We thank reviewer#1 for the thorough comments and suggestions to improve the manuscript. Below we reply to the comments point by point. The comments are presented in black, our replies in red and the revised text in green.

General comments:

Qin et al describe measurements of PM composition at a suburban site in the Pearl River Delta, China. The paper is topically relevant to ACP. The data collection methods, analysis, and interpretation all seem appropriate. However, the authors do not clearly articulate what new knowledge is generated by this manuscript, aside from a relatively small (2 month) dataset collected in a city without previously published AMS data. A revised manuscript should place more emphasis on the unique contribution of this work.

We have revised our manuscript significantly to focus on how the pollutants highly urban downtown cities can impact adjacent "buffer" peripheries, especially on nitrate and secondary organic aerosols due to traffic emissions. The manuscript was also significantly revised to highlight the potential impacts of the study, as follows.

The introduction has been changed to highlight the need to understand how urbanized megacities such as Guangzhou affect their peripheral cities. The prevailing wind direction from Guangzhou to Panyu during the campaign provides a unique opportunity for such research. While our study focuses on the PRD area, its findings can potentially be useful to developing control strategies in many city clusters in China.

Introduction change to:

Traffic emissions are one of the main contributors to air quality deterioration in rapidly expanding urban China (Kelly and Zhu, 2016; Zhang et al., 2017). Pollutants emitted from vehicles, such as $NO_x$, volatile organic compounds (VOCs), black carbon (BC) and other traffic-related particulate matter (PM), have increased over last decades(Wang et al., 2013; Zhang et al., 2012). Apart from the primary PM, the oxidation of traffic-related gaseous pollutants such as $NO_x$, VOCs, SVOCs and LVOCs leads to the formation of particulate nitrate and secondary organic aerosols (SOA), exacerbating PM pollution. Multiple studies have shown that the concentrations and proportions of nitrate in PM have increased significantly in most Chinese megacities (Pan et al., 2016; Wen et al., 2015; Xue et al., 2014).

The rapid economic development in the Pearl River Delta (PRD) region has led to a rapid deterioration in air quality, especially due to a sharp increase in PM (Chan and Yao, 2008; Ho et al., 2003; Li et al., 2017). With densely populated cities including two megacities,

Guangzhou and Shenzhen, and other smaller cities, the PRD region is developing into a giant city cluster. There are, nevertheless, less populated areas between these cities that can serve as a "buffer" zone in terms of regional air quality. Due to air dispersion patterns and regulatory strategies, air pollutants in highly urbanized regions can greatly influence PM levels in peripheral regions. For example, highly-polluting vehicles such as heavy-duty diesel trucks are banned from the inner areas of many Chinese megacities during the day but are nevertheless active at night and in the early morning, especially on the peripheries of these megacities. This regulatory policy has resulted in nighttime peaks in vehicular pollutants, which have been commonly observed in many Chinese cities (Zhang and Cao, 2015). Although vehicle emissions are substantially reduced relative to the without control scenarios, it was still significantly higher emission density in East China than in developed countries with longer histories of vehicle emission control (Wu et al., 2016). Given the complex and non-linear processes involved in the secondary production of PM such as nitrate and organic aerosols, the impacts of these adjacent "buffer" zones on air quality are crucial. However, relatively little attention has been paid to how air pollutants in major cities affect these adjacent areas.

Panyu District lies directly south of central Guangzhou and experiences predominantly northerly/northeasterly winds between September and February (Zou et al., 2015). A campaign with a host of real-time instruments including an Aerodyne HR-ToF-AMS was conducted at a site in Panyu from November to December 2014 and provides a unique opportunity to explore how pollutants from the city center have impacted this adjacent "buffer" zone, especially in terms of nitrate and secondary organic aerosols. As will be shown, the mass fraction of nitrate increased as $PM_1$ increased, and the organic mass fraction was the highest among $PM_1$ species across all $PM_1$ levels. A comprehensive analysis of nitrate and organics and their interplay with meteorological parameters will be presented. Primary OA (e.g. HOA) and SOA from traffic-related VOC precursors will also be discussed. Our findings can provide useful information for regional emission control strategies covering urban and "buffer" areas and improving parameterization in air quality models.

I have several comments on Section 2.3. Overall, this section seems more like Results than Methods, since much of the section is dedicated to comparing the various constrained versus unconstrained PMF solutions.

