

Responses to Reviewers

We thank you very much for your constructive comments and suggestions, which are very helpful to improve the scientific merits of this manuscript. We have carefully revised this manuscript according to your comments, and the amendments in the revised manuscript were presented in red font. Enclosed please find the point-by-point responses to the reviewers' comments. **This text in red is revision in the revised manuscript.** The line numbers in the response are from the revised manuscript.

Responses to Referee #1's comments

General comment: This manuscript reports the photochemical aging of PM₁ components in a suburban site in Beijing. The discussions focus on the change of AMS-measured PM₁ composition as a function of a photochemical clock calculated based on the toluene/benzene ratio. There is a major error in the analysis. The variation in the emission ratio of toluene/benzene is completely neglected when using the toluene/benzene ratio to calculate the photochemical clock. The air masses arriving at the measurement site likely originate from various sources (industrial plants vs vehicle emissions vs solvent use), which have different initial toluene/benzene ratio. The mixing of air masses with different origins will introduce significant uncertainties in the analysis. Further, the toluene/benzene ratio is influenced not only by photochemical aging but also by mixing processes. This has been extensively discussed in the literature [McKeen and Liu, 1993; Parrish et al., 2007]. Lastly, it is not clear how the initial value of toluene/benzene ratio (i.e., 3.3) is determined in this study. To ensure the analysis is

meaningful, the authors may consider to perform back-trajectory analysis and only analyze the measurements with similar origins. The authors may also estimate the photochemical age by using NO_y/NO_x ratio and compare it with toluene/benzene ratio. Besides the issues with toluene/benzene clock, many conclusions are drawn without solid evidence. Examples will be listed below. Overall, I cannot recommend publication in its current format.

Reference McKeen, S. A., and S. C. Liu (1993), Hydrocarbon ratios and photochemical history of air masses, *Geophysical Research Letters*, 20(21), 2363-2366, doi:10.1029/93gl02527. Parrish, D. D., A. Stohl, C. Forster, E. L. Atlas, D. R. Blake, P. D. Goldan, W. C. Kuster, and J. A. de Gouw (2007), Effects of mixing on evolution of hydrocarbon ratios in the troposphere, *Journal of Geophysical Research: Atmospheres*, 112(D10), doi:10.1029/2006jd007583.

Response: Many thanks for your constructive comments and valuable suggestions, which would be much helpful to improve the scientific merits of this manuscript. The concerns raised by you have been carefully addressed in the revised manuscript.

According to your suggestion, 72 h back trajectories have been performed using the NOAA-HYSPLIT4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxier and Hess, 1998). Four times for each day, 00:00, 06:00, 12:00 and 18:00 UTC, terminating in a height of 50 m, and 112 trajectories were calculated and then used to do the air mass cluster analysis. A total of 4 backward-trajectory clusters were identified (Clusters 1-4, Fig. R1), which indicated that more than half (Cluster 2, 64%) of the observation period is dominated by the southern air mass.

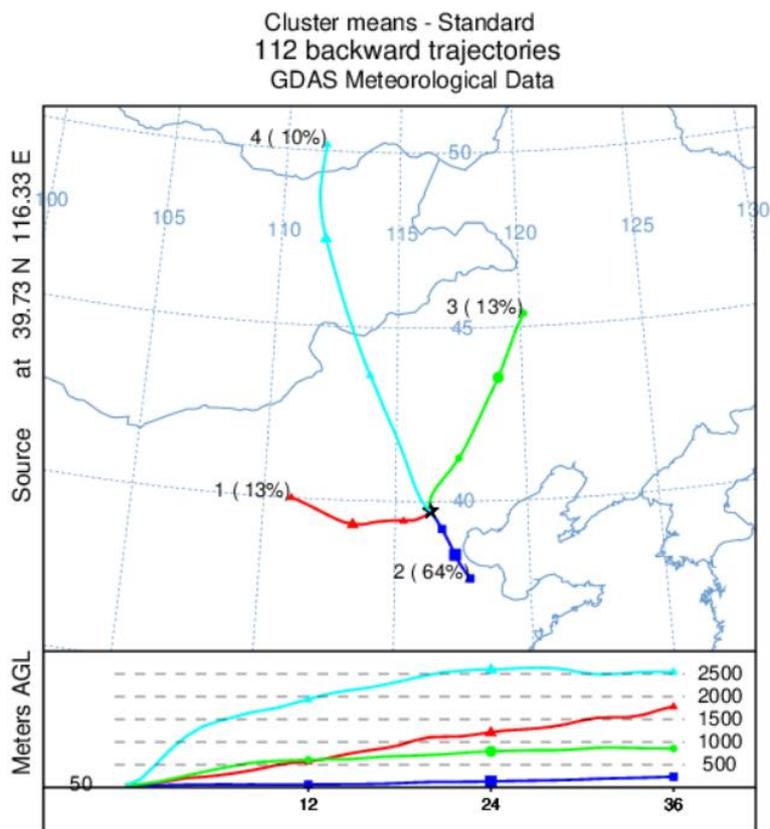


Figure R1. Mean back trajectories for 4 trajectory clusters arriving at Beijing Institute of Petrochemical Technology (BIPT) from different height. **(This figure was added as Figure S5 in the revised SI)**

