We have revised our manuscript according to the suggestions of the Referee's comments. For clarity, the Referee' comments are reproduced in blue, authors' responses are in black and changes in the manuscript are in red color text. Pages and lines of modified/inserted/deleted texts are relative to the previous version of the manuscript.

Anonymous Referee #4

This manuscript presents interesting new results on atmospheric reactions of photooxidation of cyclohexene in the presence of SO2. Unfortunately the quality of the presentation is not suitable for publication in ACP regarding scientific discussion and interpretation of results. The manuscript must be rewritten to discuss the results from a more objective, scientific point, which to a higher degree takes data uncertainty into account before reaching conclusions. Furthermore, there are many grammatical errors. In the following I have explained some of the major points.

In addition to the many comments responded below, the whole manuscript has been checked, several errors have been fixed, to improve the quality of the manuscript.

Introduction the introduction should include more references to relevant previous work. One example is in line 23 page 1, where only one study (on measurement methods for VOC from vehicles) is used as reference for the general statement about emission of alkenes from biogenic and anthropogenic sources.

We carefully reviewed the citations and several relevant previous works were added, some of which are given below:

Chin, J. Y., and Batterman, S. A.: VOC composition of current motor vehicle fuels and vapors, and collinearity analyses for receptor modeling, Chemosphere, 86, 951-958, doi: 10.1016/j.chemosphere.2011.11.017, 2012.

Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, T. F., Monod, A., Prevot, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmos. Chem. Phys., 9, 5155-5236, doi: 10.5194/acpd-9-3555-2009 2009.

Hatch, L. E., Creamean, J. M., Ault, A. P., Surratt, J. D., Chan, M. N., Seinfeld, J. H., Edgerton, E. S., Su, Y. X., and Prather, K. A.: Measurements of isoprene-derived organosulfates in ambient aerosols by aerosol time-of-flight mass spectrometry-part 1: Single particle atmospheric observations in atlanta, Environ. Sci. Technol., 45, 5105-5111, doi: 10.1021/es103944a, 2011.

Kesselmeier, J., Kuhn, U., Rottenberger, S., Biesenthal, T., Wolf, A., Schebeske, G.,

Andreae, M. O., Ciccioli, P., Brancaleoni, E., Frattoni, M., Oliva, S. T., Botelho, M. L., Silva, C. M. A., and Tavares, T. M.: Concentrations and species composition of atmospheric volatile organic compounds (VOCs) as observed during the wet and dry season in Rondonia (Amazonia), J. Geophys. Res., 107, LBA 20-21–LBA 20-13, doi: 10.1029/2000jd000267, 2002.

Kroll, J. H., and Seinfeld, J. H.: Chemistry of secondary organic aerosol: Formation and evolution of low-volatility organics in the atmosphere, Atmos. Environ., 42, 3593-3624, doi: 10.1016/j.atmosenv.2008.01.003, 2008.

Paulson, S. E., Chung, M. Y., and Hasson, A. S.: OH radical formation from the gas-phase reaction of ozone with terminal alkenes and the relationship between structure and mechanism, J. Phys. Chem. A, 103, 8125-8138, doi: 10.1021/Jp991995e, 1999.

The reference Jimenez et al. (page 1 line 28) on reactions of polyfluorinated compounds is not relevant for a general statement on SOA formation in the atmosphere, and it should thus be removed.

This reference was deleted.

On page 2 (lines 27-30) it is stated that a substantial amount of organosulfates have been observed. Most measurements estimate up to 10% of aerosol mass, and typically much lower in most places, which in my opinion is not a "substantial amount".

The original meaning of “substantial amount” was wrong. Our aim was to point out that different organosulfates were observed. Hence, "substantial amount" has been changed to “different kinds”.

Why so much focus on organosulfate formation from MBO, which is typically not found in high concentrations? Page 2 lines 11-21: This section has a good number of relevant references.

