#### Report #3

1. Some of the methods are reasonably well described and follow on methods developed and described in the literature, although additional details are needed for several aspects. However, overall nothing new seems to be offered in terms of method development nor advancement in understanding of atmospheric science (which Ref #2 recognized as well). The conclusions the authors arrive at are not surprising and have been previously published. Thus, it largely comes across as a data report, where measurements were made, previous methods are applied and the scientific analysis/interpretation is conducted in this case with less rigor than prior publications. Thus the value to the literature essentially amounts a report of measurements and simple calculations conducted in a different location. The results section consists of only a few figures and ~2 pages of 1.5-space text, a reflection of the thin-ness of new content. Importantly, some conclusions are overstated with insufficient evidence or even faulty logic presented, so in present form is in fact misleading.

#### **REPLY:**

We have done lots of modification and corrected the mistakes based on each comment to offer more solid and convincing conclusions. Please see the details as below.

2. Text was generally readable but there are lots of grammar errors. A few pervasive errors include the addition or omission of articles (e.g. "the", "a") or plurality when not needed or needed. Reviewing by native English speaker BEFORE submission is recommended. Much of the text is written clearly, while in many cases insufficient details were provided to understand exactly what was done.

#### **REPLY:**

We have corrected them in the text.

3. I agree with Referee 1 that there is not sufficient detail provided on the PMF (both justification of solutions, as well as summary of results). This should also include the NOx+ ratios for each factor, discussion of the meaning and/or possible biases related to the HOA factor containing nitrates and the inorganic factor having a different NOx+ ratios than calibrations. Correlations of organic nitrates vs all of the factors should be presented (ideally a version of Fig. 3 for each factor in the supplementary). Simply showing that organic nitrates correlate with LO-OOA does not make a convincing case that they are related since often all concentrations largely increase and decrease at a given sampling site together. Additionally diurnal cycles should be shown for all factors

#### **REPLY:**

The key diagnostic plots of chosen factors and the mass spectrum profiles of 3 to 5 factors for spring, summer and autumn are shown in Figure S2 to S4 (in the supplementary). The NOx<sup>+</sup> ratios for NIAs are given in section 3.1 in the manuscript (2.93 for spring, 3.53 for summer and 3.54 for autumn), and NOx<sup>+</sup> ratios for OA factors are shown in Table S2 in the supplement. And the uncertainties of NO<sup>+</sup> and NO<sub>2</sub><sup>+</sup> in OA factors across different fpeak values are also shown in Table S2. Correlations of organic nitrates vs. all of factors are presented in Figure S7 to S9 in the supplementary. The diurnal cycles for OA factors in each season are shown in Figure S5 and it shows that all OA factors have distinctive variation trends in different seasons.

**Table S2.** The values of NO<sup>+</sup> and NO<sub>2</sub><sup>+</sup> in OA factors when fpeak is 0 and the uncertainties of NO<sup>+</sup> and NO<sub>2</sub><sup>+</sup> in OA factors across different fpeak values (from -1.0 to 1.0)

|        |        |         | НОА      | LO-OOA   | MO-OOA               |
|--------|--------|---------|----------|----------|----------------------|
| Spring | $NO^+$ | fpeak=0 | 1.3*10-2 | 1.4*10-2 | 9.8*10 <sup>-3</sup> |

|        |                              | uncertainty | 3.4%                  | 1.4%                 | 6.8%                 |
|--------|------------------------------|-------------|-----------------------|----------------------|----------------------|
|        | $NO_2^+$                     | fpeak=0     | 1.2*10-2              | 1.5*10-4             | 3.0*10-8             |
|        |                              | uncertainty | 8.5%                  | 3.8%                 | 4.3%                 |
| Summer | $NO^+$                       | fpeak=0     | 1.5*10-2              | 1.0*10-2             | 1.2*10-2             |
|        |                              | uncertainty | 1.5%                  | 5.1%                 | 3.0%                 |
|        | $NO_2^+$                     | fpeak=0     | 1.47*10 <sup>-6</sup> | 6.7*10-4             | 1.8*10 <sup>-3</sup> |
|        |                              | uncertainty | 4.8%                  | 6.9%                 | 4.0%                 |
| Autumn | NO <sup>+</sup>              | fpeak=0     | 1.1*10-2              | 3.1*10-2             | 1.0*10-2             |
|        |                              | uncertainty | 4.5%                  | 0.5%                 | 1.2%                 |
|        | NO <sub>2</sub> <sup>+</sup> | fpeak=0     | 7.0*10-8              | 9.8*10 <sup>-8</sup> | 2.8*10-7             |
|        |                              | uncertainty | 0.6%                  | 1.9%                 | 1.2%                 |