To improve the reliability of photochemical age (t_a) as the surrogates of atmospheric oxidation capacity, the observation period included by Cluster 2 was adopted, which had the similar emission sources. Meanwhile, the influence of fresh emission during morning and evening rush hours was minimized by using the data observed from 10:00 to 18:00, as discussed by Qin et al. (2016) (Qin et al., 2016). In order to better compare with other studies, the OH concentration has been assumed as 1.5×10^6 molecule cm^{-3} in the revised manuscript. Meanwhile, the emission concentration ratio of toluene and benzene has been estimated to be 2.71 ± 0.39 ppb ppb^{-1} (Fig. R2) according to the methods described by de Gouw et al. (2017) (de Gouw

et al., 2017), where the ratio at nighttime was used to avoid the impact of photochemistry and boundary layer dilution on the emission ratio. This ratio is slightly higher than the value that calculated from the emission ratio of toluene and benzene (~2.2) in Beijing (Yuan et al., 2012), which might be related to the extensive utilization of solvents containing toluene for painting and printing in Daxing District, Beijing (Yuan et al., 2010). Uncertainties in estimating the initial emission ratio of toluene to benzene do exist and the influence to the analysis was discussed by Yuan et al. (2012), and they estimated that the uncertainties of the emission ratios were less than 50%. The concentration ratio of toluene and benzene during this observation period (i.e. Cluster 2) was given in Fig. R2. During this observation period, the average ratio of toluene to benzene is about 1.18, which is consistent with previous studies in Beijing (Liu et al., 2017; Wang et al., 2016; Wang et al., 2012).

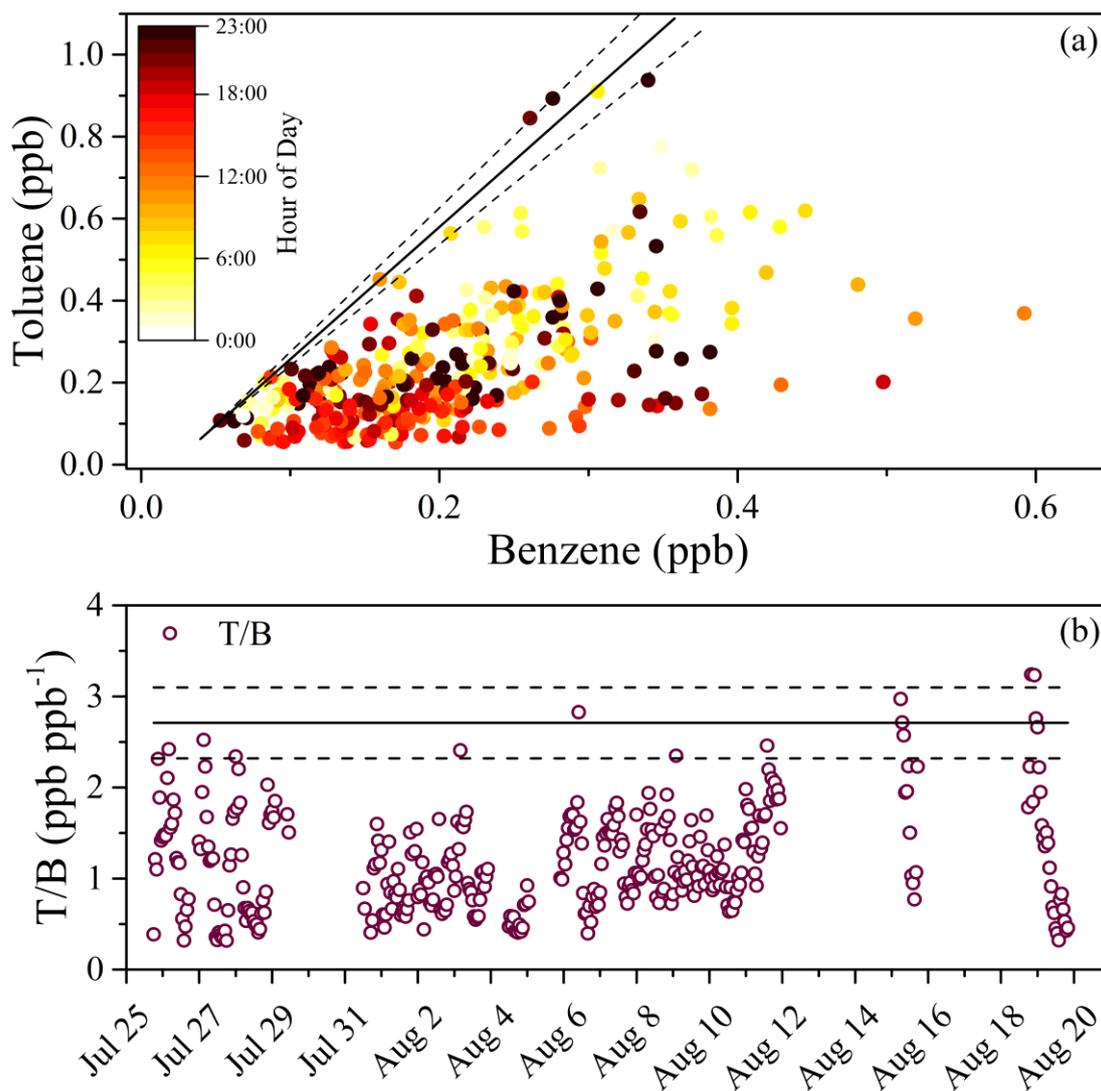


Figure R2. (a) Determination of the emission ratio of toluene vs. benzene. The scatter plot of toluene vs. benzene color-coded by the time of day, (b) Time series of concentration ratio of benzene and toluene during the observation period included by Cluster 2. **(This figure was added as Figure S6 in the revised SI)**

