Dear Prof. Tong Zhu and referees:

Thank you very much for your thoughtful and constructive comments on this manuscript. We have revised the manuscript accordingly. Detailed responses are given below point by point (in blue).

Response to Referee #1

Based on the one-year long of field measurement of inorganic ions and ammonia, this paper mainly reports the seasonal variations in concentrations and compositions of fine particulate inorganic ions, and the aerosol acidity in Wuhan, China. The aerosol acidity, i.e., pH of the $PM_{2.5}$, is the subject of this work, but I found many statements of this paper on the aerosol pH are not correct and main conclusions given by the authors do not stand up. Moreover, this field observation work lacks of QA/QC, making the data unreasonable. For an online semi-continuous measurement using a technique like MARGA in this work, a simultaneous collection of filter samples should be done to compare and correct the online results, which is an indispensible of QA/QC, because the online data are of significant uncertainty, as reported by many researchers. However, in the current work there is no any comparison by using filter collection or other measurement, and the data quality is poor (see my detailed comments below). Concentrations of inorganic ions are the main and the key data of this work, which are the basis of the conclusions given by the author, but I found the inorganic ion concentrations are too high and in many cases they exceeded the $PM_{2.5}$ mass, which are unreasonable (see the details below). Some explanations in this paper are subjective or self-contradictory (as exemplified below). In addition, there are many English grammar errors in this manuscript.

Many thanks for your valuable comments and suggestions, which help us to improve the manuscript. We conducted QA/QC for the whole running period of MARGA substantially. Sorry for that we forgot to describe QA/QC in the primary manuscript. It has been added in the text.

We quite agree that the data from online semi-continuous measurement using a technique like MARGA is of significant uncertainty. It should also be noted that the data validity and consistency of MARGA have been widely reported in many previous studies (Cowen et al., 2011; Rumsey et al., 2014; Zhang et al., 2017). Many researches have already used the data from MARGA alone to study the air pollution around the world (Khezri et al., 2013; Li et al., 2013; Liu et al., 2017; Li et al., 2019).

The sampling site is a supersite for air quality monitoring and forecasting set by local government. Therefore, a professional team for maintenance of MARGA and other online instruments was employed. Detailed QA and QC were strictly conducted. In this research, this MARGA started to work from June

2013. Till now, the engineer has been continuously paying efforts on its operation and maintenance. Internal and external calibration was persistently conducted. Internal calibration for each sample testing in 1-hour resolution was achieved by LiBr, and the relative standard deviation (RSD) was controlled within 10%. Each month the external calibration was conducted by manual sampling of mixed anionic standard solution (Cl^-, Br^-, SO_4^{2-} and NO_3^-) and mixed cationic standard solution (Li^+, K^+, Ca^{2+}, Na^+, Mg^{2+} and NH_4^+), with RSD controlled less than 5%. Simultaneously, the sampling flow was checked and calibrated monthly with RSD controlled less than 5%. In addition, accuracy, precision and reproducibility tests were annually performed by standard solution with different concentration gradients.

We have to accept the fact that the total inorganic ion concentrations with 1-h resolution occasionally exceeded the $PM_{2.5}$ mass. It was incorrect in theory definitely, while it was inevitable for different detecting principles of online equipments and corresponding deviations. We are regretful that there was no comparison between the online data with those obtained from filter collection and analysis. Then we compared the data with those from another MARGA set in Wuhan as following Table S1 shown. Both the ionic concentrations and $PM_{2.5}$ mass were comparable for the two dataset.

Meanwhile, from the response to the high proportion of ions in $PM_{2.5}$ below, the total carbon contents in Wuhan were low. We have also checked the papers published about the data obtained from MARGA. The averaged total ions and carbonaceous aerosols accounted for 56.2% and 14.5% of $PM_{2.5}$, respectively in Shanghai was also observed. (Li et al., 2019).

Therefore, we believed that the data used here can be accepted. At the same time, the shortages of online semi-continuous measurement and the possible reasons for the higher proportion of ions were also emphasized in the text (Line 209-213).

Table S1 Comparison of annual averaged concentrations of main ions obtained by two MARGA equipments in Wuhan.

