We thank the Referee for the very insightful comments. We have revised our manuscript according to the suggestions of the Referee's comments and our responses to the comments are as follows. Referee's comments are in black, authors' responses are in blue, and changes to the manuscript are in red color text.

This manuscript reported a series of indoor chamber experiments on secondary organic aerosol (SOA) formation from oxidation of furan in a smog chamber. Specifically, the authors conducted experiments under different concentrations of NOx and variable RH conditions. They found that the relatively high NOx level and increasing relative humidity can promote the formation of furan SOA. Based on mass spectra and FTIR analysis, the chemical composition of furan SOA were investigated, which are mainly organic nitrate compounds. The topic is appropriate for the journal, and the paper is in general well written. I believe this manuscript could be accepted after the following issues are addressed.

1. Page 3, line 29. "...that all the NO, NO2, NOx, and ozone concentrations were less than 1 ppb, and the particle concentration was lower than 10-3 cm-3." NOx=NO+NO2, thus, "NOx" should be deleted. The particle concentration should be 103 cm-3 but not 10-3 cm-3. The reviewer doubts that the 4 hrs' clean time is long enough to make the chamber cleaned thoroughly. The number of particles in a fully cleaned chamber should be none or only several to dozens. How the authors eliminate the effects of background particles?

Author reply:
"NOx" has been deleted as suggested.

Concerning the particle concentration, we intended to show that the particle mass concentration is lower than 10^{-3} μg m^{-3}, which demonstrates that the chamber was clean enough for conducting the experiment.

To clean the chamber, pure air was flown for at least 4 h and to confirm that there were no effects of background particles, the background samples were analyzed by SMPS before each experiment, and no particle could be detected in the chamber. To remove any confusion regarding the background gases and particles, the quoted text has been modified as:

"The chamber was cleaned by flowing pure air for at least 4 h prior to each experiment until residual NO, NO$_2$, O$_3$ or any other particles could not be detected in the chamber."

2. Page 3, line30, if this 10-3/cm3 number is correct? I think this is probably wrong.

Author reply:
As indicated in our reply above, we previously used a wrong unit, which is now corrected in the revised manuscript.

3. Page 4, line 4. "NaCl was solid when RH was below 45 % (Gao et al., 2007), and liquid droplets when RH was higher than 60 % (Ge et al., 2017a)". The DRH and

ERH of NaCl are about 75% and 45%, respectively. The authors need to make sure that they clearly understand the conditions of the references and quote them correctly.

Author reply:

The reason why NaCl was liquid at RH≥60% is that the initial addition of the particle was in the form of liquid droplets. To answer the concern of the Referee, the sentence has been corrected as:

"NaCl was solid when RH was below 45 % (Gao et al., 2007), and liquid droplets when RH was higher than 75 % (Tang et al., 1977). But the seed particles would be in the form of droplets if after generation by the atomizer, they do not go through a diffusion dryer before being introduced into the reactor (Ge et al., 2016)."

4. Page 4, line 14-15, what are the units of these numbers? if they are ppb for NO2 and O3, and particle numbers for the aerosol particles? Please clarify.

Author reply:

The gas concentration is in ppb, whereas the particle mass concentration is in $\mu g\ m^{-3}$. Generally, the wall loss rate constants are derived from measured concentrations vs. time. Their units are $s^{-1}/min^{-1}/h^{-1}$ and here we use "s^{-1}".

5. Page 4, line 27. "(Master (Master TD, Dani, Italy)", the "(Master" should be deleted.

Author reply:

This has been deleted.

6. Page 6, Experimental section. The program settings of ESI-MS during sample analysis should be addressed.

Author reply:

In our ESI-MS analysis, the LC was not combined with HR-MS. To provide more information about the sample analysis, the text at page 6 was modified as:

"The mass resolution is 10^5. The collected SOA samples were eluted with 1 mL of acetonitrile. A total volume of 300 L was sampled at a flow rate of 10 L min^{-1} and a volume of 5 μL of the extraction was used, through a direct injection, for the measurement. The addition of acetonitrile allows a lower surface tension of the solution and provides a stable electrospray ionization process (Koch et al., 2005). Both positive (+) and negative (-) ion modes were chosen to operate. N_2 was used as the sheath and auxiliary gas. The desolvation gas temperature was 320 ℃, and the gas flow was 200 $\mu L\ min^{-1}$. The capture voltage was set to 3 KV."