Revision in the manuscript:

Section 2.3.3 Photochemical age had been rewritten in the revised manuscript as follows:

In this study, the equivalent photochemical age (t_a) was calculated to characterize the OH exposure dose of the air mass, as expressed in the following equation. This

method had been widely used to provide useful information on the photochemical process in the atmosphere (Yuan et al., 2012; Parrish et al., 2007; de Gouw et al., 2005; McKeen et al., 1996).

$$t_a = \frac{1}{[\text{OH}](k_{\text{toluene}} - k_{\text{benzene}})} \times \left[\ln\left(\frac{[\text{toluene}]}{[\text{benzene}]}\right)_0 - \ln\left(\frac{[\text{toluene}]}{[\text{benzene}]}\right) \right]$$

where [OH] is the average OH concentration in the ambient air and is assumed as 1.5×10^6 molecule cm^{-3} in order to better compare with other studies (Mao et al., 2009; Chu et al., 2016; Liu et al., 2018). k_{toluene} and k_{benzene} are the rate constants for the reactions of OH with toluene (5.63×10^{-12} cm^3 molecule $^{-1}$ s $^{-1}$) and benzene (1.22×10^{-12} cm^3 molecule $^{-1}$ s $^{-1}$), respectively (Atkinson and Arey, 2003). $\left(\frac{[\text{toluene}]}{[\text{benzene}]}\right)_0$ is the emission concentration ratio of toluene and benzene before aging in the atmosphere, and was determined to be 2.71 ± 0.39 (Fig. S6, in the Supplement) according to the methods described by de Gouw et al. (2017), where the ratio at nighttime was used to avoid the impact of photochemistry and boundary layer dilution on the emission ratio.

$\frac{[\text{toluene}]}{[\text{benzene}]}$ is the measured concentration ratio of toluene and benzene, as given in Fig. S6. Based on the back trajectories and air mass cluster analysis (Draxier and Hess, 1998) (Details can be found in SI), an observation period (Cluster 2, Fig. S5) with the similar emission sources was adopted. Furthermore, the data observed from 10:00 to 18:00 was used to minimize the influence of fresh emission during morning and evening rush hours on t_a , as discussed by Qin et al. (2016). In this study, t_a was determined to be in the range of 4-90 h, which is comparable to previous studies (up to 72 h) conducted in China (Peng et al., 2016; Chu et al., 2021). Meanwhile, t_a had a higher level at noon,

which is similar to the diurnal trend of OH radicals observed in summertime of Beijing (Tan et al., 2019).

Supplement, Details of back trajectories analysis have been added to the SI as follows:

S3. Back trajectories analysis

72 h back trajectories have been performed using the NOAA-HYSPLIT4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxier and Hess, 1998). Four times for each day, 00:00, 06:00, 12:00 and 18:00 UTC, terminating in a height of 50 m, and 112 trajectories were calculated and used to do the air mass cluster analysis. A total of 4 backward-trajectory clusters were identified (Clusters 1-4, Fig. S5), which indicated that more than half (Cluster 2, 64%) of the observation period is dominated by the southern air mass. This observation period included by Cluster 2 was adopted to analyze the role of t_a in PM_{10} formation and evolution.

Comment 1: Major comments 1. Below I list some examples of the bold conclusions drawn on weak evidence or missing links between evidence and conclusion. (1) Page 14 Line 256. Why would “ NO_x concentration decreased with increasing age” implied “the formation of O_3 is in the VOC sensitive regime”?

Response: As shown in Fig. S5 mentioned in Lines 250-251, O_3 concentration significantly increased with increasing t_a , while NO_x concentration decreased with increasing t_a , this indicated that the change trend of O_3 and NO_x was opposite. Fig. R3 intuitively presented the relationship between NO_x and O_3 , an increase in NO_x resulted in a lower O_3 concentration, which was related to the reduced production of RO_2 . These

indicated that the formation of O₃ is in the VOC-sensitive regime during this observation (Li et al., 2018; Sillman, 1999; Wang et al., 2017).