Site	SO_4^{2-} (μg m^{-3})			NO_3^- (μg m^{-3})			NH_4^+ (μg m^{-3})			$PM_{2.5}$ (μg m^{-3})		
	2014	2015	2016	2014	2015	2016	2014	2015	2016	2014	2015	2016
This study	13.7	15.4	13.0	12.7	14.7	12.4	10.2	10.7	9.4	71	68	56
Hankou[a]	/	13.0	11.9	/	12.8	11.1	/	10.6	8.1	/	/	52

[a] Another unban environmental monitoring site in Wuhan, about 20 km west of the site in this study.

Line 33-35, why aerosol pH showed a different trend at the lower AWC from that at the AWC higher than 15 ug/m^{-3}?

Thanks for this query. We have a deep thinking and corrected it as following. Detailed discussion could be found in the text (Line 290 - 316).

Figure 6a exhibited the relationship between pH and AWC. From the fitting curve, the particle pH firstly showed a decreasing trend with AWC increased under much lower AWC level (less than about 15 μg m^{-3}). Then, with the AWC increasing from ~15 to ~380 μg m^{-3}, its dilution effect on aerosol acidity gradually dominated, which offset or even surpass the role of aqueous-phase reaction and formation of secondary inorganic ions, leading to the quick increase of aerosol pH. The SNA contents exhibited peak values when the AWC increased to about 380 μg m^{-3}, reflecting intense aqueous-phase reaction and formation of SNA, resulting in buffering effect on pH increase. That might be the reason for the pH slowly growth when AWC was at ~380 μg m^{-3}. Then with the AWC further increasing, SNA contents decreased (Figure 6d). When AWC is higher than about 380 μg m^{-3}, along with the increasing of AWC, a slowly increasing and gradually no significant growth of pH was observed.

The phenomenon was favored by the SNA-RH correlation coefficient variation with RH increasing (Figure 6b) and relationship of AWC-RH (Figure 6c), SNA-AWC (Figure 6d) and pH-RH (Figure 6e). Lower RH was corresponded to lower AWC (Figure 6c). It can be speculated that the pH decreasing trend at lower RH or AWC was might due to the increasing of H_2SO_4 and other acidic material for heterogeneous reactions. Simultaneously, under lower RH (< ~0.48) and AWC (< ~15 μg m^{-3}) conditions, the AWC dilution effect on aerosol acidity was negligible, which can be verified from the relationship of AWC/($PM_{2.5}$+AWC) and RH (Figure 6c). When RH was in 0.25- 0.48, AWC/($PM_{2.5}$+AWC) was only 0.08-0.16. Then with the further increasing of AWC or RH, the aerosol pH increasing trend may be dominantly controlled by the rapid increasing of AWC. It was also verified by the weaker or even negative correlation coefficient of SNA with RH when RH varied in 0.48-0.75. Along with RH gradually increased to the deliquescence relative humidity (DRH) of various inorganic salts successively, the particles' absorbing water sharply increased and produced a saturated aqueous solution (Seinfeld and Pandis, 2006). When RH was higher than 0.95, with the corresponding AWC higher than 380 μg m^{-3}, obviously negative correlation between RH and SNA was obtained ($R^2 = 0.59$, $p < 0.05$). It implied the dominated role of buffering effect of AWC, which lead to the continuously increasing and gradually slowdown trend of pH.

Line 183-193, and Table1, inorganic ions account annually for 70+/-19% of $PM_{2.5}$, I think this ratio is too high, and unrealistic, because $PM_{2.5}$ not only consist of inorganic ions, but also consist of organic matter, black carbon and crustal materials. Probably the $PM_{2.5}$ mass was underestimated. Did author collect filter samples to validate the online MARGA data? In this study there was no work on QA/QC.

Thanks for your query.

$PM_{2.5}$ concentration was not underestimated and could be verified by the data from other national monitoring sites. From September 2015 to August 2016, the mean concentration of $PM_{2.5}$ from the ten national control sites in Wuhan was 61 μg m^{-3}. In this study, by adopting the suggestion from reviewers, the data for 106 rainy days were excluded, and data in February was also not adopted as the missing of corresponding ionic concentration data due to the MARGA maintenance. Then the $PM_{2.5}$ mean value of 63.4 μg m^{-3} in this study was obtained and it is comparable with the averaged value from the ten national control sites. Meanwhile, this MARGA equipment was installed from June 2013, the proportions of total WSI in $PM_{2.5}$ (including SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+}) for these years were almost uniform (Table S2).

