Response to comments of anonymous referees # 1

General comments

Wang et al. reported their field observation of chemical composition in particulate matter (PM) in Beijing. Based on the data together with measured meteorological parameters, they estimated condensational loss rate of nitrate precursor gases for nitrate formation and aerosol liquid water. In addition, they discussed the interplay between increased particulate nitrate and aerosol particle liquid water, suggesting that particulate nitrate increased the aerosol liquid water and the aerosol liquid water promoted condensational loss of nitrate precursor gases (N_2O_5 and HNO_3), i.e., increasing particulate nitrate. This study has highlighted an increasing importance of particulate nitrate during a polluted episode in wintertime in Beijing, and would be an important contribution to the understanding of recent change in chemical composition of PM. I would recommend this manuscript for publication in ACP after addressing the following issues.

Many thanks to the reviewer for the comments and suggestions. We have improved the manuscript accordingly. Please find a point-by-point response below.

Specific comments:

Q1. Lines 85-86: The statement on “via well chemical pathways” is a bit misleading for the oxidation of SO_2 because sulfate production mechanisms from the SO_2 oxidation during haze episodes in Beijing are still ongoing debate (Cheng et al., 2016; Gen et al., 2019).

Cheng et al., Science Advances 2.12 (2016): e1601530.

Gen et al., Environmental Science & technology, (2019): 53, 8757-8766.

Thanks for the useful suggestion. The reviewer is right that the particulate sulphate formation is still ongoing debate, not well-established yet.

The original sentence has been revised as shown below.

"In atmospheric environments influenced by anthropogenic activities, particulate secondary inorganic compounds are often dominated by particulate sulfate and nitrate (Heintzenberg, 1989), which originate from the oxidation of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) via multiple chemical pathways (Calvert et al., 1985;Cheng et al., 2016;Wang et al., 2016;Gen et al., 2019a, b)."

Q2. I can understand that nitrate mass fraction in secondary inorganic components has increased. However, I am not sure why the absolute mass concentration of nitrate can increase despite the reduction of NO_x over North China. More discussion on this point deserves to appear in the introduction.

Thanks for your helpful comment. The description of increasing nitrate mass concentration by 7% in the manuscript between 2006 and 2015 was from model simulation with the input for NO_x emissions of 20.8 Tg (2006) and assumed value of 24.26 Tg (2015) (Wang et al., 2013). However, NO_x emissions showed a declining trend during the period of 2010~2017 (Liu et al., 2017;Vu et al., 2019;de Foy et al., 2016). The mass concentration of particulate nitrate showed a declining trend as well as sulfate during 2013~2017 (Wang et al., 2019).

The discussion on nitrate mass concentration trend was added in the Introduction and the corresponding context is rephrased, as shown below.

"In the recent years, China has also managed to reduce SO_2 emissions by 75% during 2007~2015 (Li et al., 2017) and declined by ~15.1% per year during 2013~2017 (Vu et al., 2019), whereas NO_x emissions declined only by ~10% between 2011 and 2015 (de

Foy et al., 2016) and by ~ 4.3% per year during 2013~2017 (Vu et al., 2019). The strict emission control reduced the $PM_{2.5}$ mass concentration and the corresponding chemical components in China significantly (Vu et al., 2019). The annual mean $PM_{2.5}$ mass loading decreased by 39.6% during 2013~2017 in Beijing-Tianjin-Hebei region, and the SO_4^{2-} and NO_3^- mass concentrations in the $PM_{2.5}$ declined by 40% and 34% respectively during 2015~2017 in Beijing (Vu et al., 2019). However, NH_3 emissions have been observed by satellites to increase by ~30% from 2008 to 2016 over the North China Plain (NCP) (Liu et al., 2018). The faster reduction rate of SO_2 than NO_x emissions in conjunction with elevated NH_3 level, made it reasonable of switching dominant inorganic component in fine aerosol particles from sulfate to nitrate in the recent years similar like European countries (Sun et al., 2015;Hu et al., 2017;Hu et al., 2016;Wu et al., 2018;Guo et al., 2014;Huang et al., 2014;Huang et al., 2010;Ge et al., 2017;Xu et al., 2019;Xie et al., 2019;Li et al., 2018). Field measurements in Beijing show that annually averaged NO_3^-/SO_4^{2-} molar ratio of NR-PM_1 (non-refractory PM_1) in 2012 (1.3~1.8) (Sun et al., 2015) has significantly increased compared to that in 2008 (0.9~1.5) (Zhang et al., 2013). Comparably, the NO_3^-/SO_4^{2-} molar ratio of $PM_{2.5}$ in Beijing increased substantially, from 1.5 before 2013 to 3.33 in 2017 (Xu et al., 2019).”