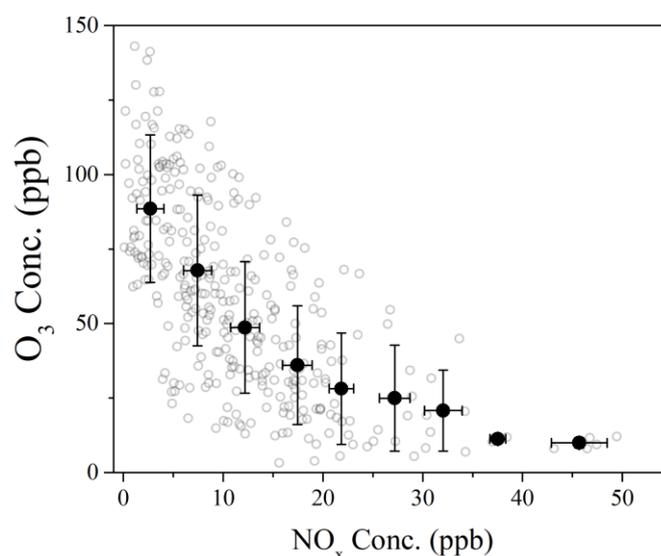


Figure R3. Relationship between NO_x and O₃ concentration during this field observation. **(This figure was added as Figure S9b in the revised SI)**

Revision in the manuscript:

Lines 278-279 (in revised version, both here and below), Change “Considering that the concentration of NO_x decreased with increasing t_a (Fig. S6)” **to:** “Further analysis showed that the concentration of O₃ significantly decreased with increasing NO_x in this observation period (Fig. S9b)”

Comment 2: (2) Page 16 Line 298-300. “The boosted formation of Cl⁻ with increasing age” is not sufficient to infer “the combination Cl⁻ with NH₄⁺ in the form of NH₄Cl”.

Response: Thanks for your comment. According to the suggestion of Reviewer #2, considering NH₄ and Cl are not produced through photochemistry, the discussion on the relationship between NH₄ and Cl and photochemical age had been removed in the

revised manuscript.

Revision in the manuscript:

Lines 298-300, Delete “The formation of Cl^- was also boosted with increasing t_a , similar to that of NH_4^+ , which confirmed the combination Cl^- with NH_4^+ in the form of NH_4Cl (Chen et al., 2020).”

Comment 3: (3) Page 12 Line 236. It is claimed that “longer aging time always corresponds to a higher concentration of O_3 and [...]”. This bold statement is not true, as the O_3 concentration depends on a number of other factors.

Response: We agree with the reviewer that there are many factors will significantly affect O_3 concentration, as reviewed by Wang et al. 2017 (Wang et al., 2017). During this field observation, we found a positive correlation between photochemical age (t_a) and O_3 concentration (Fig. R4), therefore, we inferred that higher concentration of O_3 might be related to the longer aging time during the summertime with strong solar radiation. This sentence had been rewritten in the revised manuscript.

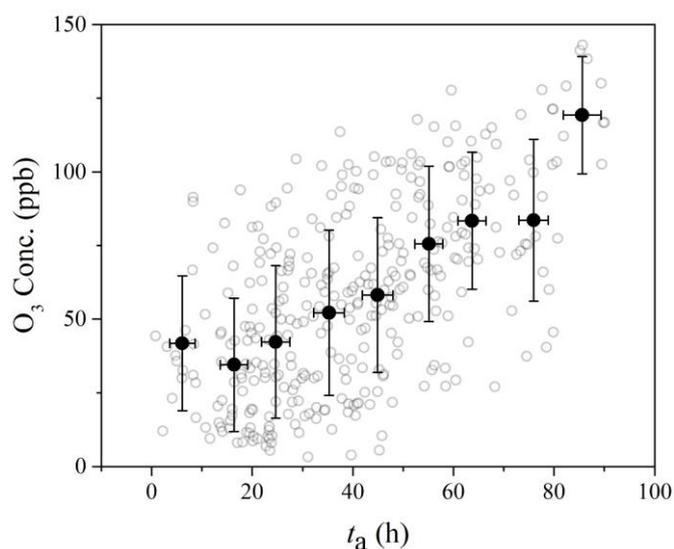


Figure R4. Relationship between photochemical age (t_a) and O_3 concentration during this field observation. The data are binned according to the value of t_a (10 h increment).

(This figure was added as Figure S9a in the revised SI)

Revision in the manuscript:

Lines 259-262, Change “longer aging time (t_a) always corresponds to a higher concentration of O_3 and stronger atmospheric oxidation during summertime.” **to:**

“Therefore, longer aging time (t_a) implies that more VOCs are consumed and more RO_2 are formed. Consequently, the transform cycles of NO_x and the accumulation of O_3 would be accelerated under this conditions (Wang et al., 2017).”

Comment 4: (4) Page 18 Line 331-333. The fraction of NO_3^- in PM_{10} shows a decreasing trend with increasing age, which is attributed to lower concentration of NO_x at higher age. This reasoning is flawed. The lower NO_x concentration at higher age is likely because NO_x conversion to long-lived NO_y species. The decreasing fraction of NO_3^- in PM_{10} could be due to enhanced production of other PM_{10} components, such as OA or SO_4 .

Response: Thanks for your comment. Indeed, the mass concentration and percentage of OOA and sulfate were greatly enhanced with the increasing photochemical age, this will lead to the decreasing fraction of nitrate in PM_{10} . This sentence has been revised.