4. Evidence is thin to install confidence that the methods for separation of organic nitrates is producing meaningful separation. While the methods have been applied before in other papers, the methods may be prone to substantial error and potentially bias when organic nitrates are a small fraction of total nitrate, as is the case here for all seasons except summertime. Diurnal cycles of the total nitrate and inorganic and organic nitrates calculated by the different methods would be helpful. Showing correlations of both the organic AND inorganic nitrate with the PMF factors may also be informative. **REPLY:** 

We have added more discussion to support that the separation between organic and inorganic nitrates is meaningful in section 3.1. Diurnal cycles of total nitrate and inorganic and organic nitrates calculated by the different methods are given in Figure 2b in the manuscript. The inorganic nitrate (NO<sub>3 inorg\*</sub>) obtained by subtracting NO3\_org\_ratio\_1 from total measured nitrates also correlated well with the inorganic nitrate estimated using the PMF method (R=0.92 for spring, 0.87 for summer and 0.86 for autumn). Furthermore, the diurnal trends of organic nitrates obtained by the two methods were also similar in each season, generally with lower concentrations in the daytime and higher concentrations at night, while they were distinctive from those of inorganic nitrate (Figure 2b), supporting that organic nitrates had been well separated from inorganic nitrate in this study.

5. The NO3 radical concentration calculation is bewildering. The loss in the steady-state calculation is based on only a-pinene and limonene with no justification for this choice (Section 2.4, Table S1). Then the concentrations are neither reported nor shown, it is unclear if they are calculated for only one fixed value or as a time series. What season was this done for? What season does Table S1 pertain to? This is a critical calculation since the conclusion that NO3 dominates over O3 for BVOC losses and for the SOA modeling. The calculation that NO3 accounts for nearly 100% of the BVOC loss at night is a little surprising and also from which major conclusions of this manuscript flow. For the Xu et al. (2015b) study (referenced in the manuscript), they calculated that only 20%/38% of the reacted a/b-binene was with NO3 at night. Might the NO3 calculated here be biased high since only two VOC losses were considered?

#### **REPLY:**

Since on-line VOCs measurement was only performed during the spring campaign, the following theoretical analysis of NO<sub>3</sub>+VOCs reactions will be just applied to the spring case. Typical nighttime VOC concentrations, their reaction rate coefficients for reacting with NO<sub>3</sub> radical are listed in Table S3. five biogenic VOCs (i.e., $\alpha$ -pinene, isoprene,  $\beta$ -pinene, Comparing to limonene and camphene )accounting for 99% NO<sub>3</sub> loss in Table S3 in Xu et al. (2015b), one anthropogenic VOC, styrene made the third largest contribution to NO<sub>3</sub> loss, which should not be ignored in the related SOA estimation analysis. But  $\alpha$ -pinene and limonene do contribute to nearly 90% of NO<sub>3</sub> loss in our case due to their much higher nighttime concentrations than other BVOCs and rapid reaction rates with NO<sub>3</sub> radical. The nighttime estimated concentration of NO<sub>3</sub> radical in this study is 1.24±0.76pptv, which is about 15 times higher than the nighttime concentration of NO<sub>3</sub> radicals (0.076 pptv) reported in Xu et al. (2015b), this is because that the value of [NO<sub>2</sub>]×[O<sub>3</sub>] (20ppbv×6.8ppbv) in this case is just about 15 times higher than that value (0.54ppbv for NO<sub>2</sub> and 21 ppbv for O<sub>3</sub>) in Xu et al. (2015b). Thus, the nighttime concentration of NO<sub>3</sub> and O<sub>3</sub> is 0.076 pptv and 21ppbv, respectively. The much higher NO<sub>3</sub> level and lower O<sub>3</sub> level in this study leads to almost all VOCs reacting with NO<sub>3</sub> radical over O<sub>3</sub> at night.