Q3. The authors need to explain how they estimate the uptake coefficient of N_2O_5 because they discussed factors affecting the coefficients.

We had briefly described how to estimate the uptake coefficient of N_2O_5 in the Method section. In the revised version, we have improved this description, provided a clearer summary of the method and refer more details to the corresponding references, as shown below.

“The uptake coefficient of N_2O_5 was estimated following the method proposed in Chen et al. (2018) and Chang et al. (2016) and references therein. The influences of RH, temperature, multiple inorganic particle compositions, secondary organic aerosol (SOA) and primary organic aerosol are considered. The uptake suppression effect of

N_2O_5 due to the presence of SOA was considered following the method in Anttila et al. (2006). Based on our source apportionment using Positive matrix factorization (SoFi tool, ME2, Francesco Canonaco, PSI), two oxygenated organic aerosol factors (OOA), usually interpreted as SOA, and three primary organic aerosol factors (POA) were determined. The fraction of SOA in the total organic aerosol (OA) was 60%~90% during the observed period, which is quite consistent with the results of a previous study in Beijing (Huang et al., 2014). Hence, 75% was used as the ratio of SOA/OA in our model to estimate the suppression effect of SOA on the uptake of N_2O_5 following the work of Anttila et al. (2006). The reaction of chloride with N_2O_5 was not considered in this study due to its limited mass concentration (on average 5% of the PM_1 mass concentration during the marked haze period), which could cause minor uncertainty in the k_N_2O_5 calculation. The detailed information regarding the estimation γ_{N2O5} is given in Chen et al. (2018), and influence of different chemical components on γ_{N2O5} is summarized in the Table 1 of Chen et al. (2018).”

Q4. Lines 268-270: I think that this depends on RH. Need to verify it.

Good point. To verify this, I used the online thermodynamic model UManSysProp (http://umansysprop.seaes.manchester.ac.uk/) to calculate the hygroscopic growth factor (GF) of pure NH_4NO_3 and $(NH_4)_2SO_4$ particles as a function of RH above their deliquescence RH. As shown in Fig. R1, for the deliquescent pure NH_4NO_3 and $(NH_4)_2SO_4$ particles, the hygroscopic growth factor under the same RH was comparable.

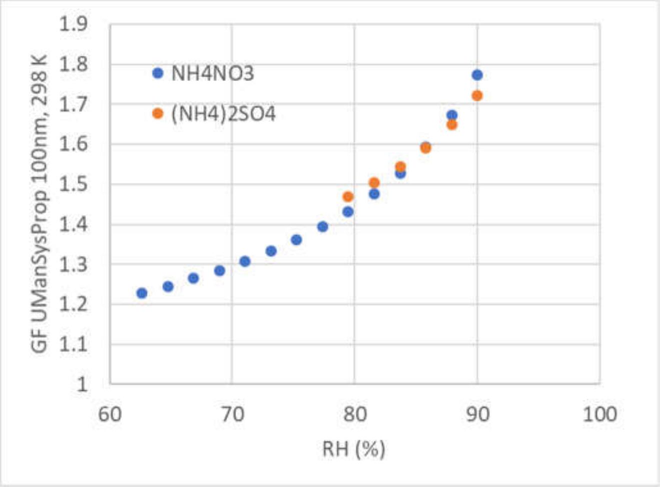


Fig. R1. Calculated hygroscopic growth factor (GF) at 298 K with 100 nm dry particle size for pure NH_4NO_3 and $(NH_4)_2SO_4$ as a function of relative humidity (RH) from UManSysProp (http://umansysprop.seaes.manchester.ac.uk/index). Here, the co-condensation of water and NH_4NO_3 was not considered in the calculation.