Revision in the manuscript:

Lines 371-373, Change “which could be attributed to the lower concentration of NO_x , higher temperature and lower RH, as mentioned above” **to:** “which could be due to the

enhanced production of OOA and sulfate as t_a increased, as well as the dilution effects and evaporation of ammonium nitrate (DeCarlo et al., 2008; Nault et al., 2018)”

Comment 5: 2. Another major issue is that only the binned data are show in the species concentration vs age plots. All raw data should be included. Also, I want to point out that the species concentration has a very large variation under the same age bin. I suspect there is any correlation between species concentration and age in the raw data.

Response: Thanks for your valuable suggestion. All raw data of each species have been included in the form of scatter plot in the revised manuscript. The scatter plot of these species also presented the similar trend as t_a increased with the binned data.

Revision in the manuscript:

Figures 2-3 and Figures 5-6 had been modified in the revised manuscript as follows:

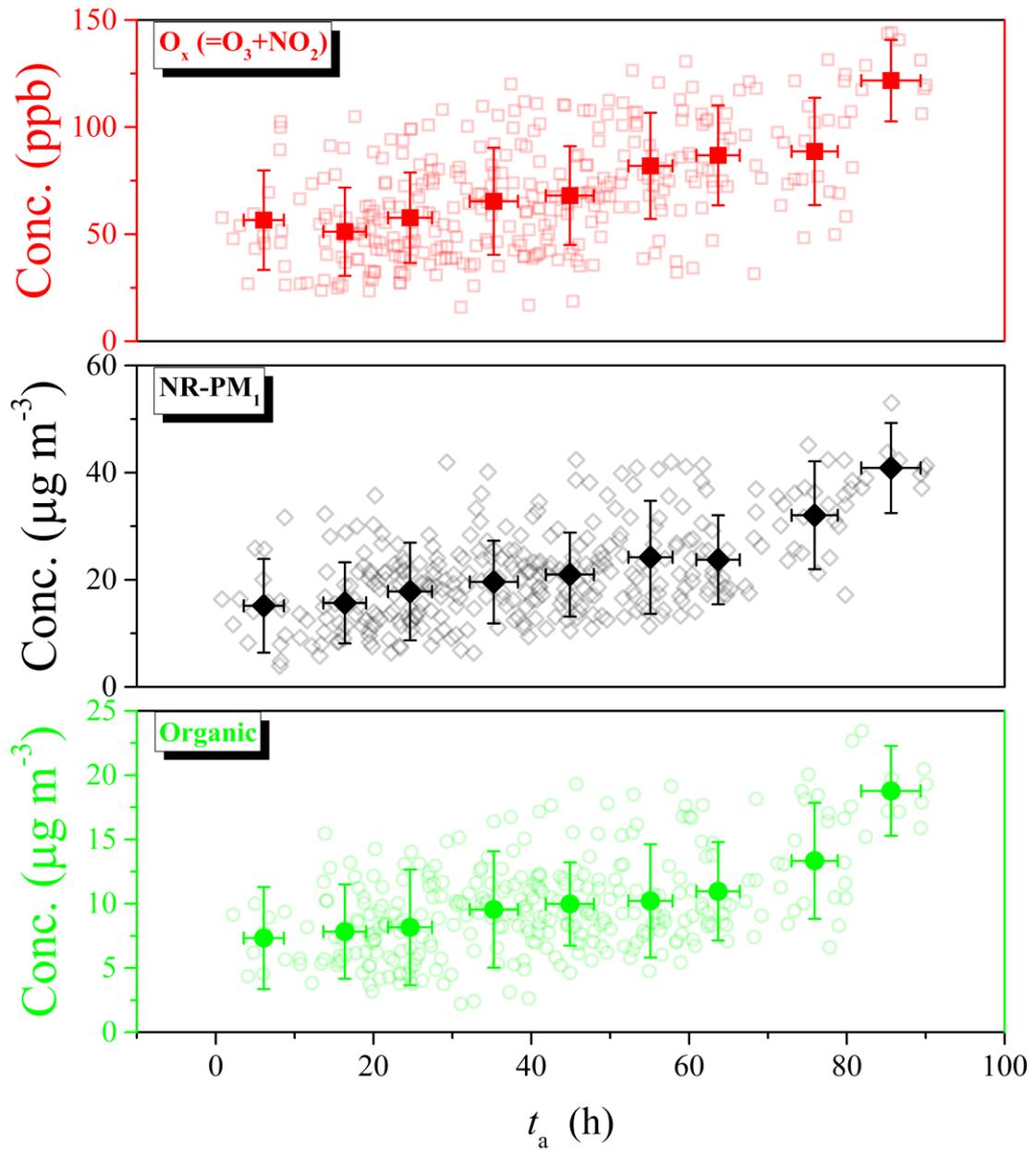


Figure 2. Relationship between photochemical age (t_a) and total oxidant (O_x), NR- PM_{10} concentration, as well as organic concentration during the observation period included by Cluster 2. The data are binned according to the value of t_a (10 h increment).