Agreed. In order not to dilute the focus of this manuscript, the whole section has been moved to the SI and has been revised according to the specific comments.

The whole section has been changed to:

In the main text:

Source apportionment for OA was performed using the newly developed ME-2 and controlled via the SoFi interface coded in Igor Pro (Canonaco et al., 2013). The procedure allows an effective exploration of the solution space, a more objective selection of the optimal solution, and an estimation of the rotational uncertainties (Canonaco et al., 2013; Crippa et al., 2014; Elser et al., 2016; Fröhlich et al., 2015; Paatero and Hopke, 2009). We only considered ions up to m/z 100 due to the low signal-to noise ratios of larger ions. Fully unconstrained runs (PMF) were first explored. However, the three-factor solution suffers from the mixing of the HOA factor with the cooking organic aerosols (COA) factor (Fig. S4) as well as the SVOOA factor with the biomass burning related organic aerosols (BBOA) factor. The four-factor solution splits highly oxidized low-volatile OOA (LVOOA) into two sub-factors (Fig. S5). The inclusion of additional factors still cannot resolve pure primary OA factors (HOA, COA and BBOA) as they may have similar time series or profiles. Zhang et al. (2013) also report that the PCA-resolved HOA can be affected by cooking emissions with a distinct noontime peak in Beijing in spring and summer. However, by introducing a priori information of source profiles for HOA, COA and BBOA, the ME-2 provides additional control over the rotational ambiguity (Canonaco et al., 2013; Paatero and Hopke, 2009). The a value, ranging from zero to unity, stands for the percentage by which each m/z signal of the final solution spectra may differ from the anchor. A value of 0 means no deviation is allowed, while a value of 1 means 100% deviation is allowed. The OA source apportionment was performed by constraining the source profile of HOA, COA and BBOA with a values of 0.1, 0.2, 0.3 for HOA, COA, and BBOA respectively. The HOA source profile was extracted from the data set using a separate PMF run in selected time series with peaks in organic mass concentration. The COA reference was adopted from the Mong Kok campaign in Hong Kong (Lee et al., 2015) while the reference BBOA profile was adopted from MILAGRO (Aiken et al., 2009). A detailed discussion can be found in the supplementary information (Text S2).

In the SI:

Text S2: Source apportionment for OA

Several source profiles from previously reported HR-ToF-AMS data with different a values were explored. However, some ions were missing from the reference source profile when

compared with our dataset. For these ions, the signal intensities were estimated based on the unit-mass-resolution (UMR) source profile from the average of multiple ambient data sets (Ng et al., 2011) as follows:

$$I_{m/z}=k *I_{UMR(m/z)}-I_{HRrest(m/z)} \tag{1};$$

$$k= I_{UMR(total)}/I_{HR(total)} \tag{2}.$$

where $I_{m/z}$ is the signal intensity of the missing ions in the reference profile; k is a correction factor, derived from the ratio of total signal intensities in the UMR and HR profiles, which accounts for the difference in total signal intensity between the profiles; $I_{UMR(m/z)}$ is the total signal intensity at UMR level for the missing ions in the UMR profile; and $I_{HRrest(m/z)}$ is the sum of the signal intensities of the rest of the ions from HR reference profile that shares the same integer m/z as the missing ions. For these ions, whose intensities are derived from the above equation, an a value of 1 (100% deviation) was used.

We used the ME-2 solver with reference mass spectra to tackle the problem of mixing sources in our PMF runs. The HOA the Paris campaign (Crippa et al., 2013) as an input reference however were not fitted in our dataset. The resolved-HOA factor contributed no more than 5% of total OA, and only exhibited a small morning rush peak in the diurnal pattern (Fig. S6). Furthermore, another factor was observed to share similar features with the HOA mass spectrum. This factor exhibited clear rush-hour peaks during the morning and late afternoon, as with traffic-related pollutants (e.g. $NO_x$ and BC). Also, the mass fraction of this factor is 3-4 times higher than the resolved-HOA factor. Alternatively, we directly extracted a local HOA source profile ($HOA_{loc}$) from the data set using a separate PMF run in selected time series with peaks in organic mass concentration. We selected the high OA events in order to capture some periods with potentially high HOA. As shown in Fig S7, the concentrations of other traffic tracers (for example BC and NOx) were also high. A similar approach of extracting factors in unconstrained PMF and subsequent combination of duplicate factors was also used in a previous study (Fröhlich et al., 2015). (Fröhlich et al., 2015)(Fröhlich et al., 2015) Using the peaks or only one peak yield similar mass spectra for local HOA factors with $R_{uc}$=0.99 (Fig. S7a, S7b). The HOA profiles remained the similar with even more factors (Fig. S7c). We then used the HOA source profile obtained from all of the short peaks as the input HOA source profile (LHOA therein). However, constrained LHOA alone cannot resolved an environmentally reasonable solution either (Fig. S8). We then tried to added other primary OA

