

Thank you very much for the reviewer's suggestions for revision. According to the reviewer's suggestions, we have revised the paper. The reviewer's comments are in blue, the answers and revised text are in black.

Anonymous Referee #1

1. General comments

The authors sampled three different types of atmospheric aerosols onto quartz filters: (1) ambient PM_{2.5} in Xi'an, China, (2) primary organic aerosols from biomass burning, coal combustion and vehicle exhaust, as well as (3) laboratory-generated secondary organic aerosols from smog chamber experiments using α -pinene, limonene, naphthalene and toluene in a mixture of different yet unspecified concentrations. The list of all these samples are presented in Tables S1, S2, and S3. However, the data presented in the paper seems to have regrouped these samples in different and again unspecified ways with unsupported conclusions. All raw data of the experiments are unfortunately missing. Nevertheless, the TMP probe is an appropriate method for quantifying 3CDOM, yet further controls need to be presented. For example, TMP kinetics with the blank filter and in water (two negative controls), TMP kinetics for triplicate of one sample and TMP kinetics with a known 3CDOM* as a positive control. As it stands, experiments couldn't be reproduced due to lack of details and the authors should address this issue. Furthermore, the reported TOC measurements are appropriate, yet an emphasis on the importance of concentration for the measured kinetics should be added. A serious revamp of the manuscript writing is also recommended to present a logical sequence of events/results. For example, a discussion of the method and why the method is appropriate using logical sequencing (for example reorganizing the logic presented in lines 61-76 where the last sentence of the paragraph remains unsubstantiated and the reader is left wondering that "TMP has a higher reactivity" than what & why "TMP is not easily affected by these substances" and). I encourage the authors to aim to be precise and concise throughout their text. There is a missing discussion on how this study builds upon past work. For example, are the authors' results consistent (or not) with what other have observed so far? I would encourage the authors to state clearly what their hypothesis was and why they specifically chose the aerosol samples listed to support their hypothesis. Furthermore, was the starting hypothesis*

validated? The authors should clearly state their scientific approach. Therefore, as it stands the paper has too little technical details, making it difficult to understand and interpret the results and thus difficult to recommend for publication in its current state.

Thanks for the reviewer's suggestion, this will be helpful for improving this article. We have improved the paper according to your suggestions. It mainly includes the following aspects.

1. We supplemented data background of TOC in SI.

Table S5. The background concentration of TOC analysis.

Background ID	Concentration/ppm
1	10.83
2	10.15
3	22.78
4	3.55
5	0.84
6	4.81
7	2.23

2. We supplemented data of triplet state background and parallel experiments in SI.

Table S6. Background of of triplet state formation.

Parallel experiments	$k_{\text{TMP}}/\text{min}^{-1}$
1	0.018
2	0.019
3	0.017

Table S7. Analysis results of parallel experiments of triplet state formation.

Parallel experiments	$k_{\text{TMP}}/\text{min}^{-1}$
1	0.041
2	0.042
3	0.043

3. We have corrected words in lines 61-76.

We have corrected “*Because the chemical probe method has the characteristics of avoiding interference with the reaction system and accurate quantification (Lin et al., 2015)Some substances in the environment can inhibit the reaction between the probes and 3CDOM*, but TMP is not easily affected by these substances (Canonica & Laubscher, 2008; Wenk et al., 2015)*” to “*Chemical probe method has become a common method to study the characteristics of ³CDOM* formation (Lin et al., 2015) and the main chemical probes are dimethoxyphenol,which is unaffected by CDOM inhibition effects (Canonica & Freiburghaus, 2001; Canonica & Laubscher, 2008; Wenk et al., 2015)*”.

2. Specific comments

Title:

1. The title is misleading, as no “implications” of the work is mentioned.

We have corrected “*implications*” to “*effect*”.

Abstract:

1. Lines 4-5: the statement of $^3\text{CDOM}^$ contributing significantly to aerosol photochemistry is a conclusion of the work, and not an introductory statement. There has yet to be studies demonstrating the impact of $^3\text{CDOM}^*$ to aging in the context of aerosol-cloud interactions.*

According to the comment, we have changed “ $^3\text{CDOM}^*$ contributes” to “*Thus, $^3\text{CDOM}^*$ may contributes*”.

In this study, we have proved that CDOM in aerosols can form $^3\text{CDOM}^*$, which drives the generation of reactive oxygen species. And this work demonstrated hydroxyl and singlet oxygen are important driving factors of aerosol aging. Therefore, we state that the $^3\text{CDOM}^*$ can affect the aerosols aging by producing reactive oxygen species.

2. Line 5: “the ability” of what?

We have corrected “*We quantify the ability to form $^3\text{CDOM}^*$ and drive the formation of ROS by primary, secondary and ambient aerosols*” to “*We quantify the ability of CDOM of primary, secondary and ambient aerosols forming $^3\text{CDOM}^*$ and $^3\text{CDOM}^*$ driving the formation of ROS*”.