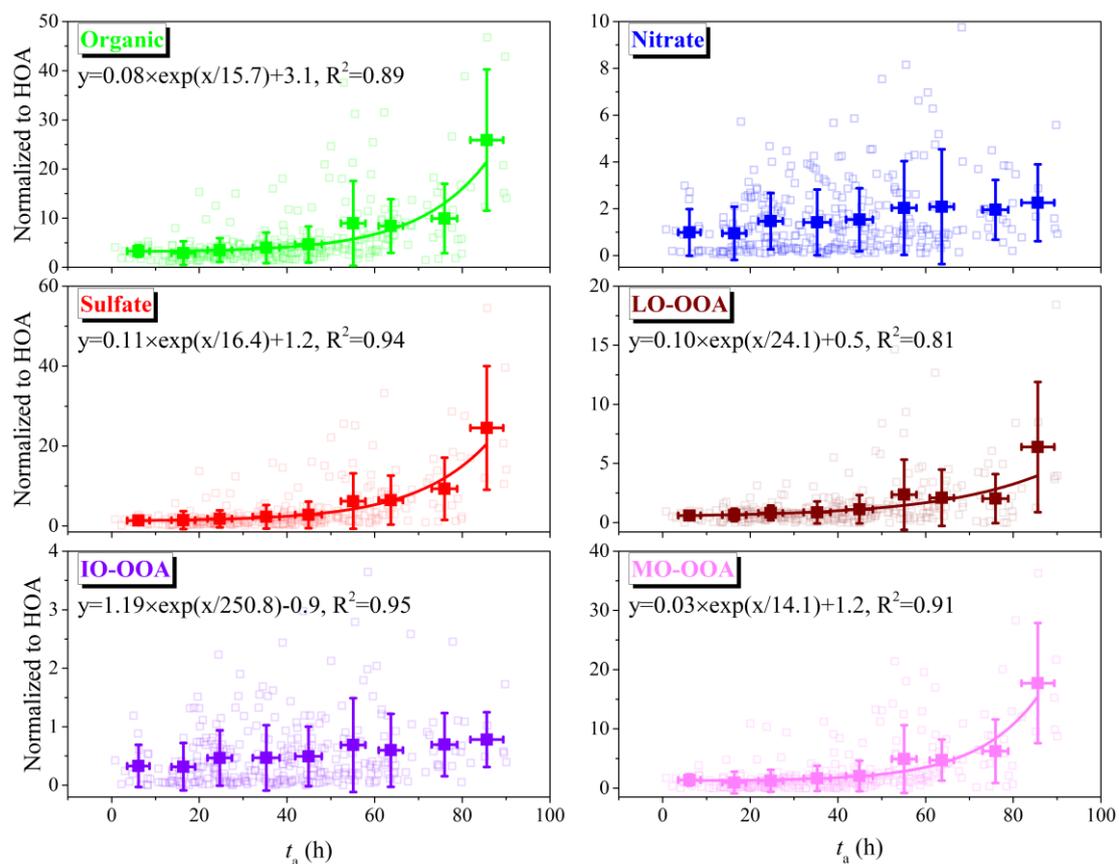


Figure 4. Variations of the ratios of mass concentrations of NR-PM₁ species and OA factors to HOA as a function of t_a . The data are binned according to the value of t_a (10 h increment). The solid lines are the results of exponential fitting with the function of $y = A \times \exp(x/B) + C$.

