

We appreciate reviewer #1 for his/her further comments on our manuscript. Following the reviewer's suggestions, we have revised the manuscript accordingly. Listed below are our response to reviewer #1's comments.

## Response to Reviewer #1

*The authors have addressed most of my major comments in the revised manuscript. Below are a few comments regarding pON quantification by different methods that have to be addressed before publishing in AMT.*

1) Page 4, NO<sub>x</sub> method: *By assuming most of the NO and NO<sub>2</sub> signals observed during high NO<sub>3</sub> loadings, R<sub>AN</sub> of 2.8 and 2.2 are likely higher than that of pure ammonium nitrate due to the presence of pON in ambient. I suggest to provide this information and highlight the potential impacts on pON quantification (e.g. over- or under-determined).*

Following the reviewer's comments, we added the following description in Page 4: "Note that NO<sub>3,org</sub> loadings calculated by "NO<sub>x</sub> method" were slightly underestimated in winter in this case due to the organic nitrate contribution even in a period with high NO<sub>3</sub> loadings."

2) Page 4, Lines 22: *It is somewhat confusing to use "pure ammonium nitrate" here as my understanding is that the R<sub>AN</sub> values used in this study cannot be obtained by pure ammonium nitrate.*

These descriptions in Page 4, line 20-22 are the previous study in summer in NYC regarding factor analysis of combined organic and inorganic aerosol mass spectra. The R<sub>AN</sub> values were determined from pure ammonium nitrate.

To avoid confusion, we added the sampling site and time in those descriptions:

"For instance, Sun et al. (2012) performed factor analysis on combined organic and inorganic aerosol mass spectra and found that the NO<sup>+</sup> and NO<sub>2</sub><sup>+</sup> ions in the nitrate factor were dominantly from inorganics, and the ratio of NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> was close to that of pure ammonium nitrate, while those in OA factors with high NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> were generally assumed as organic nitrates in summer in New York City"

3) Page 5: TD-AMS method: *I would like to follow up my previous comment on the TD-AMS method. Can the authors conduct sensitive test (e.g. varying the MFR in equation 1) to evaluate the impacts on pON quantification and RON values from equation 7?*

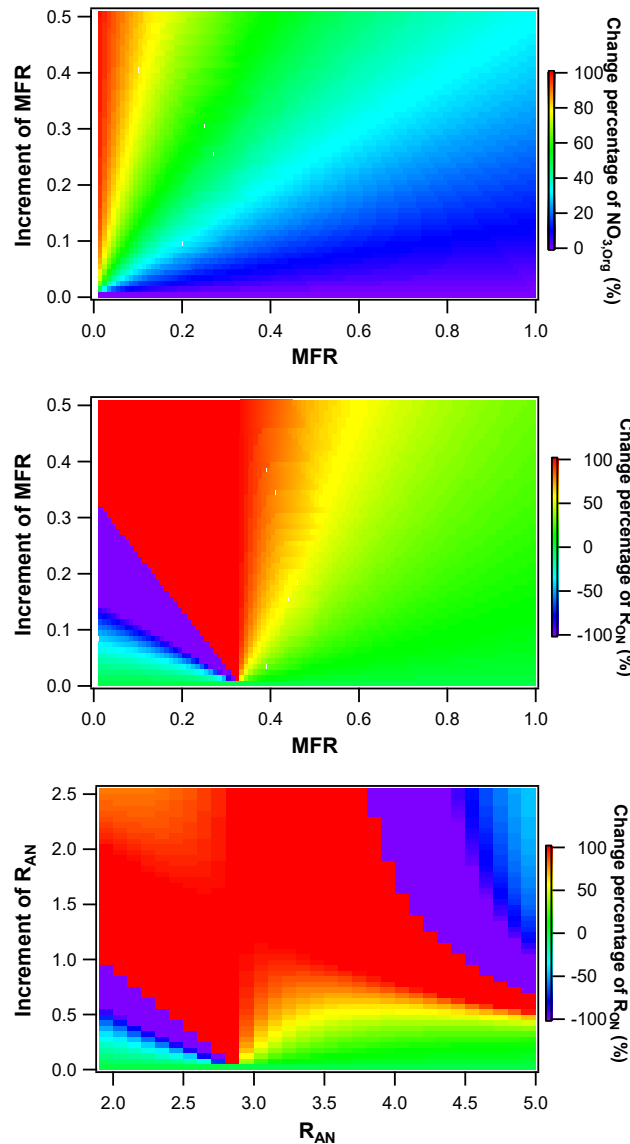


Figure R1. Dependence of increment of MFR ( $R_{AN}$ ) vs. MFR ( $R_{AN}$ ) on the change percentage of  $RON$  and  $NO_{3,Org}$ . The summer data were used as initial independent variables in equation 1-7.

Figure R1 shows the possible coverage of the change percentage of  $RON$  and  $NO_{3,Org}$  as increment of MFR ( $R_{AN}$ ) and MFR ( $R_{AN}$ ). The increment of MFR was in the range of 0 - 0.5 with a step of 0.01, and the MFR varied from 0 to 1. The  $R_{AN}$  varied from  $\sim 2$  to 5, and the increment of  $R_{AN}$  was in the range of 0 - 2.5 with a step of 0.05.

As shown in Fig. R1, the change percentage of  $NO_{3,Org}$  and  $RON$  showed overall increasing trends as the rises of increment of MFR at a fixed MFR. The average MFR of the N-containing ions varied from 0.31 to 0.37 during three campaigns, and we assume that the maximal increment of MFR was less than 0.1 (standard deviation of MFR of the N-containing ions at  $T = 90$  °C). In this case, the change percentage of  $NO_{3,Org}$  was less than  $\sim 20\%$ , suggesting that the impacts of variation of MFR on pON

quantification were relatively low. Comparatively, the change percentage of  $R_{ON}$  was relatively high, suggesting the impact of accurate determination of MFR on  $R_{ON}$ .

We also conducted sensitive test regarding the impact of  $R_{AN}$  on  $R_{ON}$  values (since the pON quantification was independent of  $R_{AN}$ ). The average  $R_{AN}$  varied from 2.2 to 3.8 during three campaigns, and we assume that the maximal increment of  $R_{AN}$  was less than 0.5. As shown in Fig. R1, the change percentage of  $R_{ON}$  was highly influenced by a  $R_{AN}$  rise of less than 0.1, implying the importance of accurate determination of  $R_{AN}$  in TD-AMS method.

Following the reviewer's comments, we added:

“The sensitive tests of  $NO_{3,Org}$  and  $R_{ON}$  with the variation of MFR and  $R_{AN}$  are shown in Fig. S5, demonstrating the impact of accurate determination of MFR and  $R_{AN}$  on  $R_{ON}$  in “TD-AMS method”.”

*4) Page 9, line 16: Refer to my comment #7 in my previous review, it is important to provide the confidence level (90% in this case) in the main text here and to mention diurnal profiles of  $R_{ON}$  has been added in the SI.*

Revised.