7. Page 6, line 20. In the manuscript, the authors mentioned that "With the decrease of the NOx concentration, the concentration of O3 increased rapidly in the beginning of

the experiment to reach the maximum, and then remained constant throughout." As an oxidant, O3 would be consumed during the experiments. The authors should explain the reasons why the O3 could remain constant during the experiments. I do not think the explanation that "the main sinks of furan are oxidations by O3 and OH, and the reaction products are RO2 and HO2 radicals, which promote the cycling of O3 formation by converting the NO back to NO2" is the main reason.

Author reply:
The main oxidant during the experiments is OH instead of O_3, which can hardly react with furan in the gas-phase. After turning on the lights, NO_2 was converted into NO, accompanied by the formation of OH radical and O_3. The OH radical is mostly produced by the photolysis of O_3 and HONO (Gligorovski et al., 2015), which are all generated from the cycling of NOx as presented in Figure S2. In the beginning of the experiments, the formation of O_3 was dominant with NO_2 converting into NO, which resulted in the increase in O_3 concentration as shown in Figure 1. However, with the consumption of NO_2, the amount of O_3 formed decreased gradually to reach a constant level. According to the Referee's suggestion, the sentence "Overall, the main sinks of furan are oxidations by O_3 and OH, and the reaction products are RO_2 and HO_2 radicals, which promote the cycling of O_3 formation by converting the NO back to NO_2." has been deleted, and further clarification is given in the revised manuscript at page 6 as:
"In the beginning of the experiments, the formation of O_3 was dominant with NO_2 converting into NO, which resulted in an increase in O_3 concentration. The formed O_3 would further photolyze to yield the main oxidant, the OH radical. However, with the consumption of NO_2, the amount of O_3 formation decreased gradually to reach a constant level."

8. Page 6, 3rd paragraph. During the experiment, the number concentration of particles decreased with time. Besides the coagulation of particles, the authors thought that "With SOA formation, organics start to dominate and result in the enhanced growth of clusters, which promote the decay of the particle number concentration". I do not agree with that. The formation of clusters can promote the new particle formation, which will increase the particle number concentrations. On the other hand, what is the role of NaCl seed during the experiments? Is the SOA mainly formed on the surface of seed particles? Does the existed particles make senses on new particle formation? It is important to understand this process, because it is related to the formation mechanism of SOA.

Author reply:
We intended to find if there was any possibility for the new particle formation to take place, and we made the contour plots of SOA bursts as shown in Figure S3. However, we could not find any evidence to support this. Furthermore, as presented in a previous study, although there is a competition between the formation of new particles and growth of existing particles by uptake of the precursors, vapor condensation onto

existing aerosol particles is favored (Liu et al., 2017). To avoid misunderstanding, we have deleted the sentence "With SOA formation, organics start to dominate and result in the enhanced growth of coagulation, which promotes the decay of the particle number concentration", and added the following on Page 6, 3rd paragraph.

"A previous study conducted in a similar smog chamber showed a decrease in particle number concentration during the experiment (Liu et al., 2017)."

Small particles that act as seed particles for SOA formation are widespread in the atmosphere. NaCl particles used in the present study were used as the seeds in the photooxidation of furan. We conducted an experiment without using NaCl seed particles, and realized no SOA formation under this condition. However, when using NaCl seed particles, the SOA were formed. Furthermore, the formed SOA could easily condense on the surface of NaCl seed particles (Abramson et al., 2013). NaCl seed particles are not only the substrate for vapor-phase condensation, but they also take part in the reactions at the surface. NaCl can react with HNO_3 and N_2O_5, which may influence the O_3 and SOA formation from furan-NOx irradiations. The $NaNO_3$ detected by IC is the evidence for secondary reactions taking place. The following sentences were added in the Introduction part, for a better understanding of the aim of adding NaCl seed particles.