We agree that MBO is not among the major molecules responsible for organosulfates formation. From the literature, laboratory chamber studies showed that OH/NOx/O_3-initiated reactions of BVOCs, such as isoprene, α-pinene, β-pinene, and limonene with sulfates or sulfuric acid are the main formation processes for organosulfates formation. Although organosulfates formation from MBO photooxidation is not as important as from isoprene and pinene reactions, MBO emissions were found to be larger than isoprene emissions in some regions (Baker et al., 1999). Hence, organosulfates formation from MBO reactions would not be negligible in those conditions. To take this into account in the manuscript, the text at page 2 lines 24 to page 3 line 10 was modified as:

“Despite the existence of organosulfates in ambient aerosols was first observed in

2005 (Romero and Oehme, 2005), proper identification of these aerosols was made two years later. In a series of chamber experiments studies, it was shown that organosulfates present in ambient aerosols collected from various locations mostly originate from acid-catalyzed reactions of SOA formed from photooxidation of α-pinene and isoprene (Surratt et al., 2007). Recently, different kinds of organosulfates have been observed in SOA around the world, and organosulfates have been identified as a group of compounds that have an important contribution to the total amount of SOA in the atmosphere (Surratt et al., 2008; Froyd et al., 2010; Kristensen and Glasius, 2011; Tolocka and Turpin, 2012; Wang et al., 2015). Laboratory chamber studies showed that OH/NOx/O_3-initiated reactions of BVOCs, such as isoprene, α-pinene, β-pinene, and limonene with sulfates or sulfuric acid are the main processes for organosulfates formation (Surratt et al., 2007; Surratt et al., 2008; Hatch et al., 2011). Despite the well-recognized presence of organosulfates in SOA, their formation and transformation processes can be complex and varied, depending on the nature of the original organic compound involved. Extensive studies on their formation have been performed and several mechanisms based on a variety of reactions have been proposed. Using nuclear resonance techniques, isoprene-derived epoxides formed during isoprene photooxidation reactions were found to be important intermediates for organonitrates and organosulfates formation via potential SOA reactions (Darer et al., 2011; Hu et al., 2011). The authors further found that organonitrates could easily be transformed to organosulfates during hydrolysis in the presence of sulfate. Some studies also showed that 2-Methyl-3-buten-2-ol (MBO), due its larger emissions than isoprene in some regions (Baker et al., 1999), is an important precursor for organosulfates and SOA in the atmosphere, through its reactions with OH under NO and aerosol acidity conditions, and from acid-catalyzed reactive uptake of MBO-based epoxides formed during MBO photooxidation (Mael et al., 2015; Zhang et al., 2012; Zhang et al., 2014). Organosulfates formation was also found from oxidation of hydroxyhydroperoxides (Riva et al., 2016) and from heterogeneous reactions of SO_2 with selected long-chain alkenes and unsaturated fatty acids (Passananti et al., 2016)."

Experimental Page 4 line 8: The VOC/NOx ratio was not about 5, but varied from 4.4 to 7.

The ratio of VOCs/NOx has been fixed.
The sentence at page 4, line 8 "The mixed concentration ratios of VOCs/NOx were adjusted to be about 5" was changed to "The mixed concentration ratios of VOCs/NOx were in the range 4.4-6.9."

Section 2.3: Why were only aerosols in the range 108-650 nm collected? What was the limit of detection of the IC analysis?

The particles were collected by a Dekati low pressure impactor (DLPI, DeKati Ltd, Finland). The size range of particles collected by this device was 0.03-10μm, which was divided into thirteen segments. According to the results of particle size range measured by SMPS as shown in Figure R1 below, the mass of particles formed were in the range of 75-300 nm. These were in the 108-650 nm range, which corresponds to the third segment, within which most of the particles from our measurements could be collected. The particles in this range constitutes about 97% of the total mass, both measured (black bars) and simulated (red line).

