pH and compositions, we chose three different emission reduction scenarios proposed by Tong et al.(2020) for future haze mitigation, naming SSP3-70-BAU, SSP2-45-ECP and SSP1-26-BHE as case studies. We estimated that the future trend of aerosol pH and NO_3^- partition ratio will change little under the weak control policy (SSP3-70-BAU), while SO_4^{2-}, NO_3^- and NH_4^+ will increase substantially. The results also demonstrate that future aerosol pH will increase under both strict control policy (SSP1-26-BHE) and moderate control policy (SSP2-45-ECP), but more drastically under former scenario. The significant increase in aerosol pH with the strict control policy will lead to the reduced aerosol NH_4^+ and NO_3^- is smaller than the reduced amount of total NH3 and total HNO_3, which is in contrast with effect of the moderate control policy. This suggests that a reduced efficiency in terms of PM controls in responses to the emission controls with the strict control policy. These results highlight the importance of proportional reductions in precursors and follow-up variations in aerosol pH in future pollution control policy."

5. Finally, the entire manuscript needs to be edited for language consistency – specifically, verb tenses change within and between paragraphs. There are too many instances to list here.

Response: Thanks for the comment. The language consistency in the manuscript has been polished, please see the modifications in the revised manuscript.

Technical/Minor Comments

1. Line 25: define all acronyms on their first use (e.g., NVCs, YRD)
2. Line 28-29: sentence needs grammatical editing.
3. Line 39: suggest deleting "studies"
4. Line 44-45: cite also Tilgner et al. (2021) in this group.
5. Line 73: cite also Vasilakos et al. (2018) and Nenes et al. (2020)
6. Line 77: "composition" should be singular
7. Line 78: suggest changing "characterizing" to "characterize"
8. Line 96: change "to be" to "were"
9. Line 96: "calibration" is not the right term here – LiBr is used as an internal standard
10. Line 102: due to unmeasured species (organic acid ions, carbonate) – it is quite possible to measure the aerosol inorganic composition accurately and not achieve an ion balance. Given what we know about organic acid concentrations, it is actually surprising that such a balance is observed.
11. Line 104: cite also Stieger et al. (2018)
12. Line 106: suggest deleting "techniques"
13. Line 107: give the instrument(s) used to measure T and RH
14. Line 128: cite also Battaglia Jr., et al. (2019)
15. Line 237-238: the diurnal behavior of aerosol pH is not just consistent with Beijing, but is far more consistent (qualitatively) with many other locations like the SE USA (Guo et al., 2015), eastern US (Battaglia et al., 2017), Chicago (Battaglia et al., 2017), which shows the important influences of T and RH on aerosol pH.
16. Line 274: "active actions" should be changed

17. Line 284: comma needed before “respectively”

Response: Thanks for the comments. We’ve revised the manuscript based on the above comments. Please see the following changes.

Changes in manuscript:

(1) Lines 24-26: We added the definition of NVCs and YRD. Please see the details as follows: “The implementation of the Air Pollution Prevention and Control Action Plan led to -35.8%, -37.6%, -9.6%, -81.0% and 1.2% changes of $PM_{2.5}$, SO_4^{2-}, NH_x, non-volatile cations (NVCs) and NO_3^- in Yangtze River Delta (YRD) region during this period.”

(2) Lines 26-30: We rewrote this sentence, please see the details as follows: “Different from the fast changes of aerosol compositions due to the implementation of the Air Pollution Prevention and Control Action Plan, aerosol pH showed a moderate change of -0.24 unit over the 9 years. Besides the multiphase buffer effect, the opposite effects from the changes of SO_4^{2-} and non-volatile cations played key roles in determining the moderate pH trend, contributing to a change of +0.38 and −0.35 unit, respectively”

(3) Line 41: We deleted the word “studies”, please see the details as follows: “Aerosol acidity is an important parameter in atmospheric chemistry.”

(4) Line 44-47: We added the cite of Tilgner et al. (2021) as: “Aerosol acidity has attracted an increasing concern in recent years because of its impacts on the thermodynamics of gas-particle partitioning, pH-dependent condensed-phase reactions and trace metal solubility(Cheng et al., 2016; Fang et al., 2017; Guo et al., 2017; Guo et al., 2016; He et al., 2018; Song et al., 2018; Su et al., 2020; Tilgner et al., 2021; Weber et al., 2016).”