Table S2 Proportions of total WSI in $PM_{2.5}$ from 2013 to 2016 at the sampling site in this study

Times	$PM_{2.5}$ (μg m^{-3})	WSI (μg m^{-3})	WSI/$PM_{2.5}$
2013.6.1-12.31	78.6	53.1	0.68
2014.6.1-12.31	70	32.6	0.47
2015.6.1-12.31	56	34.5	0.62
2016.6.1-12.31	47	28.1	0.60
2014.1.1-6.30	84	50.0	0.60
2015.1.1-6.30	78	56.0	0.72
2016.1.1-6.30	63	47.2	0.75
2014.1.1-12.31	71	41.6	0.59
2015.1.1-12.31	68	45.3	0.67
2016.1.1-12.31	56	37.7	0.67

We conducted observation of organic carbon (OC) and element carbon (EC) (Sunset RT-4) in 1-hour resolution at the same site from January 1 to April 21 in 2016 (as Figure S1 shown). The average TC (total carbon = OC + EC) concentration was 6.01 μg m^{-3}, and the averaged concentrations for the same periods of $PM_{2.5}$ and WSI were 84.2 and 59.4 μg m^{-3}, respectively. It could be found that the averaged TC and WSI accounted for 7.1% and 70.5% of $PM_{2.5}$, respectively. Therefore, the total WSI accounting for 70% of $PM_{2.5}$ from September 2015 to August 2016 in this manuscript was reasonable.

In fact, we could also find relative high ionic proportions in $PM_{2.5}$ from MARGA data (Li et al., 2019). The averaged total ions and carbonaceous aerosols accounted for 56.2% and 14.5% of $PM_{2.5}$, respectively in Shanghai.

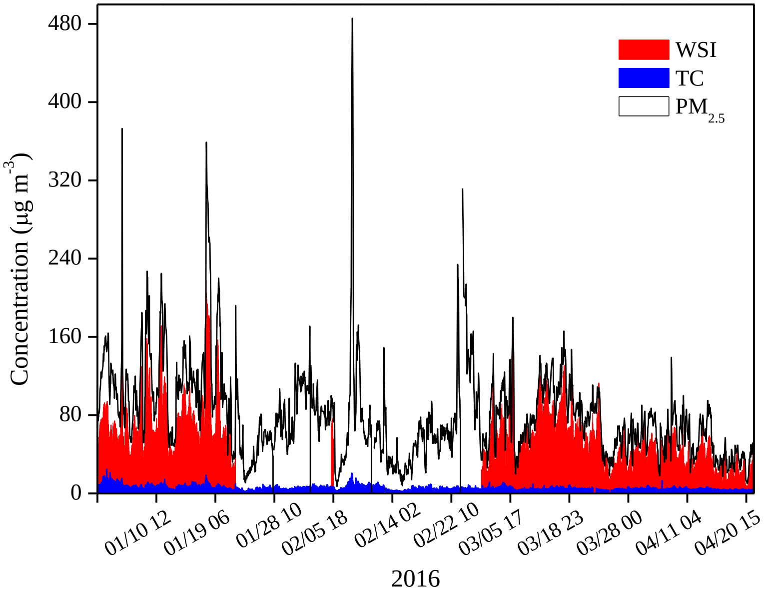

Figure S1 Time series of total carbon, water-soluble ions and $PM_{2.5}$ in January 1 to April 21 of 2016

Line 196-198, these statements are not correct. Higher temperature in summer favors more ammonium nitrate aerosol decompose and evaporate into the gas phase as NH_3 and HNO_3, because NH_4NO_3 is volatile, thus resulting in the lower concentration of NH_4NO_3 aerosols. This is the reason why the calculated NOR is lower in summer. However, this does not mean that nitrate and ammonium formation rates are lower in summer.

Many thanks for your suggestion. Corresponding corrections have been done (now Line 221-225).