| Table S3  | 3. The average campaign  | concentrations of             | f VOCs measured w      | rith an automa  | ted in situ     |     |
|-----------|--------------------------|-------------------------------|------------------------|-----------------|-----------------|-----|
| gas-chro  | matography mass spectr   | ometer (GC-MS),               | their reaction rate of | coefficients fo | r reacting with | NO3 |
| radical a | nd the production potent | tial from NO <sub>3</sub> +VO | C in spring.           |                 |                 |     |

| VOC species            | Mean concentration (ppbv) | <b>Rate Coefficient</b> | Production potential |
|------------------------|---------------------------|-------------------------|----------------------|
|                        |                           |                         | (pptv/s)             |
| 1,2,3-Trimethylbenzene | 0.057                     | 1.90E-15                | 3.72E-06             |
| 1,2,4-Trimethylbenzene | 0.177                     | 1.80E-15                | 1.10E-05             |
| 1,3,5-Trimethylbenzene | 0.051                     | 8.80E-16                | 1.54E-06             |
| 1,3-Butadiene          | 0.052                     | 1.00E-13                | 1.79E-04             |
| 1-Butene               | 0.415                     | 1.32E-14                | 1.89E-04             |
| 1-Hexene               | 0.022                     | 1.20E-14                | 9.06E-06             |
| 1-Pentene              | 0.022                     | 1.20E-14                | 9.27E-06             |
| 2,2,4-Trimethylpentane | 0.068                     | 9.00E-17                | 2.10E-07             |
| 2,2-Dimethylbutane     | 0.199                     | 4.40E-16                | 3.01E-06             |
| 2,3,4-Trimethylpentane | 0.022                     | 1.90E-16                | 1.44E-07             |
| 2,3-Dimethylbutane     | 0.299                     | 4.40E-16                | 4.54E-06             |
| 2,3-Dimethylpentane    | 0.293                     | 1.50E-16                | 1.51E-06             |
| 2-Methylheptane        | 0.034                     | 1.90E-16                | 2.20E-07             |
| 2-Methylhexane         | 0.514                     | 1.50E-16                | 2.66E-06             |
| 2-Methylpentane        | 1.582                     | 1.80E-16                | 9.81E-06             |
| 3-Methylheptane        | 0.027                     | 1.90E-16                | 1.74E-07             |
| 3-Methylhexane         | 0.534                     | 1.50E-16                | 2.76E-06             |
| 3-Methylpentane        | 1.411                     | 2.20E-16                | 1.07E-05             |
| Acetaldehyde           | 1.249                     | 2.70E-15                | 1.16E-04             |
| Acetylene              | 0.941                     | 5.10E-17                | 1.65E-06             |
| Acrolein               | 0.042                     | 3.30E-15                | 4.73E-06             |
| Benzene                | 0.599                     | 3.00E-17                | 6.19E-07             |
| cis-2-Pentene          | 0.005                     | 3.70E-13                | 6.57E-05             |
| Cyclohexane            | 1.164                     | 1.40E-16                | 5.61E-06             |
| Cyclopentane           | 0.416                     | 1.40E-16                | 2.00E-06             |
| Ethane                 | 1.567                     | 1.00E-17                | 5.40E-07             |