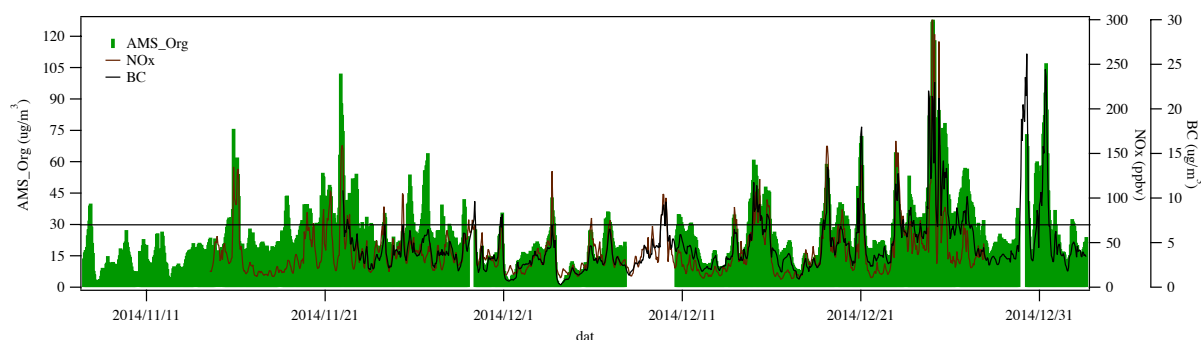
source profiles as constrain. For COA source profile, we chose the COA source profile from the Hong Kong campaign (HK) to constrain our ME-2 runs as cooking styles and ingredients in Guangzhou are similar to those in Hong Kong. For the BBOA factor, we used the reference BBOA profile from MILAGRO (Aiken et al., 2009). The resolved BBOA factor tracked well with its tracer ion ($C_2H_4O_2^+$) and potassium ($K^+$) in time series.

  A four-factor (HOA, COA, BBOA and a free factor) solution had a higher $Q/Q_{exp}$, while a six-factor (HOA, COA, BBOA and three free factors) solution seemed to split OOA factors without obvious physical meaning. An five-factor solution with a values of 0.1, 0.2, 0.3 for $HOA_{local}$, $COA_{HK}$, and $BBOA_{MILAGRO}$, respectively, was finally adopted. The a values for these POA factors were also in line with previous ME-2 studies (Canonaco et al., 2013; Crippa et al., 2014; Fröhlich et al., 2015). We further run the ME-2 with the same conditions with 10 runs to explore the stability of solution. The time series and mass spectra for these 10 runs were quite steady (Fig. S10). The final solution came from results obtained with averaging these ten runs. Two oxygenated organic aerosol factors, SVOOA and LVOOA, were assigned based on their degree of oxygenation.

Specific comments for this section include:

1. The main goal of the text from _line 131-189 is buried. My interpretation is that the authors are describing their efforts to separate an HOA factor from a cooking (COA) factor, and that these two factors do not cleanly "fall out" from unconstrained PMF (e.g., moving from 3 to 4 to 5 unconstrained factors starts splitting OOA factors rather than splitting HOA and COA).
Yes. Because the PMF solutions showed signs of source mixing (viz. HOA mixed with COA, BBOA mixed with OOA), we therefore used ME-2 to constrain the primary OA source. The above statement has been added to this section.

2. I am confused by Lines 167-175. I think the authors performed a separate unconstrained PMF analysis of high concentration events, assumed that the high concentration peaks are dominated by HOA, and used the resulting HOA factor from these high events to constrain the more general HOA mass spectrum. If this is the case, the description is extremely confusing (I had trouble determining if lines 167-175 describe a single PMF solution approach or multiple), and the utility of this approach is not described well. Also, the authors do not describe what evidence they used to attribute the high concentration events to traffic/HOW.