3. Between lines 4-8: please add information on the types of aerosols investigated as well as the method used.

We've already introduced the collection method and detailed information of aerosol samples used in the study in section 2.1.

4. Line 6: be specific when mentioning primary, secondary and ambient aerosols. To some extent all these qualifies could indicate identical samples.

The detail of POA, SOA and Ambient PM is shown in Table S1-S3 of SI.

5. Line 11: *The structure-activity relationship description should be made clear that it was developed in this work.*

We have added “*The structure-activity relationship reveals the contribution of CDOM to ³CDOM* formation*” in the improved paper.

6. Line 16: *be precise as to *how* the study verified that 3CDOM* drives ¹O₂. At the end: add a sentence relating to the implications of the work for aerosol photochemistry.*

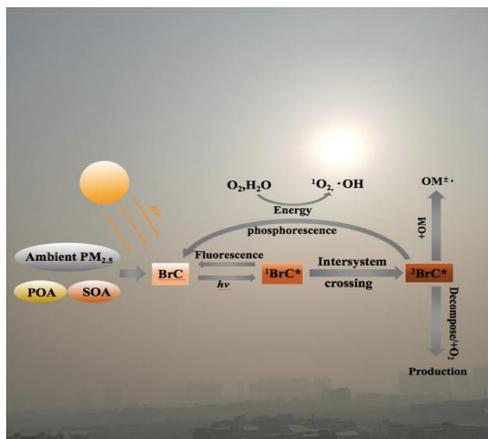
By comparing the amount of ¹O₂ and ·OH in the reaction system with or without triplet state, we prove the contribution of triplet state to reactive oxygen species.

We have added “*which reflects the significant contribution of triplet state to aerosol aging*” in the improved paper.

TOC art:

1. *What is the meaning of the different colors/shades?*

We has modified the TOC. As shown in TOC, the ellipse represents three sources of aerosols and the square represents different states of brown carbon.



Introduction:

1. Lines 24-26: *unsubstantiated sentence and unsupported by the chosen references. Best to use reviews on brown carbon (or even better, modeling studies) to support a claim on climate impacts, since the current references deal with laboratory studies.*

We have corrected “*Aerosols contain organic substances that can absorb sunlight and promote photochemical reactions and have a potentially significant impact on the global climate and atmospheric environmental quality*” to “*Aerosols contain organic substances that can absorb sunlight and promote photochemical reactions and have a potentially significant impact on atmospheric photochemical reaction process and atmospheric quality*”.

2. Line 32: the Kaur et al. 2019 reference is only valid for the IO2 claim in this sentence. Best to include accurate references for the other oxidants.

We have added references in the improved paper.

- Fujii, M.; Rose, A. L.; Waite, T. D. and Omura, T.: Oxygen and superoxide-mediated redox kinetics of iron complexed by humic substances in coastal seawater. *Environ. Sci. Technol.*, 44, 9337–9342, <https://doi.org/10.1021/es102583c>, 2010.
- Li, R., Zhao, C., Yao, B., Li, D., Yan, S. W., O’Shea, K. E. and Song, W. H.: Photochemical Transformation of Aminoglycoside Antibiotics in Simulated Natural Waters, *Environ. Sci. Technol.*, 50, 2921–2930, <https://doi.org/10.1021/acs.est.5b05234>, 2016.
- Sun, L. N.; Qian, J. G.; Blough, N. V. and Mopper, K.: Insights into the photoproduction sites of hydroxyl radicals by dissolved organic matter in natural waters, *Environ. Sci. Technol. Lett.*, 2, 352–356, <https://doi.org/10.1021/acs.estlett.5b00294>, 2015.
- Zhang, D. N.; Yan, S. W. and Song, W. H.: Photochemically induced formation of reactive oxygen species (ROS) from effluent organic matter, *Environ. Sci. Technol.*, 48, 12645–12653, <https://doi.org/10.1021/es5028663>, 2014.

3. Lines 35-54: I encourage the authors to be more specific when referencing earlier studies. The authors should add and specify the mechanisms at play, the specific atmospheric environment (line 37), the explanation of how Corral Arroyo et al 2018 proved that the triplet state affected aerosol aging (lines 41-42), the explanation of how 3CDOM contributes to SOA formation (line 45), the specific “certain chemical reactivity” (line 46), the specific “important role” (line 49).*

According to the comment, we have corrected those points in the improved paper.