Page 2
"Sea salt aerosols are the second most abundant primary inorganic aerosols in the atmosphere, wherein they play an important role in atmospheric chemistry near coastal regions (Beardsley et al., 2013; Rossi, 2003). They constitute the main components of particulates in the marine troposphere. A previous study has shown that hygroscopic particles act as a sink for volatile organic compounds and take part in the aqueous phase reactions (Volkamer et al., 2007)."

Page 3.
"NaCl particles acted as the nuclei and took part in secondary reactions."

9. Page 7, line 9. The authors mentioned that "Experiments were started with an average of 12.6 µg m-3 of NaCl seed aerosol". However, in part 2.1 on page 4, they said the initial mass concentrations of seeds were 6 µg m-3 on average.

Author reply:
We apologize for this mistake. The data were checked again, and the average initial concentration of NaCl seed aerosol was 6 µg m^{-3}. This has been updated in the revised manuscript.

10. Page 7, line 16. The sentence that "It exhibits a sharper increase of O3 formation at 42 % RH than that at 5 % RH" is confusing, because it lacks of preconditions.

Author reply:

The sentence has been re-written as:

"With similar NOx level, the amount of O_3 formed at 5 % RH was larger than that at 42 % RH."

11. Page 7, line 17. The authors should show the reasons why the influence of RH on O3 is not obvious compared to that of NOx. What is the intrinsic mechanism?

Author reply:

The influence of NOx level on O_3 formation is more important than that of RH because NOx is directly linked to the O_3 formation in two ways: (i) with the photolysis of NO_2, the O atom produced, reacts further with O_2; (ii) direct reaction of RO with NO_2. Both pathways directly affect the NOx level, which has repercussions on O_3 formation. However, the RH affects O_3 formation by propelling NO_2- and ONO_2- containing products into SOA particles as presented in a previous work (Jia and Xu, 2014). At high RH conditions, the SOA formation is promoted whereas O_3 formation is suppressed. The O_3 formation is instead favored under low RH conditions.

We have added the following text at Page 7:

"This obvious influence of NOx level on O_3 is likely related to its formation mechanism. The formation of O_3 is directly connected with the NOx in two ways: the photolysis of NO_2, producing the O atom that reacts with O_2, and the NOx reacting directly with RO. However, the RH has little to no effect on O_3 formation. This formation is slightly favored at low RH whereas, at high RH, the NO_2- and ONO_2- are easily transferred into the aerosol phase, thereby suppressing the O_3 formation (Jia and Xu, 2014)."

12. Page 8, line 1 and line 17. Much higher SOA yield can be obtained under different RH conditions than that under different NOx levels. Why? The authors should give the intrinsic mechanisms of SOA formation under different NOx levels and RH conditions.

Author reply:

Much higher SOA yield was observed under high RH conditions because the presence of water plays an important role in SOA formation. Previous studies have shown that solid NaCl particles can carry water as a thin liquid-like surface layer (Allen et al., 1996; Vogt and Finlaysonpitts, 1994). With RH increasing, the water content on the NaCl surface increases and the liquid-like surface layer thickens. This condition favors the aqueous-phase reactions and affects the uptake of water-soluble gases, which contributes to SOA formation (Hennigan et al., 2008; Lim et al., 2010). Furthermore, the presence of ALW in the particles can facilitate the partitioning of semi-volatile organic compounds, resulting in the enhancement of SOA formation (Sareen et al., 2017). The ALW may also promote photodegradation of dissolved SOA

(Romonosky et al., 2015). A previous study also showed that much higher yields of SOA were generated under high RH conditions (Kamens et al., 2011).

Generally, NOx plays two roles in a reaction process. Increasing NOx concentration will promote the formation of O_3 and HONO, leading to more OH radical formed, which in turn is favorable to SOA formation (Sarrafzadeh et al., 2016). High NOx levels can also facilitate the competition between NO/NO_2 and HO_2 to react with RO_2. Products with high volatility will be generated more by the $NOx+RO_2$ reaction than the HO_2+RO_2 reaction (Kroll and Seinfeld, 2008). However, the formation of lower volatility products is favored by the increase of SOA yields (Chen et al., 2018). In this regard, the increasing NOx level is not conducive to SOA formation. In the present study, the OH radical plays a determining role in the photooxidation process of furan, and the NOx level does not reach the critical value for the $NOx+RO_2$ reaction to dominate the process. The main fate of RO_2 is mainly through reaction with HO_2 instead of NO/NO_2, which contributes to SOA formation.