Yes. We performed a separate unconstrained PMF analysis of high OA concentration events to capture a local HOA factor. These high OA events is likely affected by high HOA, so that it is possible to resolve a HOA factor from these high OA peaks by perform a PMF analysis. As shown below, the concentrations of other traffic tracers (for example BC and NOx) were also high, indicating the potential influence by high HOA. A similar approach of extracting factors in unconstrained PMF and subsequent combination of duplicate factors was also used in a previous study (Fröhlich et al., 2015).

The original text has been change to:

Alternatively, we directly extracted a local HOA source profile ($HOA_{loc}$) from the data set using a separate PMF run in selected time series with peaks in organic mass concentration. We selected the high OA events in order to capture some periods with potentially high HOA. As shown in Fig S7, the concentrations of other traffic tracers (for example BC and NOx) were also high, indicating the potential influence by high HOA. A similar approach of extracting factors in unconstrained PMF and subsequent combination of duplicate factors was also used in a previous study (Fröhlich et al., 2015). (Fröhlich et al., 2015)(Fröhlich et al., 2015) Using the peaks or only one peak yield similar mass spectra for local HOA factors with $R_{uc}$=0.99 (Fig. S7a, S7b). The HOA profiles remained the similar with even more factors (Fig. S7c).

3. Lines 190-201 describe an "optimum" solution (the authors should be careful with this word because their source apportionment was not formally optimized) and compare 10 runs of that optimum solution. What is changing between these 10 runs? It is not explained why the 10 runs might result in different answers.

Changed from "optimal" to "final". The additional ten runs were performed with the same setting as used in the final solution to confirm the stability of the final solution.

4. Does the final PMF solution incorporate the high concentration events, or exclude them? E.g., does the pie chart in Figure 5 include the high concentration events?

Yes. The high concentration events are included in the final solution.

Comments on Section 3.3 - Formation of SOA

1. The main idea of this section seems to be that SOA formation is dominated by gas-phase oxidation chemistry (hence a strong SOA-to-Ox relationship) rather than heterogeneous or aqueous oxidation pathways. This should be stated more clearly.

We have classified the above sentence as suggested.

In line 301-307:

The evolution of AMS OA factors has been used to infer SOA formation via photochemical oxidation in Hong Kong (Lee et al., 2013; Li et al., 2013; Qin et al., 2016). Fig. 11a shows that SOA (SVOOA+LVOOA) increased as $O_x$ increased during the day (7:00 to 18:00) in both November and December. The SOA concentrations do not show clear correlations with temperature or liquid water content in aerosols (Fig 11b, 11c). When taken together, this suggests that SOA formation in our study is dominated by gas-phase oxidation chemistry rather than heterogeneous or aqueous oxidation pathways.

2. The SOA/Ox slope is higher than other locations (Table 2). The humid tropical climate is given as the reason. Can this claim be substantiated more quantitatively? Another possible explanation is that emissions are different in the PRD than the North American cities listed in Table 2. The authors should address possible explanations besides climate.

Agree. The higher emissions in the PRD than the North American cities may also lead to the above results. Liu et al.(2015) and Deng et al.(2017) reported that the emission factor for BC and POA and production factor for SOA under photochemical aging were significantly higher than those studies in Europe and the US and those of light-duty gasoline vehicles.

The above statements have been added to the revised manuscript in Line 313-318.

3. One could argue that Figure 7b shows that SOA formation was essentially independent of temperature for this city over the temperature range sampled in November and December.

Yes. The temperature effect on gas-particle partitioning did not appear to be important in this study. As we stated in the original manuscript:

Higher temperatures should favor the partitioning of SOA to the vapor phase, but higher SOA mass loading was associated with higher temperature in our case (Fig. 7b). In November, LWC stayed in a relatively narrow range (Fig. 7c) when SOA experienced drastic changes. In December, LWC was lower than that in November. Yet, SOA increased as LWC increased. The enhanced LWC may facilitate the SOA formation in December, but such influence was not clear in November. Furthermore, there is no correlation between SOA/OA and LWC in either month.

Other comments

1.      Line 91 - Please provide details for the manufacturer of the MARGA

Metrohm applikon. Added in the revised manuscript line 81.