The original sentence has been revised as shown below.

As one of the main hydrophilic compounds in atmospheric aerosol particles, the ability of water uptake is comparable between deliquescent $(NH_4)_2SO_4$ and NH_4NO_3 particles with same sizes and ambient RH (Kreidenweis and Asa-Awuku, 2014;Wu et al., 2016), (http://umansysprop.seaes.manchester.ac.uk/).

Q5. Lines 312-313: It can be understood that in their calculation the authors ignore the interaction between dissolved organic components and the activity of NO_3^-. However, small organic acids affect the aerosol hygroscopicity. Can the authors provide the potential impact of such small organic acids on their calculation?

In this study, the aerosol liquid water was calculated based on the measurements of the particle number size distribution, hygroscopicity by H-TDMA, ambient RH and T. Here, the hygroscopicity measurement gives an bulk hygroscopic growth of given particle size. Therefore, the estimated aerosol liquid water has already considered the contribution *from organic compounds. Then, the aerosol liquid water was used as input of E-AIM model to calculate the equilibrium vapor pressure of HNO_3 and NH_3 over wet particles.*

Q6. Molar fraction of nitrate over total of HNO_3 + NO_3^- highly depends on particle pH (e.g., Nah et al., 2018). The authors attributed a strong positive correlation between particulate nitrate and aerosol liquid water to the feedback loop between them, but no particle pH effect has been discussed. Can you exclude the possibility that increased particle pH would lead to an increase in particulate nitrate?

Nah et al., Atmospheric Chemistry and Physics 18.15 (2018): 11471-11491.

Good point. We agree with the reviewer that the higher pH of aerosol particles is favorable for the equilibrium of HNO_3 into the particle phase (Nah et al., 2018). Therefore, We calculated aerosol pH using ISORROPIA Ⅱ and found out that the aerosol pH is not the driving factor for particulate nitrate formation in the investigated case.

A detailed discussion has been added into the Sec. 3.2 (Line 318~330, clean manuscript) as shown below.

"Aerosol pH is also an important factor on the particulate nitrate formation, higher pH is favorable for the equilibrium of HNO_3 into the particle phase (Nah et al., 2018). pH of the fine aerosol particles was calculated by ISORROPIA Ⅱ (Fountoukis and Nenes, 2007) during the investigated period. The model was running in 'forward mode' with chemical composition of NR-PM1 (NO_3^-, SO_4^{2-}, Cl^-, NH_4^+) and gas precursors (HNO_3, HCl, NH_3) by GAC-IC as inputs. And the model was running in 'metastable mode', assuming no solid existed in the system. Generally, the fine aerosol particles became more acidic with pH dropping from ~8 down to ~4 when NR-PM$_1$ mass concentration increased from ~12 $\mu g/m^3$ up to >300 $\mu g/m^3$ as shown in Figure 5 and Figure 6. This declining trend of pH is not favorable for the HNO_3 partitioning into the particle phase

(Nah et al., 2018). However, a clear enhanced trend of molar ratio of particulate nitrate in the total nitrate as a function of NR-PM_1 mass concentration was observed correspondingly (as shown in Figure 5 and Figure 6). Therefore, in this case the increase of aerosol liquid water is more likely to be the driving factor of particulate nitrate formation compared to the influence of pH."