| Ethylbenzene       | 0.563 | 1.20E-16 | 2.34E-06 |
|--------------------|-------|----------|----------|
| Isoprene           | 0.032 | 6.96E-13 | 7.76E-04 |
| m/p-Xylene         | 0.602 | 3.80E-16 | 7.88E-06 |
| Methacrolein       | 0.012 | 3.40E-15 | 1.44E-06 |
| Methylcyclohexane  | 0.172 | 1.40E-16 | 8.29E-07 |
| Methylcyclopentane | 0.673 | 1.40E-16 | 3.25E-06 |
| n-Butanal          | 0.044 | 1.10E-14 | 1.68E-05 |
| n-Butane           | 1.848 | 4.60E-17 | 2.93E-06 |
| n-Decane           | 0.060 | 2.80E-16 | 5.74E-07 |
| n-Heptane          | 0.351 | 1.50E-16 | 1.81E-06 |
| n-Hexane           | 1.916 | 1.10E-16 | 7.25E-06 |
| n-Nonane           | 0.033 | 2.30E-16 | 2.59E-07 |
| n-Pentanal         | 0.128 | 1.50E-14 | 6.61E-05 |
| n-Pentane          | 0.593 | 8.70E-17 | 1.78E-06 |
| n-Propylbenzene    | 0.029 | 6.00E-16 | 6.01E-07 |
| Octane             | 0.064 | 1.90E-16 | 4.17E-07 |
| o-Xylene           | 0.464 | 3.80E-16 | 6.06E-06 |
| Propanal           | 0.144 | 6.31E-15 | 3.12E-05 |
| Propane            | 3.678 | 7.00E-17 | 8.86E-06 |
| Propene            | 0.477 | 9.54E-15 | 1.57E-04 |
| Styrene            | 0.194 | 1.50E-12 | 1.00E-02 |
| Toluene            | 3.120 | 7.00E-17 | 7.52E-06 |
| alpha-Piene        | 0.391 | 6.21E-12 | 8.36E-02 |
| beta-Piene         | 0.013 | 2.51E-12 | 1.10E-03 |
| Camphene           | 0.276 | 6.20E-13 | 5.91E-03 |
| Limonene           | 0.137 | 1.22E-11 | 5.77E-02 |

6. As Referee 2 points out regarding the analysis in Sect. 3.3 on the estimation of pON formation, the sources of measured pON depend on reacted VOCs, not on the amount of VOCs present in the atmosphere. The authors seem to be equating the two. Essentially the authors appear to be calculating the relative amounts of production expected if the sampled airmass was allowed to react to completion with no further emissions. This does not equate to the regional productions since it would systematically underweight more reactive compounds and the two metrics may only be loosely connected. In general, Section 3.3. is highly undersupported and speculative considering the evidence shown. It is not clear what the modeled SOA (blue trace in Fig. 5) even is. Is that the amount produced per unit time from the model? It cannot be the cumulative production since it increases and decreases (and the model does not have dynamics, dilution, mixing, etc.). In any case, the features of organic nitrates and the ambiguous modeled SOA don't match all that well. Also, it seems likely that the day-to-day variability may be of similar or larger magnitude to the variations in the average diurnal cycle (variability bars such as standard deviations would be helpful here). Potentially the very rough similarities may be an averaging artifact? Thus, the diurnal cycle should be supplemented additional evidence, such as time series of the relevant metrics and correlations plots to make a convincing case that the model may be indeed be representing the key processes and explain the measurements. Also, inclusion of inorganic nitrates together with the organic nitrates (diurnal cycle and other relevant plots), would help make a stronger case that the

separation of the organic nitrates is meaningful and robust. Based on this section, the authors conclude (as stated in the abstract) that BVOC + NO3 at night are the dominant formation pathway of organic nitrates in the polluted atmosphere. This simply has not been demonstrated. Moreover, it appears that the authors are equating boundary layer concentration with overall regional production importance. As the authors note in Sect. 3.3, the boundary layer is expected to be lower during nighttime. Consequently, the concentrations observed represent a smaller volume of air, so equating lower concentrations during daytime with lower overall (column integrated, regional) importance is faulty logic. BL-effects were not considered here nor production during the daytime modeled, thus no conclusions beyond nighttime boundary layer concentrations should be drawn based on this analysis. Yet, this manuscript seems to do just that – an example of the vastly overstated implications claimed.