Line 200-207, summer is rainy season for Wuhan, thus the lower SNA is also due to the stronger precipitation.

The manuscript had deleted all the date for rainy days according to the previous reviewer's comments before ACPD, so no data about rainy days were mentioned in this paper.

Thank you very much for this comment. Although the data in rainy day has been deleted, the cleaning effect is also one of the reasons for the lower SNA in summer. Corresponding corrections have been done in the revised manuscript (Line 223-225).

Line 214-217, the strong correlation between SO_4^{2-} and NH_4^+ does not necessarily means that $(NH_4)_2SO_4$ is the form of ammonium, in fact, many studies have pointed out that NH_4HSO_4 is the major chemical form rather than $(NH_4)_2SO_4$. Moreover, this statement is inconsistent with the main conclusion of this work. Because as reported by Liu et al. (Liu et al., 2017; Son et al., 2018). The pH of $(NH_4)_2SO_4$ aerosol is more than 4, while the pH of $PM_{2.5}$ in Wuhan estimated by the current work is very acidic, less than 3.7, thus NH_4HSO_4 seems more consistent with the aerosol pH values given by the authors.

We really appreciate this suggestion.

In this study, HSO_4^- really existed in liquid aerosol from the output of ISSORROPIA-II, while its concentration was much lower than SO_4^{2-}, with the mean concentrations of 0.005 μg m^{-3} and 13.76 μg m^{-3} for HSO_4^- and SO_4^{2-}, respectively.

In literatures, the ratio of NH_4^+ to SO_4^{2-} was often used to estimate the existing form of $(NH_4)_2SO_4$ or NH_4HSO_4. Khlystov et al. (2005) reported that the NH_4^+/SO_4^{2-} ratio of 1.4 was corresponded to a proximately equal mixture of $(NH_4)_2SO_4$ and NH_4HSO_4, and all the sulfate was neutralized by ammonium when the NH_4^+/SO_4^{2-} ratio was higher than 2. Rees et al. (2004) also defined that NH_4^+/SO_4^{2-} ratio < 1 indicated the present of bisulfate, and ratio of 0.5 or less implied the absence form of sulfate but completely form of bisulfate.

In this study, from Figure 7a in the manuscript, the molar ratio of NH_4^+/SO_4^{2-} in Wuhan mainly varied from 2:1 to 6:1, indicating that SO_4^{2-} was mainly presented in the form of $(NH_4)_2SO_4$. Definitely, a few data held NH_4^+/SO_4^{2-} ratio lower than 2:1, or even lower to 1.3, which indicated the existing of NH_4HSO_4, but it should not be the dominant form.

Other study indicated that when NH_4^+/SO_4^{2-} ratio was about 1.9 - 2, there were negligible HSO_4^- signals (Chan and Chan, 2013). From literatures letovicite may also exist (Pathak et al., 2004), but it can be negligible. Furthermore, from the pH and acid-base titration curves of sulfuric acid (Figure S2, draw by CurTiPot, http://www.iq.usp.br/gutz/Curtipot.html), it can be seen that sulfate and bisulfate were co-existed when pH was lower than 4, and all the sulfuric acid was in the form of sulfate when pH continuously increasing. Aerosols in Wuhan were moderate acidic, with pH averaged as 3.30 ± 0.49, at the point that SO_4^{2-} was the dominant form. Thus, we insist that $(NH_4)_2SO_4$ is the main form rather than NH_4HSO_4 in this study.