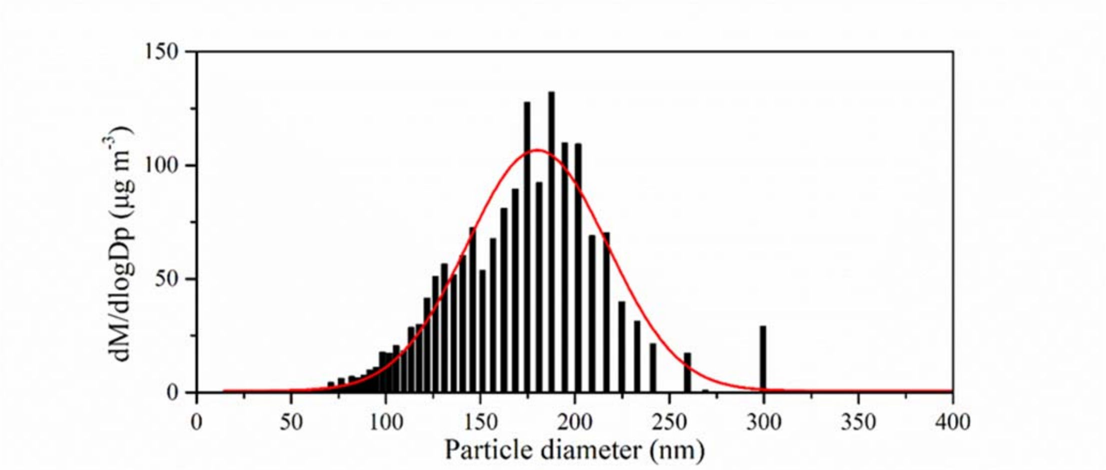

Figure R1. Particle size distribution

For the IC analysis, the limit of detection was 0.2 mg mL^{-1}.
The following is added in the manuscript at page 5 line 13 to clarify:
"For the IC analysis, the limit of detection was 0.2 mg mL^{-1}."

Were samples for FTIR and SOA analysis collected right after each other?

No. While FTIR is very sensitive and does not require high concentrations of the samples, the SOA analysis needs higher concentrations. For SOA analysis, the concentrations were increased in order to have more SOA formation. Hence, the samples for SOA analysis were collected after the samples for FTIR analysis. However, both samples types were collected using the same method, and all experiments were conducted using the same procedure.

Results and discussion Section 3.1 should be moved to the experimental section.

We have moved the content of Section 3.1 to the experimental section.

Section 3.2 Page 6 line 22. The trend is not clear, especially regarding maximum particle number, which shows quite some scatter.

Page 6 line 26-32: The conclusions in this section are beyond what I see in the data, given the scatter.
Why are some of the experiments not shown in Figure 1? Only 11 out of 17 experiments can be seen.

Based on the available data and despite the scattered behavior of the maximum particle number concentration relative to the particle number concentration at maximum yield with increasing SO_2 concentration, we can find an acceptable correlation between the two types of number concentrations, as we mentioned at page 6 lines 21-22. The change in the trend of SOA number concentrations is quite evident from the particle number concentrations at the maximum SOA yield as can be observed in Figure R2 below.

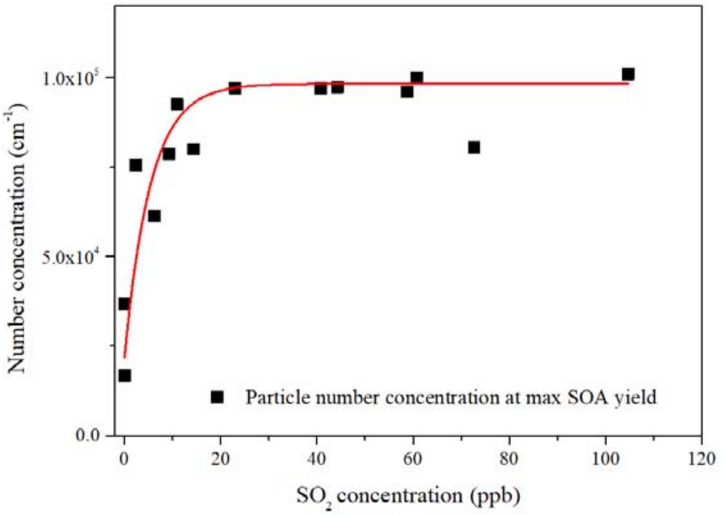

Figure R2: Particle number concentrations of SOA in the photooxidation of the cyclohexene/NOx/SO_2 system with different initial SO_2 concentrations.