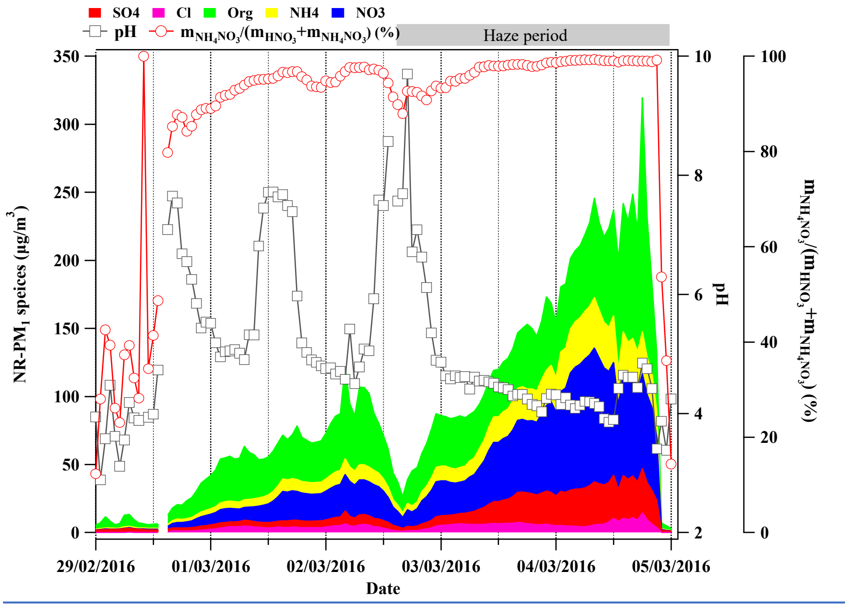


Figure 5 in the revised manuscript (revised from Figure 3 in the old version) The time series of chemical composition measured by HR-ToF-AMS (left axis), calculated aerosol pH by ISORROPIA II (inner right axis) and molar ratio of particulate nitrate in the total nitrate (gas+particle phase) shown on outer right axis.

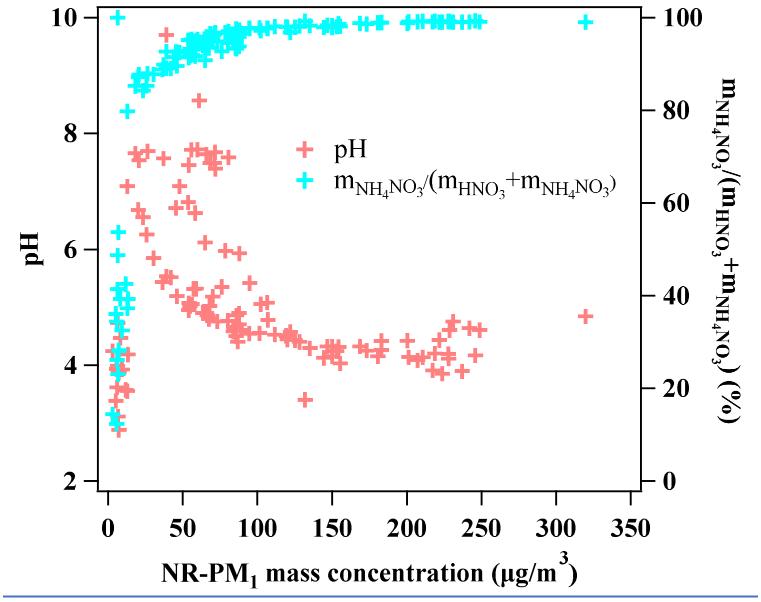


Figure 6 in the revised manuscript. The pH of the fine aerosol particles (left axis) and the molar ratio of particulate nitrate in the total nitrate (gas+particle phase) (right axis) as a function of NR-PM$_1$ mass concentrations.