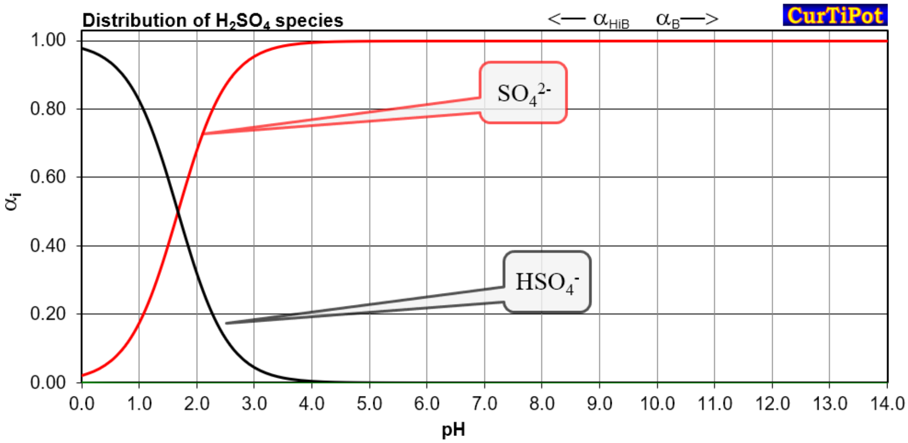

Figure S2 pH and acid-base titration curves of sulfuric acid

(Note: the "X" curve might shift slightly to the left or right with the ions strength changing)

Line 246-248, as I mentioned above, under such a lower pH=2.84 conditions, ammonium in $PM_{2.5}$ cannot be exist as $(NH_4)_2SO_4$.

Thanks for your query.

In Wuhan, the lowest pH value occurred in summer, averaged as about 2.84. As mentioned above when pH = 2.84, SO_4^{2-} and HSO_4^- coexisted and SO_4^{2-} was dominant.

Line 299-302, this statement is not correct. In fact, AWC is not only dependent on RH but also dependent on the mass of hygroscopic matter. Inorganic ions such as ammonium sulfate and ammonium nitrate are much less in summer, thus AWC are the lowest in July, August and September.

Thanks for this comment and suggestion.

The lower AWC in summer was indeed closely related with the lower inorganic ion concentrations such as ammonium sulfate and ammonium nitrate and we corrected it in the manuscript (Line 320-323).

However, we also believed that the high temperature in summer is one of the reasons for the lower AWC. Seinfeld and Pandis (2006) indicated that the atmosphere water vapor exponentially decreases with temperature. From the relationship of Figure 6(f) in the manuscript, the AWC significantly decreased when the temperature was higher than about 303 K (about 30 ℃). In addition, from the following Table S3, the similar SNA/$PM_{2.5}$ was not always equal to similar AWC. RH was important factor to determine AWC, and RH may also be affected by temperature in Wuhan, considering the abundant watershed area of this region.

Table S3 RH, SNA/$PM_{2.5}$ and AWC contents under different ambient temperature in Wuhan

Temperature (T)	T (average)	RH	SNA/$PM_{2.5}$	AWC ($\mu g\ m^{-3}$)
T>303K	306.5 K	0.67	0.59	18.9
293K<T<303K	298.2 K	0.69	0.58	45.3
283K<T<293K	289.4 K	0.74	0.61	98.1

Response to Referee #2

I have to endure to read the manuscript thoroughly because of too many wrong or confusing statements, as pointed by another Reviewer. I only listed a few of them.

We really appreciate your patience for reading this manuscript. Thanks for your valuable comments and suggestions. We have thoroughly corrected the manuscript by adopting the suggestions from the reviewers and re-read more references.

Meanwhile, all the authors have polished the paper for another time carefully.

Line 23, “for different time scales”, you have told the resolution is 1-h, the mean values does not mean another scale.

Thanks for your suggestion and we have revised it (Line 23-25).

Line 25-26, it is difficult for the readers to conclude the seasonal pH values from your monthly results.

We have corrected the description (Line 27-28). We have calculated the seasonal pH in Section 3.2 (Line 263-276).

Line 29-31. I do not know why high RH could lead excess ammonium? What is environmental conditions?

Thanks for this query.

Sorry for the unclear description, and we have revised it (Line 32-34). Primarily, we used the "environmental conditions" here to indicate the specific geographic position and air pollution situation in Wuhan.

Now the sentences have been corrected as "The higher RH, higher excess ammonium (averaged as 6.06 ± 4.51 μg m^{-3}) and abundant aerosol water content (AWC, averaged as 71.0 ± 82.8 μg m^{-3}) can explain the lower $PM_{2.5}$ acidity in Wuhan than other megacities of China."

Line 36-37, "With atmospheric RH increasing, the aerosol pH exhibited decreasing trend firstly and then increased, with the turning point RH as about 0.48", why? We need explanations in the abstract.

Thanks for this comment. It was similar with the query of Referee #1.

We have corrected it (Line 34-43)

Detailed explanations were shown in Line 290-316.