The scattered behavior of the maximum particle number concentrations was likely caused by measurement errors. These concentrations were obtained upon particles formation after the black light lamps were switched on. SOA number concentrations increased rapidly to reach the maximum, and subsequently, the particle number concentrations decreased, as shown in Figure S1. The particles were collected by SMPS at a sampling interval of 5 min. The decrease rate of the particle number concentration was very fast, about 25% after 5 min, which also corresponds to the time at which the maximum number concentration was obtained.
To avoid any misinterpretation of our data and our conclusions due to the data being scattered, Figure 1 of the main manuscript was replaced by Figure R2 in this response.

There are two main reasons why there are some maximum particle number concentrations were missing. Firstly, as shown in Figure S1, the maximum number concentration was obtained in the beginning of SOA formation, but the maximum mass concentration was obtained one hour later. For our experiments, we focus on the

ultimate yield of SOA and hence, the SMPS analysis was not performed immediately after the UV light was turned on.
Secondly, the collection volume by the impactor was slightly smaller than the volume of the chamber. In order to reduce gas consumption in the smog chamber, the particles were not sampled continuously by SMPS from the beginning of the experiment. Despite the missing data on the maximum particle number concentration the trend of SOA number concentration with initial SO_2 is obvious.

Page 7 lines 1-4: This discussion is very unclear.

We re-wrote this as:
"It is evident from Figure 1 that even small amounts of SO_2 affect the new particle formation substantially. This is in agreement with the finding that wood soot, a minor source of SO_2 (Reddy and Venkataraman, 2002), resulted in a measurable positive deviation to the VOCs/NOx photooxidation reaction system without background aerosol (Jang et al., 2002)."

Page 7 line11: It is of course difficult to reproduce concentrations of VOC and NOx in an experiment, which is also clear from the present work. Therefore the statement of "similar" conditions is too strong. VOC concentration varies from 472 to 665 ppb, which also affects SOA formation to some degree (seen by plotting the data presented here).

We agree that the statement with "similar" can be misleading. Because this statement does not actually give extra information on the stated finding, it was deleted, however, without changing the meaning of the main result.

Page 7 line 18-19: This seems speculative.

This sentence was deleted.

Page 7 line 22-23: This meaning is unclear.
This was re-written as:
"New particles were formed by vapor condensation onto existing aerosol particles."

Section 3.3. Page 7 line 30: According to your data in Table S1 the SOA yield without SO2 present was 2.7-3.4%, not 2.5-2.7% as stated here.

This was fixed.

Page 8 line 10: The ratio was 4.4-7 according to Table S1, not 5 as stated here.

This was fixed.

Figure 2. The SOA yield shows a trend, but only to some degree, since the values for low SO2 concentrations are somewhat scattered, while experiments at high concentrations have not been repeated. This means that conclusion about a trend in the data is based on very few data points at high SO2 concentrations. The discussion e.g. on page 8 lines 23-31 should be revised considerably with this in mind.

Due to the error associated with measuring SO_2 concentrations, many experiments were needed at low concentrations (below 40 ppb) for a better reproducibility of the experimental data since even 1 ppb error in SO_2 concentration can have significant effects. However, this error is less important at higher SO_2 concentrations and hence, few data points were used. The scattered behavior of the SOA yield at low SO_2 concentrations is primarily a consequence of the errors due to measuring the concentrations, and the trend of Figure 2 is not expected to change. Based on this, the discussion on page 8 lines 23-31 is modified as:

"SOA yields for the cyclohexene/NOx/SO_2 system were measured for initial SO_2 mixing ratios of 0-105 ppb. Due to the error associated with the SO_2 concentrations measurement, with stronger impact on low values than on higher values, several experiments were performed at SO_2 concentrations below 40 ppb. The experimental results showed a clear decrease at first and then an increase in the SOA yield with increasing SO_2 concentrations (Figure 2). When SO_2 concentrations increased from 0 to 40.8 ppb, there was a remarkable decrease in SOA yield, dropping by about half with the increase of SO_2 concentration. For SO_2 concentrations higher than 40.8 ppb, SOA yield increased with increasing SO_2 concentration. The highest SOA yield was obtained to be 3.5%, at 104.7 ppb SO_2 concentration. The lowest SOA yield of cyclohexene photooxidation was obtained at the initial SO_2 concentration of 40 ppb. Although the SOA yield increased gradually with the initial SO_2 concentration at concentrations higher than 40 ppb, a higher SOA yield than that in the absence of SO_2 could not be obtained when the initial SO_2 concentration was lower than 85 ppb."