#### **REPLY:**

We have accepted this comment and changed the analysis in section 3.4 significantly. First, we used the NO<sub>3</sub> loss rate at night, which can be calculated as  $K_i \cdot [VOC_i]$  in Eq. (9), to roughly judge the production potential of organic nitrates from a NO<sub>3</sub>+VOC reaction:

## $[Production Potential]_{NO3+VOCi} = K_i \cdot [VOC_i] \cdot [NO_3] (9)$

Where  $K_i$  represents the reaction rate coefficient for NO<sub>3</sub> radical and a VOC,  $[VOC_i]$  is the concentration of the specific VOC and  $[NO_3]$  is the concentration of NO<sub>3</sub> radical. In the spring campaign, the diurnal variations of NO<sub>2</sub>, O<sub>3</sub> and estimated NO<sub>3</sub> radical concentrations are shown in Figure S10 (with standard deviations). It was found that the high concentrations of NO<sub>2</sub> (19.93±2.31 ppb) at night leaded to high yield of NO<sub>3</sub> radical (1.24±0.76 ppt) in Shenzhen.

According to the distribution of production potential, five biogenic VOCs (BVOCs) (i.e.,  $\alpha$ -pinene, limonene, camphene,  $\beta$ -pinene and isoprene) and one anthropogenic VOC (styrene) were identified as notable VOC precursors with high production potential, while the sum of production potential from the other VOCs was negligible as shown in Figure 5b.

In addition, Figure 6 shows the average nighttime variations of BC, LO-OOA, NO<sub>3.org\_ratio\_1</sub>, NO<sub>3.org\_PMF</sub> and production potential of the six notable VOCs identified during the spring campaign. The concentrations of BC and LO-OOA generally decreased slowly after sunset till sunshine due to the combined effect of both the planetary boundary layer variation and traffic emissions, while particulate organic nitrates showed a different trend with two clear growth processes (19:00-22:00 and 3:00-6:00) at night, suggesting their unique sources. In contrast, the production potentials of the six notable VOCs with NO<sub>3</sub> had two roughly similar increases at the same periods as those of particulate organic nitrates, which supported the key role of NO<sub>3</sub>+VOCs reactions for nighttime organic nitrate formation.

Finally, based on the production potential evaluation above, we further estimated roughly the nighttime SOA bulk yield of NO<sub>3</sub>+the six notable VOC precursors. And the estimated SOA production from NO<sub>3</sub>+VOCs reactions using SOA mass yields in the literature was 0-0.33  $\mu$ g m<sup>-3</sup> for  $\alpha$ -pinene, 0.09-1.28  $\mu$ g m<sup>-3</sup> for limonene, 0.24  $\mu$ g m<sup>-3</sup> for styrene, 0.004-0.06  $\mu$ g m<sup>-3</sup> for  $\beta$ -pinene and 0.002-0.02  $\mu$ g m<sup>-3</sup> for isoprene. The SOA yield from camphene is currently unknown in the literature. It is seen that the average observed nighttime concentration of particulate organic nitrates during the spring campaign (0.39-0.83 $\mu$ g m<sup>-3</sup>, converting NO<sub>3.org\_ratio\_1</sub>, NO<sub>3.org\_PMF</sub> in Figure 6 into organic nitrates assuming the average molecular weight of organic nitrates of 200 to 300 g mol<sup>-1</sup>) was well within the estimated SOA concentration ranges produced by  $\alpha$ -pinene, limonene and styrene, indicating that these three VOCs were the key VOC precursors in urban atmosphere in Shenzhen. Considering both the production potentials and SOA yields, the contributions of  $\beta$ -pinene and isoprene to nighttime formation of particulate organic nitrates study be negligible. Besides the BVOCs species, this study

highlights the key role of anthropogenic styrene in nighttime particulate organic nitrate formation in urban atmosphere in China, and relevant smog chamber studies for anthropogenic VOCs+NO<sub>3</sub> reactions are needed to support parameterization in modeling.

7. Kiendler Scharr et al. (2016), Ng et al. (2017), Xu et al. (2015a, 2015b) and others have reported that at many sites throughout the US and Europe, including polluted urban areas, organic nitrates can be substantial fractions of aerosol nitrates and NO3+BVOC reactions can be an important source. The manuscript fails to make a case for what new information this study from one polluted city provides and how it would add to the body of atmospheric literature.