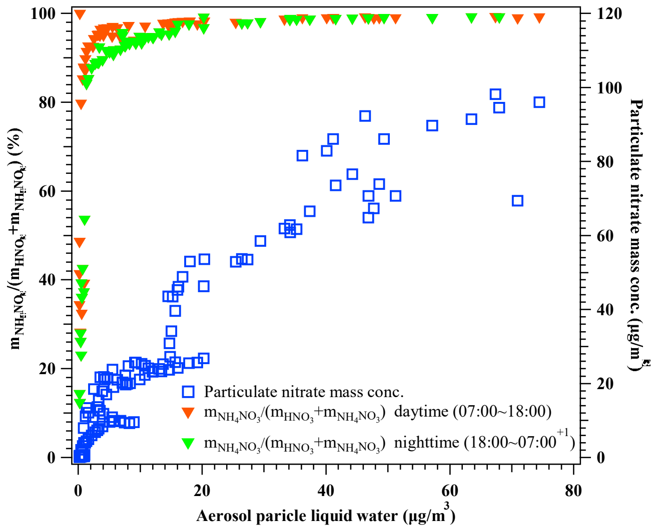
Q7. If the mutual effect of both nitrate and aerosol particle liquid water is the determine factor for increasing nitrate, why did the mass concentration of ammonium nitrate reach a plateau at aerosol liquid water of > 20 µg/m^3?

Thanks so much for your comment on this point. I made a mistake by using the PM$_{2.5}$ nitrate mass concentration from GAC-IC measurement as a function of PM$_1$ aerosol liquid water. The revised figure was shown in revised Figure 4 below, a clear co-increase of particulate nitrate and aerosol liquid water was observed, which has been shown in Figure 1.

The previous description (Line 319~322, original manuscript) has been revised in the manuscript, as shown below.

"Consistently, a significant co-increase of particulate nitrate and aerosol liquid water was observed during haze development as shown in Figure 4. At first, a steep increase

of particulate nitrate in total nitrate mass ratio (from ~12% to ~98%) was observed as the aerosol liquid water enhanced up to ~20 µg/m^3. And then, the particulate nitrate mass kept increasing with further increase of aerosol liquid water. We observed that, ~98% of nitrate was present as particle phase when aerosol liquid water was higher than ~20 µg/m^3."



Revised Figure 4 in the manuscript. The relationship between aerosol particle liquid water and the molar ratio of particulate nitrate in the total nitrate, $m_{NH_4NO_3}/(m_{HNO_3}+m_{NH_4NO_3})$ *(left axis) during the nighttime 18:00~07:00^{+1} (green solid triangle) and the daytime at 07:00 ~ 18:00 (red solid triangle), and mass concentration of particulate nitrate as a function of aerosol liquid water (right axis) during the period of February 29 to March 5, 2016. Here, particulate nitrate was measured by HR-ToF-AMS and the HNO_3 in the gas phase was measured by GAC-IC. Aerosol liquid water was calculated by H-TDMA-derived method.*

Q8. Finally, an increase in RH also leads to increasing aerosol liquid water. As shown in Fig. 1, the increased nitrate is coincident with the elevated RH during the haze period.

How do the authors discriminate the mutual effect from this elevated RH effect?

The aerosol liquid water is determined by the ambient RH and chemical composition. So, the variations of ambient RH has been included in the changes of aerosol liquid water. We demonstrated the increasing of ambient RH as a prerequisite of the mutual promotion effect between particulate nitrate and liquid water.

Minor comments:

Q1. Lines 109-113: Is the discussion about NO_3^-/SO_4^{2-} in China?

This discussion is based on NO_3^-/SO_4^{2-} *in Beijing. This has been clarified in the manuscript.*

Q2. Line 123: Need to be more explicit about how the reactive uptake o precursors and the thermodynamic equilibrium of ammonium nitrate change. Enhancing or increasing?

Here, changing represents enhancing. This sentence has been clarified in the update version as "This leads to an increased aerosol liquid water content (Wu et al., 2018), which will enhance the particulate nitrate formation by increasing the reactive uptake of precursors and the thermodynamic equilibrium of ammonium nitrate (Cheng et al., 2016;Wang et al., 2016;Wang et al., 2017;Yun et al., 2018;Yue et al., 2019)."

Q3. Line 279: theoretical calculations of what?

Here, the theoretical calculations of condensational loss rate of N_2O_5 *and* HNO_3*. This has been clarified in the update version.*

Q4. Line 293: Please use either liquify or liquefy for consistency.

Thanks for your kindness. This has already been corrected in the update version.

Q5. Line 346: HNO3 instead of particulate nitrate?

Thanks for your kindness. This has already been corrected in the update version.

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