In the revised manuscript, the SOA formation mechanism is backed up at page 11 as:
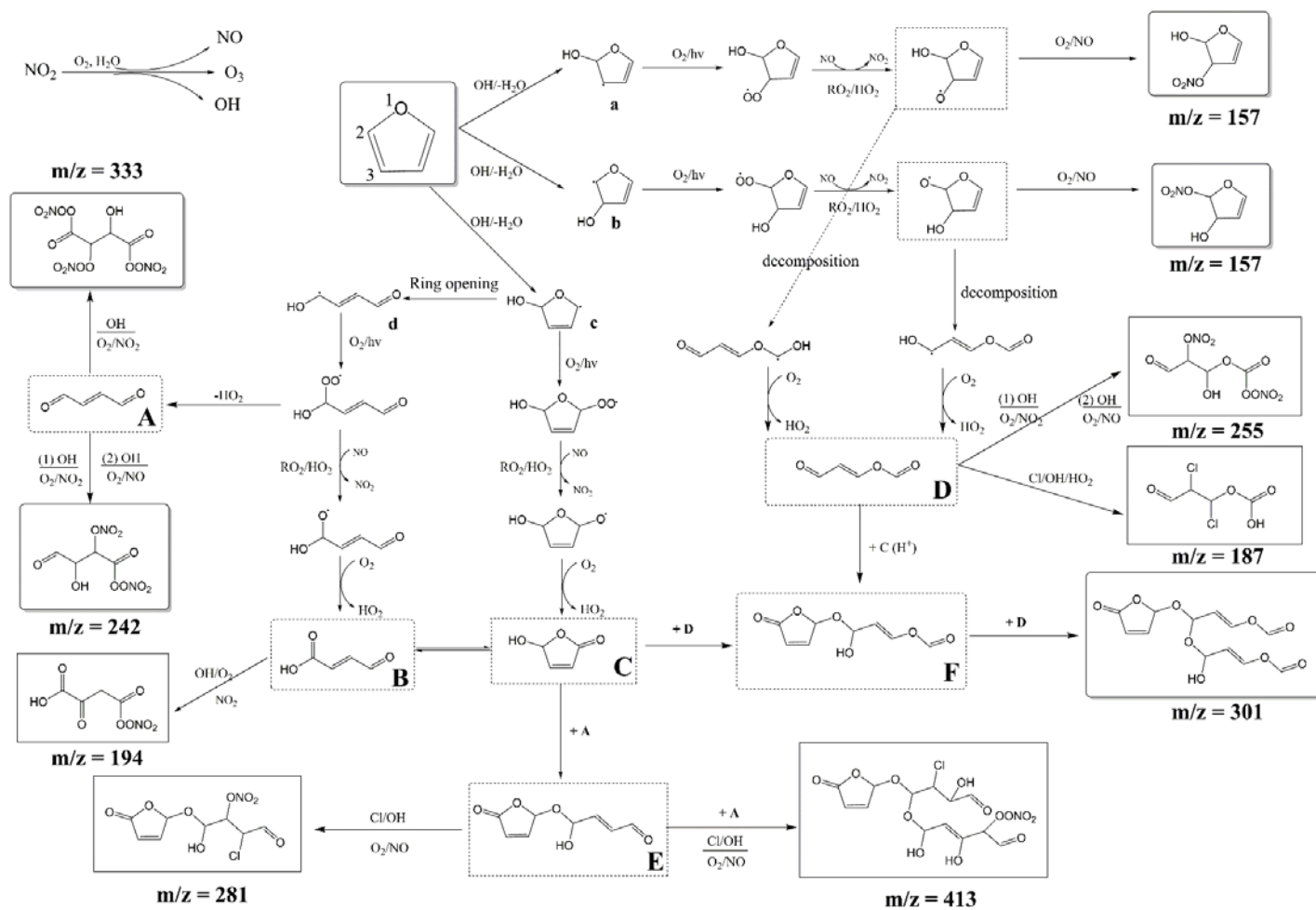
" 3.4 Mechanism of furan SOA formation