(5) Line 73-76: We added the cites of Vasilakos et al. (2018) and Nenes et al. (2020), please see the details as follows: “Aerosol pH may change due to the significant changes of the chemical composition in $PM_{2.5}$, which may feedback to the multiphase formation pathways of aerosols such as sulfate, nitrate and ammonium (Cheng et al., 2016; Nenes et al., 2020; Vasilakos et al., 2018)”

(6) Line 78-80: We edited this sentence as: “A thermodynamic model, ISORROPIA II (version 2.1) (Fountoukis and Nenes, 2007) was applied to estimate the pH based on 9-year continuous online measurements of $PM_{2.5}$ composition at an urban site in Shanghai.”

(7) Line 80-81: We edited this sentence, please see the details as follows: “The main purposes of this study are to: (1) characterize the long-term trend of aerosol pH;”

(8) Line 98-99: We edited this sentence into: “To better track the retention time changes of different ion species and ensure their concentrations were measured successfully,”

(9) Line 98-100: We rewrote this sentence into: “To better track the retention time changes of different ion species and ensure their concentrations were measured successfully, an internal standard check was conducted every hour with Lithium Bromide (LiBr) standard solution (Qiao et al., 2014; Zhou et al., 2016).”

(10) Line 105-108: Indeed, the measurements of organic acid ions were lacked in our study. However, we find that in previous studies, the concentrations of organic acid in Shanghai area were low. Ding et al., (2021) found that total dicarboxylic acids in Chongming Island in Shanghai during the day and night was 375±282 ng/m^3 and 341±270 ng/m^3, respectively, and the ketocarboxylic acids ranged from 3.3 ng/m^3 to 125 ng/m^3. Yao et al., (2002) also found that the sum of oxalate, malonate and succinate only account for 0.3-2% of the total mass of the water-soluble ions in Shanghai. The concentrations of organic ions were significantly lower than that of SO_4^{2-} and NO_3-, which were the main anions in aerosol of Shanghai. Due to the low concentrations of organic acids in $PM_{2.5}$, they

may have minor effects on ion balance. Meanwhile, we also find that the average equivalent ratios of cation/anion(C/A) were close to unity in many cities of China (Huang et al., 2014; Shen et al., 2010; Sun et al., 2006; Zhang et al., 2018). In our study, the modelled and measured NH_3 and NH_4^+ concentrations were in good agreement based on observed aerosol composition, further indicating that the measurement of the ions was accurate. We rewrote this sentence as: "The correlation between cation and anion was strong (R^2=0.94), with a slope of 1.00, indicating that these ion species were charge balanced and well represented major components in $PM_{2.5}$."

(11) Line 108-110: We added the cites of Stieger et al. (2018) as: "In previous studies, intercomparison experiments between MARGA and filter-based method have been carried out, and the data measured by MARGA showed acceptable accuracy and precision (Rumsey et al., 2014; Huang et al., 2014; Stieger et al., 2018)"

(12) Line 110-113: We edited this sentence as: "The mass concentrations of $PM_{2.5}$ were simultaneously measured using an on-line beta attenuation PM monitor (FH 62 C14 series, Thermo Fisher Scientific) at a time resolution of 5 min."

(13) Line 112-114: We added the instrument information used to measure T and RH, please see the details as follows: "The temperature and RH were also measured using meteorological parameters monitor (Metone 597, Met One Instruments) at a time resolution of 1 min."

(14) Line 135-137: We added the cite of Battaglia Jr., et al. (2019) in this sentence as: "The use of $ALWC_i$ to predict pH is therefore fairly accurate and common(Battaglia Jr et al., 2019; Battaglia et al., 2017; Ding et al., 2019)"

(15) Line 270-272: We rewrote this sentence into: "Figure 4 shows the diurnal variations in the aerosol pH and its potential drivers. Aerosol pH in Shanghai exhibits notable diurnal variations, being higher during nighttime.

(16) Line 314-315: We rewrote this sentence into: "SSP3-70-BAU is a reference scenario that without additional efforts to constrain emissions."

(17) Line 330-333: We edited this sentence into: "Moreover, the predicted pH will increase by ~0.5, and the NO_3^- and NH_4^+ partition ratios will decrease by 0.14 and 0.23, respectively (green lines in Fig. 6d-f)."

Reference

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