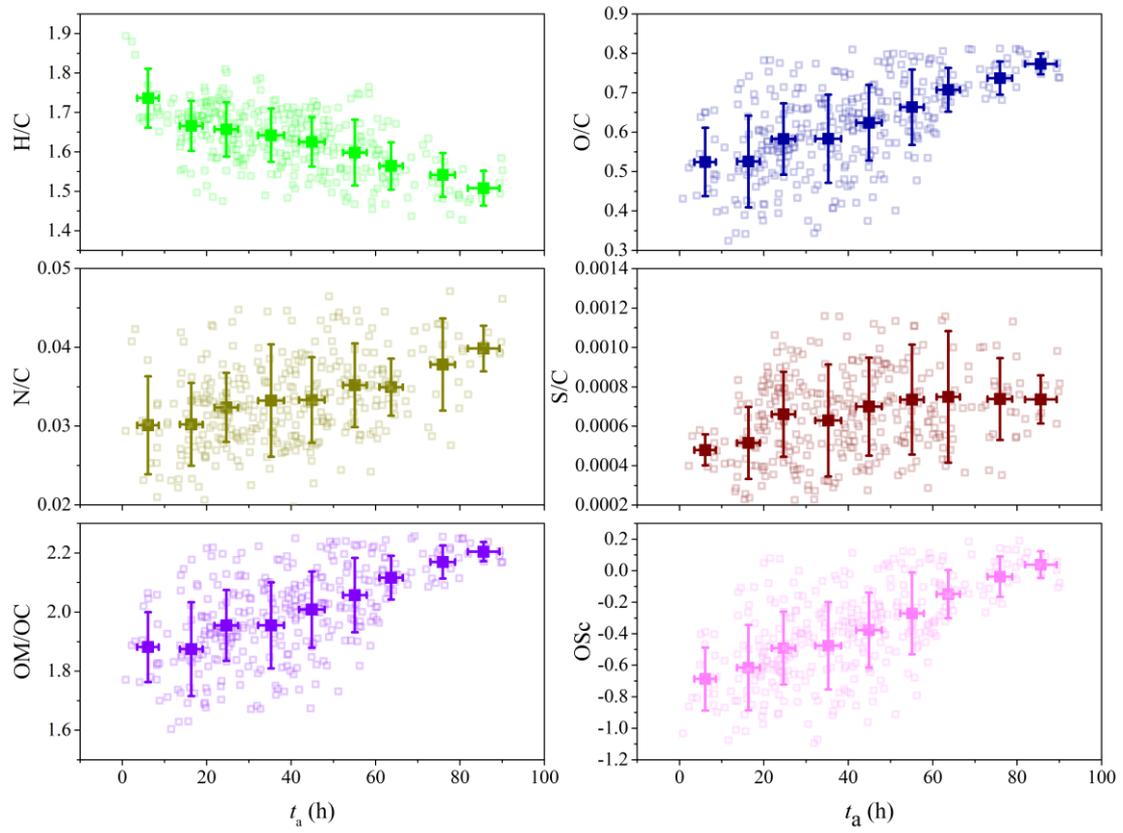


Figure 6. Variations of H/C, O/C, N/C, S/C, OM/OC and OSc as a function of t_a . The data are binned according to the value of t_a (10 h increment).

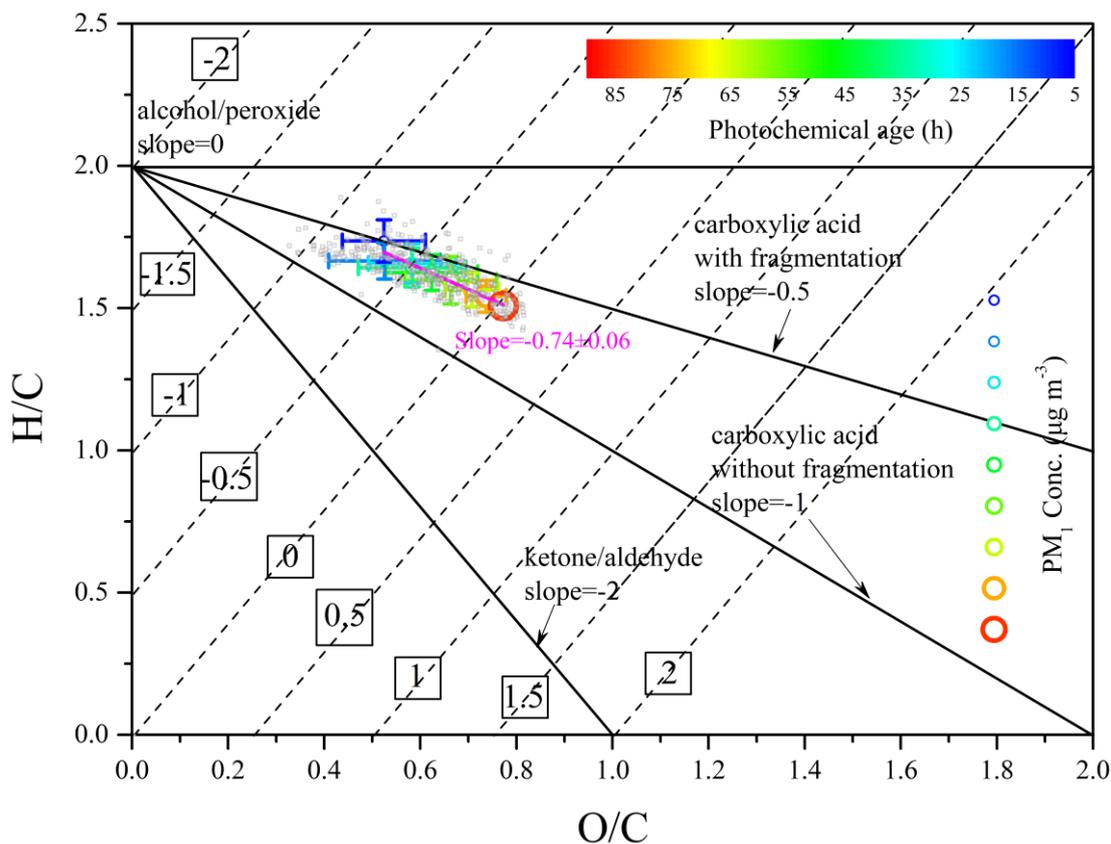


Figure 7. Van Krevelen diagram during this field observation. The data are binned according to the value of t_a (10 h increment) and color coded by t_a . The elemental composition of OA was fitted with a slope of -0.74 ($R^2 = 0.95$).

Comment 6: 3. Figure 2. When examining the photochemical evolution of a species, it is more intuitive to group species concentration measurements based on age, not the other way around as in Figure 2.

Response: Thanks for your valuable suggestion. According to your suggestion, a more intuitive method was used to group species concentration based on photochemical age. Meanwhile, raw data of each species concentration has been presented in the revised figure.