According to the identified products in this work and based on previous kinetic (Atkinson et al., 1983; Lee and Tang, 1982) and products (Villanueva et al., 2009; Aschmann et al., 2014b; Tapia et al., 2011; Villanueva et al., 2007; Strollo and Ziemann, 2013) studies reported in the literature, together with tropospheric reactivity principles, a proposed chemical mechanism for SOA formation from furan in the presence of NOx is shown in Scheme 1. After turning on the lights, NO_2 was converted to NO, accompanied by the formation of the OH radical and O_3. The most efficient oxidant in the present study is the OH radical, for which the rate constant of the reaction with furan is 7 orders of magnitude higher than in the case O_3 as oxidant (Atkinson et al., 1983). The OH radical is mostly produced by the photolysis of O_3 and HONO (Gligorovski et al., 2015), which are both generated from the cycling of NOx as presented in Fig. S2 (Stemmler et al., 2006; Li et al., 2008). More extensive studies on gas-phase photochemistry of alkyl furans have shown that the process is initiated by the OH-radical addition to the aromatic ring (Aschmann et al., 2014a; Gomez Alvarez et al., 2009). Additionally, on the basis of well-established mechanisms for atmospheric volatile organic compounds (Atkinson and Arey, 2003), it can be concluded that the reaction is initiated by OH addition to a C=C bond at C2 or C3 positions. Addition at the C2 position forms two cyclic alkyl radicals (a, c), one of which (a) can isomerize to form a ring-opened alkyl radical (d), whereas addition at the C3 position forms a single alkyl radical (b). The OH radical addition leading to an hydrogen abstraction generates the alkyl radicals (R·) followed by reaction with O_2 to form alkylperoxy radicals (RO2·) (Pan and Wang, 2014). The ring-opened alkylperoxy radical generated from (d) can decompose to generate an unsaturated 1,4-dialdehyde (A). Moreover, the formed alkylperoxy radicals (RO2·) can either react with RO_2/HO_2 or NO to yield the corresponding alkoxy radical (RO·), which can (i) react with NO/O_2 to form a first-generation peroxydinitrate compound (m/z = 157) (Atkinson, 2000), (ii) react with O_2 to form a first-generation unsaturated

products 1,4-aldoacid (B) and hydroxyfuranone (C), or (iii) decompose and then react with O_2 to yield a 1,4-aldoester (D). It is worth mentioning that the hydroxyfuranone (C) can go through a ring-opening reaction via acid-catalyzed heterogeneous/multiphase reactions to form the unsaturated 1,4-aldoacid (B). Scheme 1 also shows the formation of second-generation products (E) and (F) via reactions of the hydroxyfuranone (C) with 1,4-dialdehyde (A) and 1,4-aldoester (D), respectively.

As detected by the ESI-Exactive-Orbitrap MS, the hydroxynitrates (m/z = 333, 242) were generated by the reactions of 1,4-dialdehyde (A) with OH radicals and O_2/NO/NO_2 (Carlton et al., 2009; Ervens et al., 2011). There are two pathways for hydroxynitrates formation from RO_2 radicals in the presence of NOx according to which RO_2 radicals may react with NO and NO_2 to form $RONO_2$ and $ROONO_2$, respectively (Kroll and Seinfeld, 2008). Hydroxynitrates with m/z = 194 and 255 were produced by similar mechanisms based on 1,4-aldoacid (B) and 1,4-aldoester (D) reactions, respectively. The identified chlorinated organic compounds were generated from the former products (D) and (E), in which hydrogen atoms were substituted by Cl atoms. The OH radicals can convert Cl^- ions in the NaCl droplets into Cl atoms, which can rapidly react with organic compounds in the particle-phase and yield identified chlorinated organic compounds (m/z = 281, 413, 187).