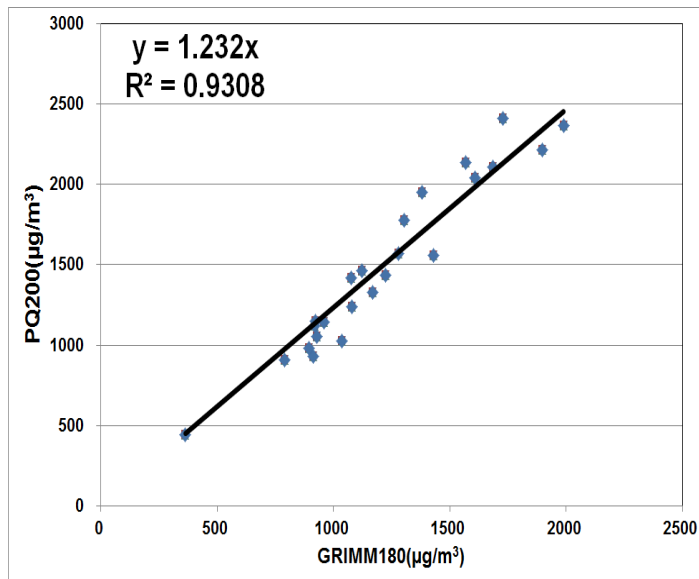
Other data presented in this work were obtained from collocated instruments, which included a Grimm 180 for $PM_{2.5}$, a thermo-optical ECOC analyzer (Sunset Laboratory Inc.), a Magee AE33, a dual spot filter based instrument for black carbon (Drinovec et al., 2015), a gas analyzer system (Teledyne Instruments), and a monitor for aerosols and gases in ambient air (MARGA, Metrohm applikon) with a $PM_{2.5}$ cutoff.

2.      Line 89 - what method does the Grimm 180 use for determining PM mass?

The Grimm monitor was based on particle scattering measurements or called optical particle counter. A $PM_{2.5}$ cut-off was placed before the GRIMM 180. We integrated the hourly Grimm 180 data into daily basis, and then compared with the daily $PM_{2.5}$ mass concentration with quartz filter measurement (PQ200 Ambient Air Particulate Sampler with a $PM_{2.5}$ cut-off). Added in the revised SI (Text S1).

3.      Line 92-93 - Please provide more detail on how the Grimm 180 data were corrected with quartz filter data. Did the correction use bare quartz filters only?

Hourly Grimm 180 data was integrated into daily basis, and then compared with quartz filter measurement (PQ200 Ambient Air Particulate Sampler with a $PM_{2.5}$ cut-off). The daily mass concentration by quartz filter sampling was then obtained gravimetrically. The PQ200 and Grimm 180 measurements were in good agreement with $R^2$ of 0.93, with a slope of PQ200 vs. Grimm 180 data of 1.232. We then used the linearly fitted parameter to correct the hourly

Grimm 180 data to derive the hourly PM$_{2.5}$ mass concentration. Added in the revised SI (Text S1).

4.      Section 2.2 needs to be broken into multiple paragraphs.

Changed as suggest.

5.      Line 118 - Need citation for the Middlebrook paper

Changed as suggest.

6.      Figure 2b - label says NH3, should be NH4

Changed as suggest.

7.      Line 343 - it is not clear what "this method" and Method 1 refer to, specifically.

"this method" or Method 1 refer to the ON estimation based on NO$^+$ to NO$_2^+$ ratio. Revision in wording has been made.