Line 41, "reduce the aerosol pH" and "further mitigate air pollution", they are conflicting, as stated in the text.

Thanks for this query.

There was no contradiction for "reduce the aerosol pH" and "further mitigate air pollution". Wang et al. (2016) indicated that the increase of pH value will promote the solubility of precursor gases and increase the aqueous reaction rates, which will accelerate the formation of secondary inorganic ions and aggravate air pollution, resulting in more severe haze.

Line 42-43, why the authors insisted Wuhan is a place having unique high RH, what about the coastal cities in southern China? I do not think this statement is enough to support the originality for an ACP paper.

Thanks for this query.

Initially, we thought that Wuhan located in the regions which can be easily impacted by the air pollutants transported from North China Plain and the relative humidity and ambient temperature were both higher than the northern Chinese cities, considering its specific geography, water coverage area and air pollutant levels (as Figure 1 shown). Therefore, we believed that the aerosol acidity in Wuhan may be quite different from those in northern Chinese cities, where most aerosol acidity researches were conducted recently. Then we used this title to highlight the possible differences.

We re-think your suggestion and collected the atmospheric annual relative humidity from 2010-2016 for the major provincial capitals and coastal cities in China from National Meteorological Information Center (http://data.cma.cn/site/index.html). In the following Figure S3, the RH in Wuhan was indeed higher in the major cities of China, averaged as 0.79 (78.86%), and exceeded the values for many eastern and southern coastal cities, such as Guangzhou, Shenzhen, Zhuhai, Xiamen, Fuzhou, Shanghai, Dalian and Qingdao, etc. Therefore, we believed that the highlight and the title could be accepted.

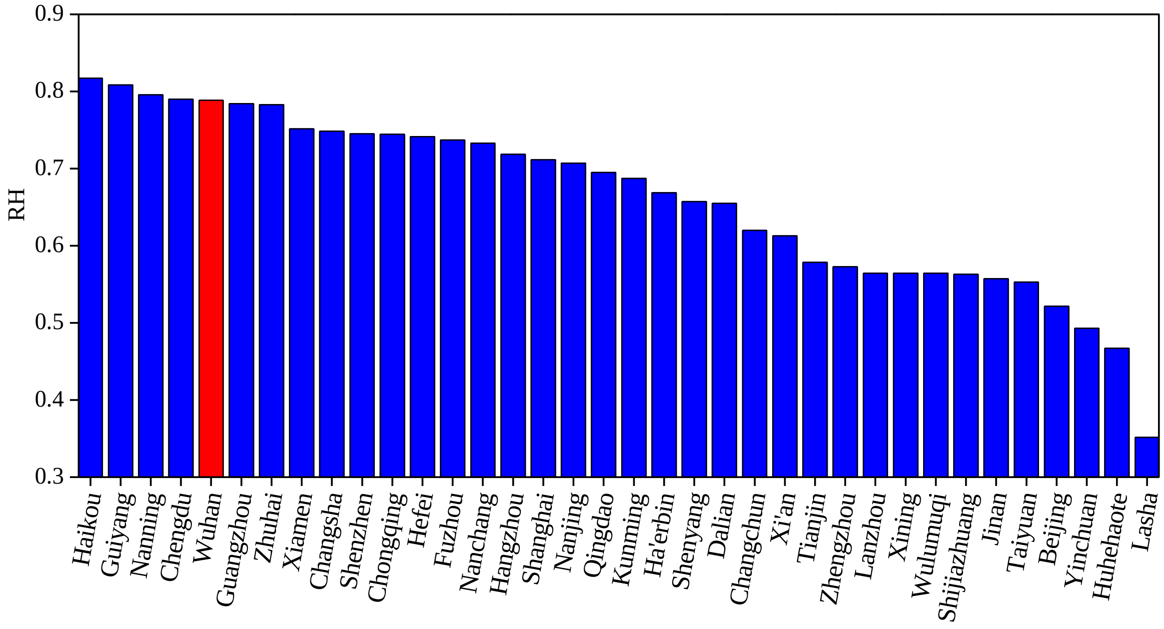

Figure S3 Averaged RH for major Chinese cities for the year of 2010-2016

(Note: 1. Chengdu was referred to Wenjiang site; Chongqing was referred to Shapingba site; Changsha was referred to Wangcheng site. 2. The data for Shenzhen, Qingdao and Dalian were averaged for the year of 2010-2013.)