Page 9 line 1-3: Which experiments were with NO and which with NO2?

A new plot for Figure 2 was made, in which experiments initiated with NO and NO_2 are clearly distinguished. Also, different NO- and NO_2-initiated experiments are marked in Table 1.

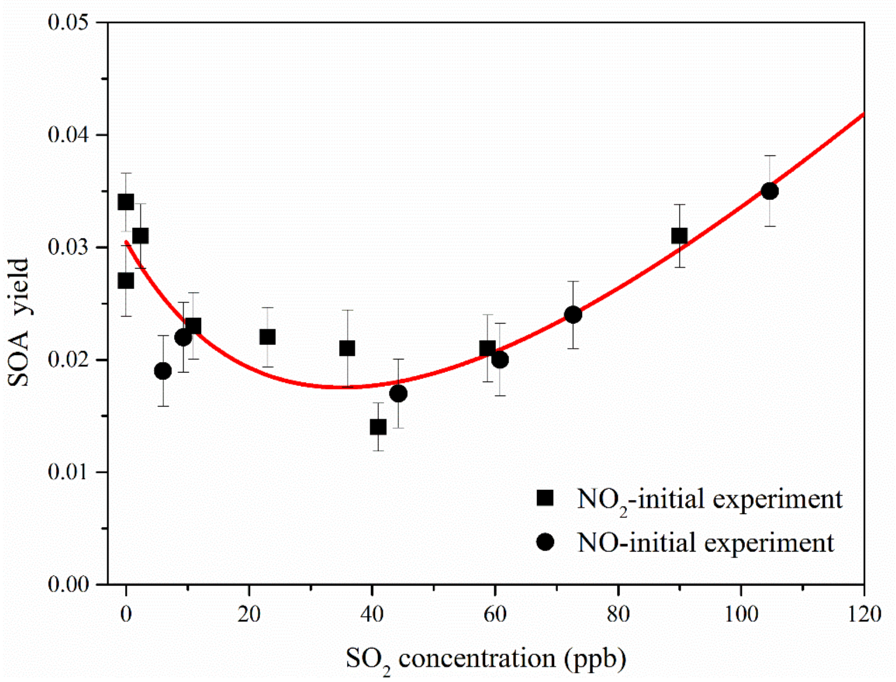

Figure 2: SOA yields of cyclohexene photooxidation in the presence of NOx at different initial SO_2 concentrations. Solid line is least-square fitting to the data. The error bars were determined on the basis of propagation of uncertainties arising in the ΔHC measurements, including GC calibration uncertainties propagation and the variance in the initial cyclohexene measurements.

Table 1 Experimental conditions for the photooxidation of cyclohexene/NOx/SO_2 system. All experiments were performed under dry conditions (relative humidity < 10%). ΔM_0 is the produced organic aerosol mass concentration and Y is the SOA yield.