As discussed above, the yields of SOA increased with the increase of the NOx concentration in the present study. Generally, the NOx level has two different effects in the reaction process. Increasing NOx concentration will promote the O_3 and HONO formation, leading to more OH radical formed, which in turns is favorable to SOA formation (Sarrafzadeh et al., 2016). In addition, high NOx level can facilitate the competition between NO/NO_2 and HO_2 to react with RO_2. Products with high volatility will be generated more by the NOx+RO_2 reaction than by HO_2+RO_2 reaction (Kroll and Seinfeld, 2008). However, the formation of lower volatility products favors the increase of SOA yields (Chen et al., 2018). In this regard, the increasing NOx level is not conducive to SOA formation. It was shown that the yields of SOA generated from the photooxidation of *m*-xylene increased firstly and then decreased with the increase of the NOx level (Chen et al., 2018). In the present study, the OH radical plays a determining role in the photooxidation process of furan, and the NOx level does not reach the critical value for the NOx+RO_2 reaction to dominate the process. The main fate of RO_2 is mainly through reaction with HO_2 instead of NO/NO_2, which contributes to SOA formation. A previous study on the photooxidation of *m*-xylene/NOx demonstrated that extremely high NOx level (*m*-xylene/NOx < 5.0) suppressed the SOA formation compared to experiments conducted under *m*-xylene/NOx > 8.0 conditions (Song et al., 2005). Nonetheless, the experimental conditions conducted in the present study did not include the HC:NOx ratio < 5.0. Consequently, we did not observe a decreasing trend of SOA yield as the NOx level increased, which is different from the photooxidation of other aromatic compounds where increasing NOx concentration suppresses the SOA formation. The effect of RH on SOA formation in furan photooxidation is dominantly determined by the aqueous photochemistry under high RH conditions as discussed above. The heterogeneous reactions including acid catalyzed reactions at the surface of particles promote the formation of first-generation

gloxal-like products, which likely play an important role in the process of SOA formation. The proposed gloxal-like compound (D) plays a substantial role in the obvious increasing of m/z=187 product formation under high RH conditions. Previously, unsaturated first-generation reaction products of 3-methyl furan have also been suggested to undergo acid-catalyzed condensed-phase reactions, with SOA yields up to 15% (Strollo and Ziemann, 2013). In addition, the reinforced effect of RH on SOA yield was also ascribed from the photooxidations of other aromatic compounds, such as, benzene (Ng et al., 2007), toluene (Hildebrandt et al., 2009; Kamens et al., 2011), and xylene (Zhou et al., 2011)."

Scheme 1: Proposed mechanism for the SOA formation from furan photooxidation. SOA constituents in dotted and solid boxes are proposed first-generation products and ESI-Exactive-Orbitrap MS detected products, respectively.

13. Fig. 4. The SOA yields had a sharper increase under <45% RH conditions, but a gentle increase at >45% RH. How the authors draw a conclusion that the ALW played an important role in gas/particle partitioning during the experiments? The abbreviation of "ALW" in the caption or "LWC" in the figure should be consistent.

Author reply:
The 42% RH used in the present work is very close to the ERH of NaCl particles. It has been previously shown that SOA formation decreases both the ERH and DRH of the seed particles and results in the uptake of water by the particles (Liu et al., 2018; Takahama et al., 2007; Smith et al., 2012). There is a high possibility that the NaCl seeds will effloresce early after being coated by the new formed SOA. The effect of efflorescence contributes to the water uptake of the particles, leading to aqueous-phase reactions and the uptake of water-soluble gases. To make it clear, we have added the following text on Page 8.
"An obvious increase of SOA yield was observed when the RH increased from 37% to 42%. This phenomenon was mainly caused by the efflorescence transition when the seed particles were coated with SOA. It has been previously shown that SOA formation decreases both the efflorescence RH and deliquescence RH of the seed particles and results in the uptake of water by the particles (Liu et al., 2018; Smith et al., 2012; Takahama et al., 2007). There is a high possibility that the NaCl seeds effloresce early after being coated by the new formed SOA. The effect of efflorescence contributes to the water uptake by the particles, leading to the obvious trend-changing of SOA yield."

The caption in Fig. 4 has been corrected.

14. Page 8, line 33. Although the organic groups of -COOH, -OH, -CO and –COH are hydrophilic, the hygroscopic growth factor (GF) of SOA is about 1.05 at 75% RH (Meyer et al., 2009), which is quite weak compared with that of NaCl and NaNO3. Thus, the contribution of SOA to ALW is not important.

Author reply:
We totally agree with the Referee that the GF of SOA is quite weak compared with that of NaCl and $NaNO_3$, and that the contribution of SOA to ALW is not important. Hence, we have deleted the original sentence on page 8.