#### **REPLY:**

In the revised manuscript, we have proposed several new information: 1.adding some description in Introduction to address the contribution of this paper on organic nitrates studies in detail. "Ng et al. (2017) reviewed the nitrate radical chemistry and the abundance of particulate organic nitrates in the United States and Europe, and further concluded that particulate organic nitrates are formed substantially via NO3+BVOC chemistry, which plays an important role in SOA formation. Unfortunately, relevant Chinese datasets are scarce yet and not included in this review. This was because (1) the contributions of organic nitrates in SOA and total nitrates in Chinese atmosphere remain poorly understood; (2) the anthropogenic and biogenic precursor emissions in China are largely different from those in the United States and Europe, and thus cannot be easily estimated. To our best knowledge, few studies have investigated the concentrations and formation pathways of particulate organic nitrates in China. Xu et al. (2017) estimated the mass concentration of organic nitrogen in Beijing using AMS, but in this study they ignored the contribution of  $NO_{X^+}$  family, which are the major fragments of organic nitrates"; 2. in section 3.3, we discussed the size distribution characteristics of organic nitrates. We used the NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> ratio as an indicator to investigate the size distribution characteristics of organic nitrates. It is clearly found that the NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> ratio generally increases towards smaller size in spring, summer, and autumn, while the  $NO^+/NO_2^+$  ratio keep similar to the value of  $R_{NH4NO3}$  throughout the full size range in winter. It should also be noted that in spring, summer, and autumn, the lowest values of  $NO^+/NO_2^+$  ratio at > 1 µm are also approximate to the seasonal values of  $R_{NH4NO3}$ . These characteristics clearly indicate that organic nitrates occurred mostly in fresh particles with smaller sizes, and thus should be mainly of local origin. The diurnal trends of size distributions of NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> ratio show higher values in small size range at night comparing that in the day in spring, summer and autumn, suggesting a dominant nighttime origin of organic nitrates; 3. according to the analysis in section 3.4, we can find that besides the BVOCs species, anthropogenic styrene plays an important role in nighttime particulate organic nitrate formation in urban atmosphere in China; 4. we compared this study with other particulate organic nitrate studies in section 3.5 and the results show that the formation of particulate organic nitrate is more likely NOx-control than BVOCs-control and high NOx emissions could promote biogenic SOA formation at night.

8. The authors broadly refer to their results as pertaining to "South China" which is a large and diverse areas. Unless there is evidence that this site is generally representative of that geographic area, text and conclusions should be limited to just this one urban area. This is an example of overselling the story without providing the supporting analysis.

#### **REPLY:**

We have toned down this conclusion and only addressed this case as a typical urban site in South China.

9. L32-3: "Play a larger role" for what? Reference?

## <u>REPLY:</u>

We have added the related references.

10. L40-41: The Rollins et al. (2002) paper demonstrated the application of the aerosol-only organic nitrate measurement. The technique for total nitrates was developed and demonstrated a decade earlier (Day et al., 2002).

# **REPLY:**

We have corrected it.

11. L41: "measured" would be better than "obtained"

## **REPLY:**

We have corrected it.

12. L66: "to obtain more representative samples" seems vague. Clarify.

## **REPLY:**

We have deleted "more representative".

13. L74: "literatures" should not be plural.

## **REPLY:**

We have corrected it.

14. L82-4: state if the RIE of ammonium was calibrated or assumed.

# **REPLY:**

RIE of ammonium was assumed and we have added the statement in section 2.2.1: The relative ionization efficiencies (RIEs) used in the study were 1.2 for sulfate, 1.1 for nitrate, 1.3 for chloride, 1.4 for organics, and 4.0 for ammonium (Jimenez et al., 2003).

15. L105-6: Site Xu et al. (2015a) since isn't this is exactly what they did?

## **REPLY:**

We have added this reference.

16. L107-8: Stating that negative calculated organic nitrates means that concentrations must be low is not very analytically sound. E.g., can't this just mean the method isn't working well or there are large uncertainties due a variety of possible factors? Please revise to be more precise and inclusive of the possible causes.

## **REPLY:**

We have modified this statement and provided more detailed and precise discussion in section 3.1.