Exp.	T (K)	SO_2 (ppb)	cyclohexene (ppb)	NOx (ppb)	cyclohexene/NOx	ΔM_0 ($\mu g\ m^{-3}$)	Y (%)
1[b]	308	0.0	596	122.0	4.9	57.0	2.66
2[b]	305	0.0	651	93.7	6.9	79.7	3.40
3[b]	309	2.4	553	95.7	5.8	62.6	3.15
4[a]	307	5.8	612	92.7	6.6	41.0	1.87
5[a]	309	9.3	599	93.5	6.4	48.1	2.23
6[b]	309	11.0	574	94.7	6.1	47.1	2.28
7[b]	309	23.0	514	90.5	5.7	42.6	2.30
8[b]	305	36.6	665	99.7	6.7	96.3	2.01
9[b]	308	40.8	472	91.4	5.2	22.6	1.33
10[a]	308	44.3	592	98.6	6.0	35.3	1.66
11[b]	305	55.0	497	113.0	4.4	77.3	2.16
12[b]	308	58.8	577	96.7	6.0	44.3	2.13
13[a]	309	60.8	626	102.0	6.1	43.9	1.95
14[a]	308	72.7	581	98.4	5.9	49.2	2.35
15[b]	306	90.0	543	99.6	5.4	102.0	2.62
16[a]	309	104.7	608	93.7	6.5	77.1	3.52
17[bc]	305	236.0	1048	198.0	5.3	-	-
18[bc]	306	93.7	1235	215	5.7	-	-

a: the experiment was initiated by NO.

b: the experiment was initiated by NO_2.

c: the formed particles were detected by ESI-HR-MS.

Section 3.4 Please distinguish between sulfonate and organosulfate and make this more clear in the text.

Sulfonate and organosulfates are all members of organosulfur compounds, with respective formulae $R\text{-}SO_3^-$ and $R\text{-}O\text{-}SO_3^-$, where R is an organic alkyl or aryl group. The two terms are distinguished in the text.

Figure 3: The line for ratio should be removed as it is based on very few data points. Figure 5: This figure is very confusing. Some data points are placed on top of each other. Information on the secondary axis (scale + label) is missing. Furthermore the uncertainty on the measurements should be presented. I suggest to make two figures instead of one.

The ratio line from Figure 3 was removed. The discussion at page 9 lines 30-32 was modified as:

"Figure 3 shows that the changes with initial SO_2 concentrations were not uniform between the SO_4^{2-} concentration and ΔSO_2, which indicates that besides SO_4^{2-}, other products were formed from SO_2."

In Figure 5, all the results were normalized to SOA mass. In order to compare the changes of the results with different initial SO_2 concentrations and the relationship between the different results at the same SO_2 concentration, all the results were set as 1 at the initial SO_2 concentration of 44 ppb. This led to some points being overlapped in Figure 5. For clarification, we used different colors, Figure 5 was remade as follows. We do not need the secondary axis here since the magnitudes of the band at 1100 cm^{-1} and SO_4^{2-} are similar. The results of ΔSO_2 were measured before DPLI sampling, both the volume of the chamber and the sampling time being inconsistent with those of the measurement of the band at 1100 cm^{-1} and SO_4^{2-}. Hence, in order to be more rigorous, we deleted the results on ΔSO_2 in Figure 5.

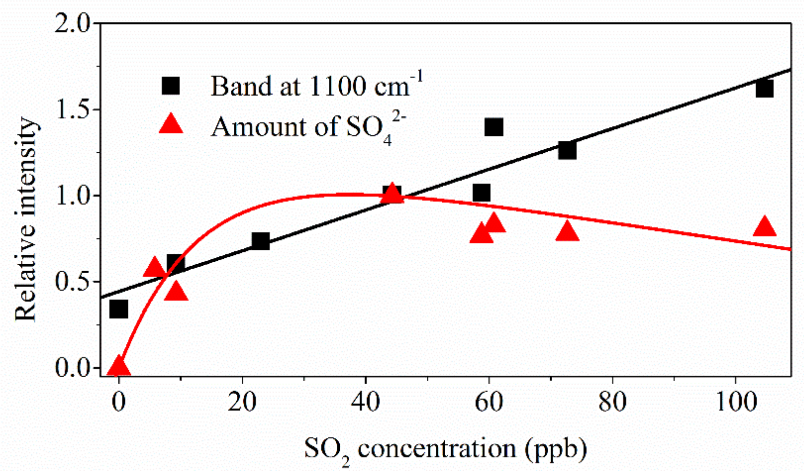

Figure 5: The relative intensity of the FTIR band at 1100 cm^{-1} (square) and the amount of SO_4^{2-} (triangle) normalized to SOA mass. The 1100 cm^{-1} band intensity and the amount of SO_4^{2-} were divided by the formed SOA mass. Subsequently, the results of both FTIR band at 1100 cm^{-1} and the amount of SO_4^{2-} divided by SOA mass were set to 1 when the initial SO_2 concentration was 44.3 ppb.