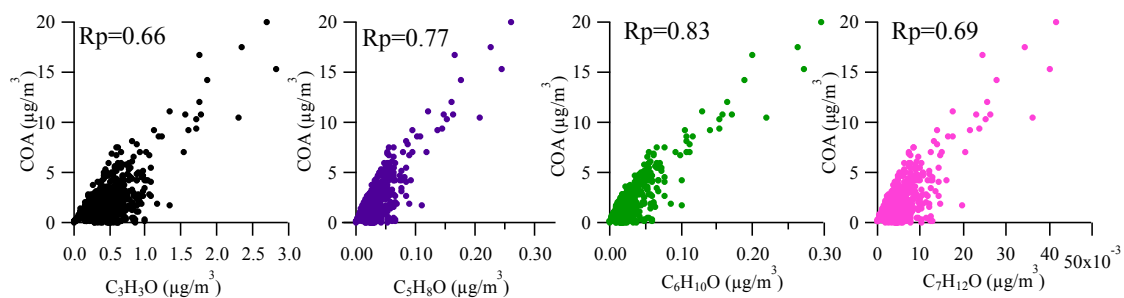
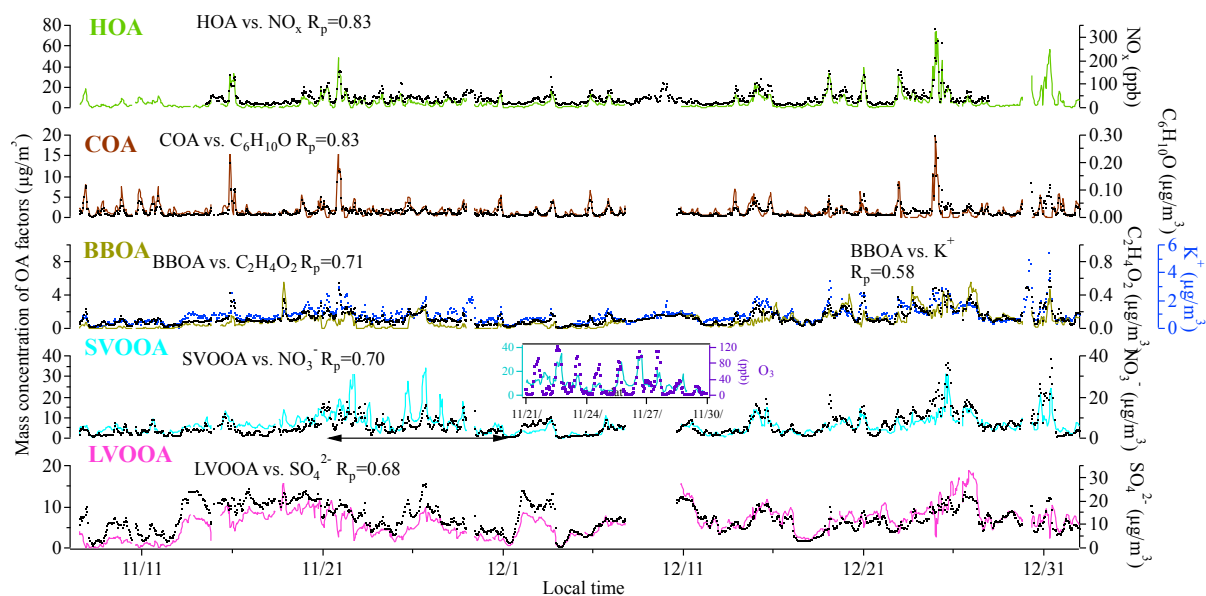
Change to:

The method using NO$^+$/NO$_2^+$ (Method 1, hereinafter) was adopted in this study to estimate the contributions of inorganic and organic nitrates.

8.      Figure 6 - In December it seems that the mean HOA concentration is often outside of the 75th percentile. Are the diurnal patterns in this month skewed by a few very high concentration days?

Yes. The mean concentrations were greatly affected by a few very high concentration days, especially from 0:00-12:00. There were several significant HOA peaks during December (19 Dec., 21 Dec., 23 Dec, 25 Dec., and 31 Dec., as can be seen from Fig. 4). Furthermore, from Fig. 5c, HOA almost linearly increased with OA concentration, and played a significant contribution to the high OA situation. This observation highlights the importance of traffic control in mitigating the high PM pollution in "buffer" areas.

9. Figure 6 - It looks like the average LVOOA is about 1 ug/m^3 higher in December than November. Given that there is less photochemical activity, does this seem physically reasonable? Or could it be an artifact of the source apportionment?
We analyzed photochemical activities based on locally measured parameters, e.g. $O_x$ etc. Therefore, it is the local photochemical activities that were lower. LVOOA in Dec can be from long range transport.

10. Fig 4 - C3H3O does not seem like a great COA tracer. Is there a scatter plot showing the relationship of COA with this tracer ion?
Agree. The scatter plot showing the relationship of COA and $C_3H_3O$ and other COA tracer (Sun et al., 2011) is shown as below. Indeed, $C_3H_3O$ does not seem like a great COA tracer. Rather, $C_6H_1O$ tracks better with COA.

The original text has been change to:
The time series of COA tracked well with one of its tracer ions, $C_6H_{10}O^+$.
And the time series plot has been changed to:

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