15. Page 10, 1st paragraph. The results of FTIR showed that all functional groups increased by factors of about 2 when the RH ranged from 42 % to 85 %. Why the SOA yields showed in Fig.4 only had an increase of 14% when RH ranged from 42 % to 85 %?

Author reply:
In the revised manuscript, Figure S4 and Figure S5 show the variations of FTIR absorption abundance of different functional groups at different NOx and RH levels

relative to the absolute abundance detected at $C_4H_4O/NOx=7.8$ and 85% RH, respectively. The difference between trends of FTIR results and the SOA yield is due to the fact that FTIR results were based on the normalized relative abundance instead of absolute abundance. As a result, the FTIR data exhibit an obvious variation of the SOA components compared to the SOA yield. Furthermore, it is possible that the absorbance intensity of a certain wavenumber range does not only arise from different substances with the same functional group, but may also be affected by one substance with more than one same functional group. For example, the –CO group in carbonyl and carboxylic compounds can absorb in the wavenumber range 1600-1800 cm^{-1}. This detection mechanism will cause a superimposed effect of the absorbance intensity. Therefore, it is not proper to compare the results of FTIR and SOA yield quantitatively.

16. Fig. 9 is not clear enough. Positive and negative ion modes were used to analyze different kind of compounds, respectively. The authors should make a comparison between the same modes, and mark the identified compounds in the figure.

Author reply:
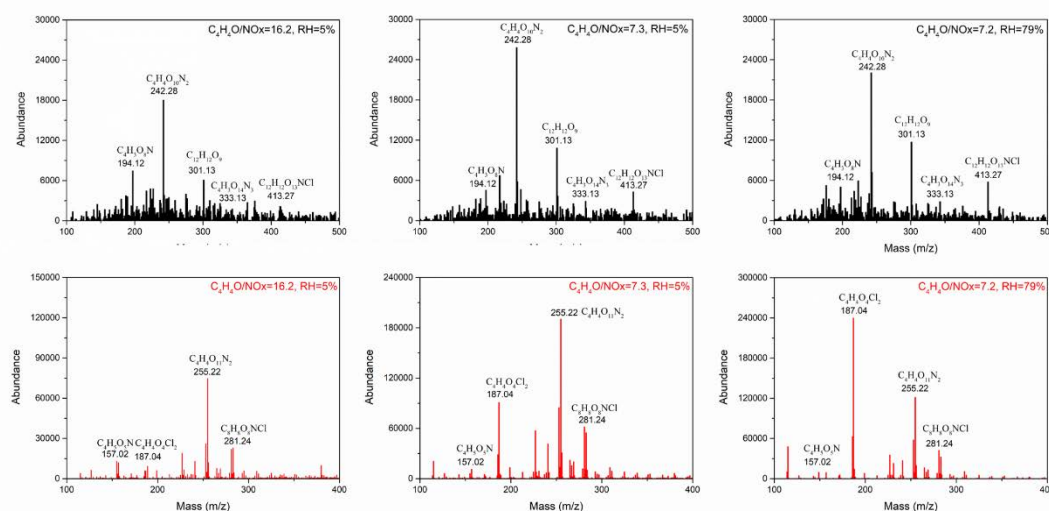Figure 9 has been modified as suggested by the Referee and the new figure is as follows:

Figure 9: Background-subtracting mass spectra of SOA in both positive ion mode (black) and negative ion mode (red) from the photooxidation of furan under different experimental conditions.

17. From the results of FTIR and ESI-MS, the authors obtained that the compounds were similar both under different NOx levels and RH conditions. Does it demonstrate that the SOA formation mechanisms are similar? If so, what's the role of seed particles in the experiments? What is the mechanism of furan SOA formation?

Author reply:
Based on the results from both the FTIR and ESI-MS, we conclude that the major

products are generated by a similar mechanism under different NOx and RH conditions. In the experiment, NaCl particles acted as nuclei to facilitate the deposition of semi-volatile organic compounds. Moreover, the NaCl particles took part in the secondary reactions. For a better understanding of the role of seed particles in the experiments, please refer to our response to comment 8. Also, mechanistic insights of furan SOA formation have been discussed in comment 12 above and were added in the revised manuscript.

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