Line53, only "Acidic aerosols have the capability to reduce atmospheric visibility", the black carbon and organics cannot do so?

Thanks for your query.

Yes, they can. In this manuscript, we just discussed the aerosol acidity and ions. Sorry for this unclear description. We have corrected it (Line 58).

Line58, "The aerosol acidity exhibited spatiotemporal discrepancy, owing to the diversities of source emission and meteorological conditions." I am confused. Which source, which conditions?

Sorry for this unclear description. We have corrected the description (Line 63-64).

Line 68, "Even in the same city, the aerosol acidity was different." It is true? What you mean?

Sorry for this unclear description. We have corrected the description (Line 73-74).

Line 71, "One of the key factors for these diversities is that these studies were done at a given period with different pollution levels." Definitely, it is not correct, the authors should read the papers in detail.

We have re-read the papers and have corrected the description (Line 77-78).

Line 72-76, since Liu et al...., all of them are nonsense. The authors really do not understand the importance of aerosol pH, and what factors would affect the pH values. I strongly suggest they should read papers in depth.

This section has been corrected (Line 78-81).

Line 78-79, I disagree "It may limit the understanding of how the aerosol acidity affects the atmospheric chemistry." Not always is it necessary to measure the ambient air in a whole year for the research.

Sorry for this arbitrary description. We have deleted it and corrected relative sentences (Line 80-81; Line 148-150).

Previous researches (Liu et al., 2017; Wang et al., 2018) of aerosol acidity during a pollution event was definitely significative for atmospheric chemistry. And our study about aerosol acidity firstly in Central China from different angles with a whole year continuous observation was also helpful to understand the relationship of chemical composition-aerosol acidity-air pollution and to enrich the knowledge about formation mechanism of serious air pollution out of North China Plain

Line 81, "always" is not correct, many studies pointed it is improper.

We have corrected the description (Line 82-83).

Line 90, “AWC serves as a medium for aqueous phase reaction of SO_2 oxidation, which can also lead to the increase of aerosol acidity”. I do not know how to lead to the increase of aerosol acidity.

Thanks for this query. Sorry for this unclear description.

We have corrected the description (Line 93-95). “Which” does not mean “AWC”. It actually means that the aqueous oxidation of SO_2 would lead to the formation of SO_4^{2-} and then increase the aerosol acidity.

Generally, When pH was 2-7 in aqueous, SO_2 was mainly in the form of $SO_2 \cdot H_2O$ and can be oxidized to form sulfate and simultaneously release H^+.

Line 92, it is too simple to explain the relations between AWC and pH.

We have corrected it (Line 91-105). The further discussion has been shown in Section 3.3 (Line 290-316).

Line 99-101, it is not a scientific question.

We have corrected it. The scientific question has been corrected as following:

Till now, previous studies were usually conducted in a limited season or pollution episodes (Zhou et al., 2012; Bian et al., 2014; Guo et al., 2015; Cheng et al., 2016; Wang et al., 2016a; Liu et al., 2017; Wang et al., 2018), which can only cover a specific variation ranges of meteorological parameters and levels/mass percentages of ionic species. The complex relationship between ambient temperature, RH, AWC, ionic species concentrations/percentages and aerosol acidity deserved a thorough investigation. However, to our knowledge, this kind of study is still limited. (Line 105-111)

For the regions with abundant agricultural activities which are the main emission sources for ammonia, the relationship of ammonia, aerosol acidity and air pollution needs a clear illustration. (Line 122-124)

Line 106, who neutralizes acid? How ammonium neutralizes acid?

We have corrected it (Line 112-113).

Line 140, why the results in this MS are urgently for the air pollution formation? Many studies have shown it.

Sorry for this arbitrary description. We have corrected it (Line 148-150).

Line 183, I do not think “70%” is correct, the high values is not reasonable, please compare the previous studies.

Thanks for this query. It is similar with the comment by Referee #1.