To clarify these changes, the statement at page 10 lines 23-24 was deleted. The statement at page 10 lines 27-28 was changed to "Figure 5 show the inconsistency between the FTIR band at 1100 cm^{-1} and the amount SO_4^{2-} as the initial SO_2 concentration, which implies that the 1100 cm^{-1} band originated not only from SO_4^{2-}, but also from other organosulfur compounds. These include organosulfates, which also have the S=O bond, and might contribute to the 1100 cm^{-1} band in the FTIR spectrum. The gap between the FTIR band at 1100 cm^{-1} and SO_4^{2-} can be attributed to the formation of organosulfates."

Page 10 line 30- and Figure 6: Did the composition and response of organosulfates vary between samples?

We investigated the composition and response of organosulfates between samples by performing another experiment with different initial SO_2 concentrations. We found that these did not vary much with the change in initial SO_2 concentrations, as can be seen in Figure S5 below.

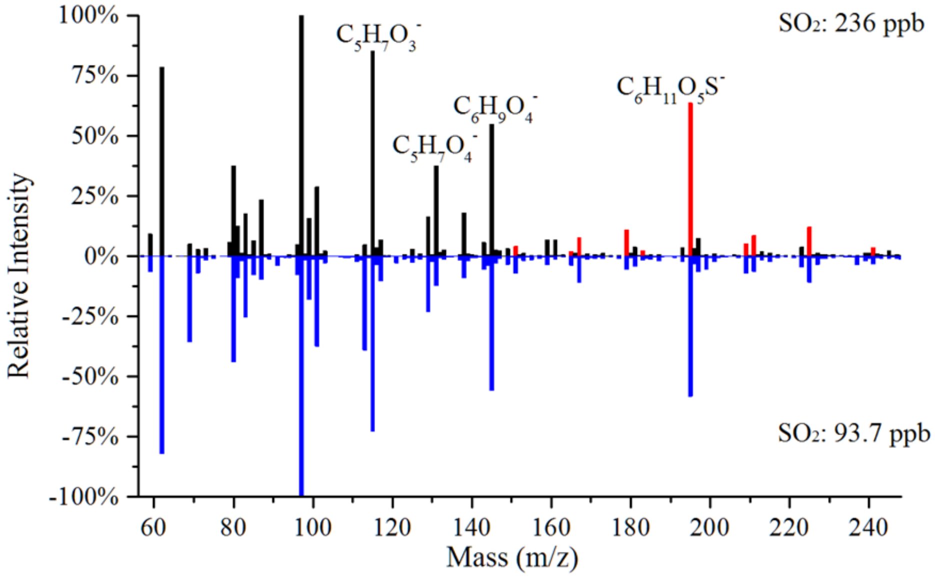

Figure S5: Comparison of SOA ESI-HR-MS spectra with different initial SO_2 concentrations.

The following was added at page 11 line 18 for clarification.

"The ESI-HR-MS spectra of particles formed from two different initial SO_2 concentrations are shown in Figure S3. We found no obvious difference in the composition and response of organosulfates with different initial SO_2 concentrations. The relative intensity of m/z = 97, which corresponds to sulfate was set to 100% in both ESI-HR-MS spectra. The relative intensities of the organosulfates peaks in both spectra were almost unchanged regardless of the initial SO_2 concentration, indicating that the organosulfates yield was associated with sulfate content. Our result is consistent with the results of Minerath et al. and Hatch et al. who observed an increase in organosulfates yield with increasing sulfate concentration (Minerath and Elrod, 2009; Hatch et al., 2011). These observations demonstrate that particle sulfate content is likely a key parameter influencing organosulfates formation."

In conclusion, some of the results are interesting, but the quality of the presentation and discussion is not adequate for publication of this work in ACP.

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