Revision in the manuscript:

Figure 2 had been modified in the revised manuscript as follows:

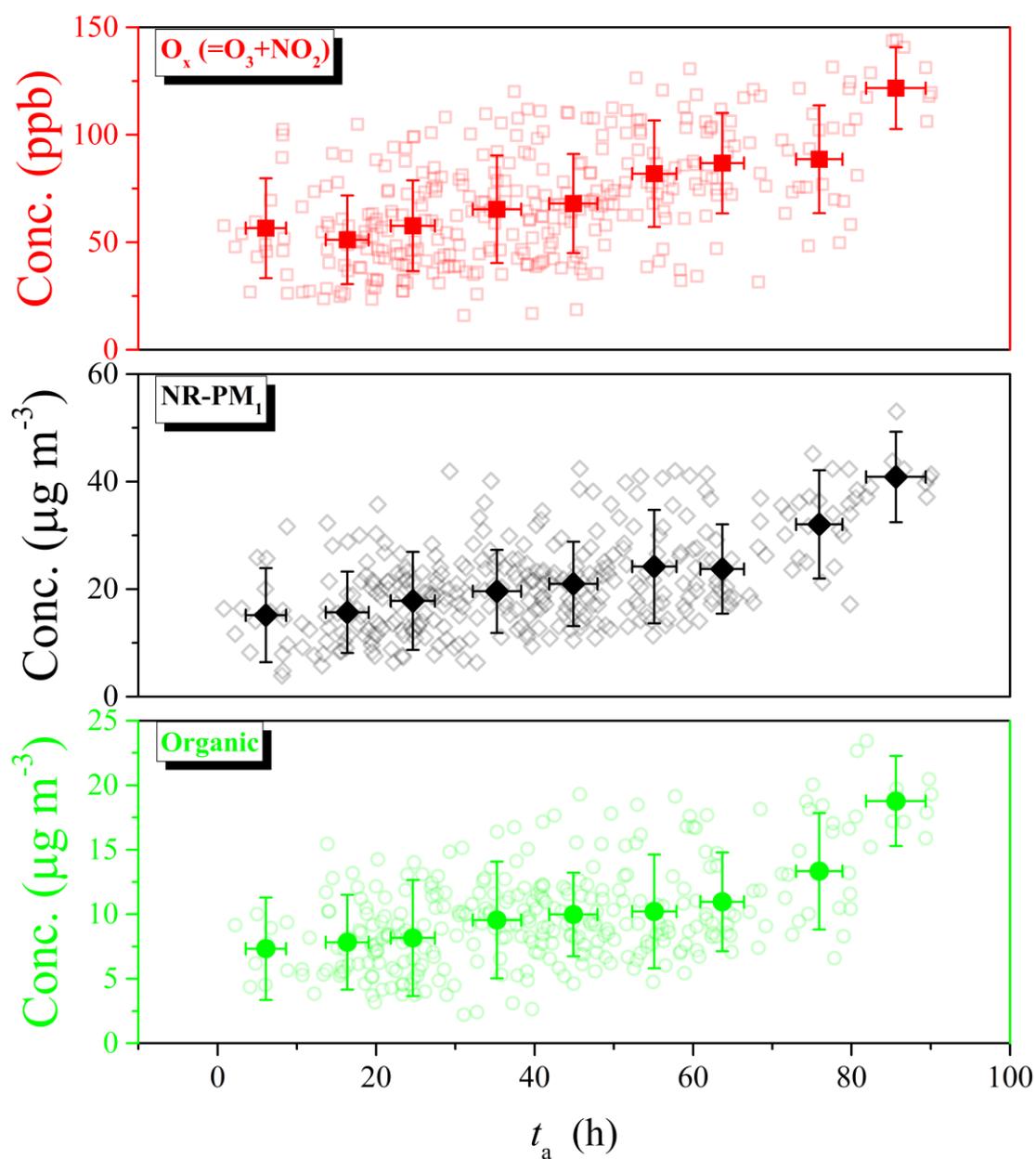


Figure 2. Relationship between photochemical age (t_a) and total oxidant (O_x), $NR-PM_1$ concentration, as well as organic concentration during the observation period included by Cluster 2. The data are binned according to the value of t_a (10 h increment).

Comment 7: 4. Many confounding factors are omitted in the analysis. For example, Figure S6 shows that T has a positive relationship with age and RH has a negative

relationship with age. Do the authors believe that these relationships are also due to photochemical aging? My point is that one cannot draw conclusion on the evolution of PM composition solely based on its relationship with photochemical clock.

Response: We agree with the reviewer that the evolution of PM₁ composition cannot solely based on its relationship with photochemical clock. In this study, the effect of photochemical age on the evolution of PM₁ composition was mainly considered and discussed, while other related factors, such as RH and T were also considered. Fig. S6 presented the variation trends of RH and T as a function of t_a , instead of the causal relationship between them. Based on these relationships, the effect of aqueous-phase process and evaporation process on the evolution of specific PM₁ composition can be illustrated. For example, we found that the evaporation of nitrate into the gas phase under high temperature condition might be the reason for the lower nitrate fraction at high levels of t_a . Therefore, the photochemical clock was not the only factor that was considered in this study to affecting the evolution of PM₁.

References:

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