17. L127: Fry et al. 2013 is not the proper reference for heterogeneous N2O5 reaction with aerosol. They just performed the calculation.

## **REPLY:**

We have replaced the right literature (Dentener and Crutzen, 1993) with it in the manuscript.

18. L130: Units for velocity are wrong.

## **REPLY:**

We have corrected it.

19. L131: Where did the aerosol surface area concentration for the modeling come from? It does not appear that an aerosol sizing instrument was used in the study.

# **REPLY:**

SA is calculated from the size-resolved particle number concentrations assuming spherical particles measured by a scanning mobility particle sizer (SMPS) (TSI Inc., USA, 3775 CPC and TSI Inc. 3080 DMA). And 220  $\mu$ m<sup>2</sup> cm<sup>-3</sup> is actually under dry conditions, the ambient (wet) aerosol SA is 475  $\mu$ m<sup>2</sup> cm<sup>-3</sup> by using the hygroscopic growth factor in Liu et al. (2010). We have added and corrected the related description in section 2.2.2 and Text S1.

20. L133-135: Why do the authors include the calculation of the N2O5+H2O gas-phase reaction? The lifetime is 500 years!

# **REPLY:**

We have corrected the value of daily maximum  $[H_2 O]$  (5.5\*10<sup>17</sup>molecule cm<sup>-3</sup>) and the calculated value of N<sub>2</sub>O<sub>5</sub> lifetime with respect to the reaction with H<sub>2</sub>O (1470 s).

21. L145: "6.82 and 19.38 ppb": Too many significant figures. It would be more useful to report averages and standard deviations.

## **REPLY:**

The concentrations of  $NO_2$  and  $O_3$  with their standard deviations are shown in Figure S10 in the supplementary.

22. L159: "may explain" instead? Without any quantitative assessment, it is not justified to say that it does explain it.

## **REPLY:**

We have replace "explain" with "may explain".

23. L165: "adequate" for what or by what measure? Too vague – needs clarification.

# **REPLY:**

We have replace "adequate" with "good".

24. L173-4: Statement points out that the organic nitrate is similar to SEUS "even though" BC and NOx are higher in S. China. How is this meaningful? If terpenes are dominantly reacting with NO3 then the production would be largely controlled by the amount of terpenes present - which appear to be quite modest at this location. Statements like this need more context/discussion to be meaningful. Or remove such comparisons if not informative and making a clear point.

## **REPLY:**

We have deleted this statement.

25. L180: "both organic spectra" should instead read "both OOA spectra"? **REPLY:** 

We have corrected it

26. L180-2: Unclear. Is this referring to running PMF with and without the NOx ions? Please clarify. **REPLY:** 

We have clarified this statement: The mass spectrum profiles and diurnal patterns of each OA factor using PMF based on OA spectra only in spring, summer and autumn are shown in Figure S2-S5.

27. 182-3: Please report these correlations (table?) and show the correlations in the SI (i.e. duplicate Fig. 3 for MO-OOA and HOA).

## **REPLY:**

We have added Figure S7-S9 to show the correlations between organic nitrates with HOA and MO-OOA.

28. L196-198: Limonene and a-pinene shown to account for 90% of NO3 loss? Assuming the authors are referring to Sect. 2.4 (not 2.3 as written), this is not shown there - or anywhere else. Only 2 compounds were considered according to the text (which references Table S1).

#### **REPLY:**

We have added the related information in section 3.4 and table S3.

29. Table 1: "NO3-": Is that inorganic nitrate or total nitrate? If total, then the ionic denotation isn't appropriate.

#### **REPLY:**

We have replace "NO3-" with "total NO3-".

30. Figure 5: beta-pinene misspelled. **REPLY:** 

We have corrected it.

31.Table S1: a-pinene SOA yield: Why was this single reference value picked, considering that there is a substantial range reported in the literature? See Table 2 in Ng et al. (2017). A range would seem more appropriate here unless the authors justify why this particular one is more appropriate for this study.

# **REPLY:**

We have added more references of SOA yield, please see Table 3 in the manuscript.

#### References

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