$PM_{2.5}$ concentration was not underestimated and could be verified by the data from other national monitoring sites. From September 2015 to August 2016, the mean concentration of $PM_{2.5}$ from the ten national control sites in Wuhan was 61 μg m^{-3}. In this study, by adopting the suggestion from reviewers, the data for 106 rainy days were excluded, and data in February was also not adopted as the missing of corresponding ionic concentration data due to the MARGA maintenance. Then the $PM_{2.5}$ mean value of 63.4 μg m^{-3} in this study was obtained and it is comparable with the averaged value from the ten national control sites. Meanwhile, this MARGA equipment was installed from June 2013, the proportions of total WSI in $PM_{2.5}$ (including SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+}) for these years were almost uniform (Table S2).

Table S2 Proportions of total WSI in $PM_{2.5}$ from 2013 to 2016 at the sampling site in this study

Times	$PM_{2.5}$ (μg m^{-3})	WSI (μg m^{-3})	WSI/$PM_{2.5}$
2013.6.1-12.31	78.6	53.1	0.68
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We conducted observation of organic carbon (OC) and element carbon (EC) (Sunset RT-4) in 1-hour resolution at the same site from January 1 to April 21 in 2016 (as Figure S1 shown). The average TC (total carbon = OC + EC) concentration was 6.01 μg m^{-3}, and the averaged concentrations for the same periods of $PM_{2.5}$ and WSI were 84.2 and 59.4 μg m^{-3}, respectively. It could be found that the averaged TC and WSI accounted for 7.1% and 70.5% of $PM_{2.5}$, respectively. Therefore, the total WSI accounting for 70% of $PM_{2.5}$ from September 2015 to August 2016 in this manuscript was reasonable.

In fact, we could also find relative high ionic proportions in $PM_{2.5}$ from MARGA data (Figure S2) (Li et al., 2019). The averaged total ions and carbonaceous aerosols accounted for 56.2% and 14.5% of $PM_{2.5}$, respectively in Shanghai (Figure S2).

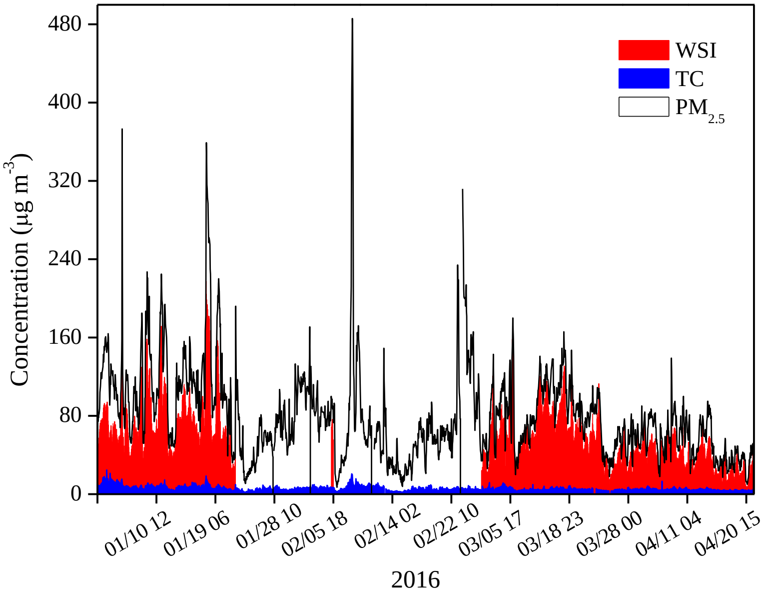

Figure S1 Time series of total carbon, water-soluble ions and $PM_{2.5}$ in January 1 to April 21 of 2016

Line 189-190, what is the "aqueous phase reactions"?

We have corrected it (Line 207-208).

Line 213-214, ".. in summer, ammonium preferred to exist in the form of gas phase due to the thermal equilibrium", the ammonium could be gaseous?

Sorry for this incorrect expression.

We have corrected it (Line 221-225 and Line 229-231).

Reference:

Chan, L. P., and Chan, C. K.: Role of the aerosol phase state in ammonia/amines exchange reactions, Environ. Sci. Technol., 47, 5755-5762, doi: 10.1021/es4004685, 2013.

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