We have corrected “*such as sewers, terrestrial natural waters and oceans (Bodhipaksha et al., 2015; Erickson et al., 2018; Zhou et al., 2019)*” to “*CDOM in sewers (Bodhipaksha*

et al., 2015) and river (Erickson et al., 2018; Zhou et al., 2019) have the ability of formation $^3\text{CDOM}^$.*

We have corrected “*Corral Arroyo et al. (2018) proved that the triplet state has an effect on photochemical reaction and aerosol aging on the particle phase*” to “*Corral Arroyo et al. (2018) proved that Atmospheric particles contain BrC, which is the triplet state forming organic compounds that can sustain catalytic radical reactions and thus contribute to oxidative aerosol aging*”.

We have corrected “ *$^3\text{CDOM}^*$ has a certain chemical reactivity, which leads to its participation in various photochemical reactions*” to “ *$^3\text{CDOM}^*$ participation in various photochemical reactions have been revealed*”.

4. Line 55: low concentrations compared to what?

We have deleted “*low concentrations*”.

5. Lines 58-59: consider rewriting this sentence. A method “becoming an early analysis method” appears to be an oxymoron, especially when a 1969 reference is used.

We have corrected “*Studying the $^3\text{CDOM}^*$ quenching process by phosphorescence has become an early analysis method*” to “*The quenching process of $^3\text{CDOM}^*$ could be studied by phosphorescence*”.

6. Line 61: specific the chemical probe method

In this study, the chemical probe method is as follows: specific chemical substance reacts with triplet state, and the formation characteristics of triplet state are indirectly studied by quantitative analysis of the specific chemical substance.

7. Line 75: Why isn't TMP not easily affected by “these substances”. Add more specific information. Dissolved organic matter (DOM) is both a promoter and an inhibitor of triplet-induced organic contaminant oxidation.

We have corrected “*Some substances in the environment can inhibit the reaction between the probes and $^3\text{CDOM}^*$, but TMP is not easily affected by these substances*” to “*In addition, CDOM electronic absorption in the visible range is largely due to donor-*

acceptor complexes between electron-rich aromatic donors and carbonyl-containing acceptors. The inhibitory effect decreased with the increasing extent of CDOM pre-oxidation, and it was correlated to the loss of phenolic antioxidant moieties, as quantified electrochemically, and to the loss of DOM ultraviolet absorbance. The triplet photosensitizing ability of pre-oxidized DOM was determined using the conversion of the probe compound TMP, which is unaffected by CDOM inhibition effects”.

- Wenk, J., Aeschbacher, M., Sander, M., Gunten, U. V., and Canonica S.: Photosensitizing and Inhibitory Effects of Ozonated Dissolved Organic Matter on Triplet-Induced Contaminant Transformation, *Environ. Sci. Tech.*, 49, 8541-8549, <https://doi.org/10.1021/acs.est.5b02221>, 2015.

8. Lines 84-85: specifically mention what (Korak et al., 2014; Ma et al., 2010; McKnight et al., 2001; Rosario-Ortiz & Canonica, 2016; Wenk et al., 2015) observed.

We have corrected “*such as humic-like substances (HULIS), quinones, phenols and amino acids*” to “*For example, humic-like substances (HULIS), quinones, phenols and amino acids could be identified by fluorescence spectral characteristics*”.

9. Lines 88-95: what is the study’s scientific hypothesis? Why were Xi’an samples studied? How do these samples help support the hypothesis?

Scientific hypothesis: We think that there are a lot of chromophore substances in aerosols, which not only affect the solar radiation, but also affect the atmospheric photochemical reaction process. Therefore, we insist that chromophores in aerosols have the ability to form trilinear States, and further affect the formation of reactive oxygen species and the aging of aerosols.

As the largest central city in Northwest China, Xi'an city has serious air pollution and high annual average concentration of carbonaceous aerosols. The higher carbonaceous components contain more chromophores, so we chose the aerosol in Xi'an as the research object.

- Mu, Z., Chen, Q. C., Wang, Y. Q., Shen, Z. X., Hua, X. Y., Zhang, Z. M. et al.: Characteristics of carbonaceous aerosol pollution in PM_{2.5} in Xi'an, 40, 1529-1536, *Environ. Sci.*, <http://doi.org/10.13227/j.hjkx.201807135>, 2019.

We think that the types, contents and the ability to form triplet states of chromospheres are different in different sources of aerosols. Therefore, different sources of aerosols have been studied to comprehensively explain the characteristics of the formation of triplet states in different sources of aerosols.

Material and methods:

1. Lines 103-104: a 1000 L/min flow rate for a 24 h sample appears to be very high. The PM2.5 samplers I've worked with don't typically exceed 50 L/min. The authors should comment on this high flow rate and specify the instrument used for collection.

In section 2.1, we illustrate the details of particulate matter collection by an intelligent large-flow particle sampler (Xintuo XT-1025, Shanghai, China). A large sampling flow is accompanied by a large area of quartz fiber filter, which is 348.8 cm².

2. Line 114: why is the flow rate here only 16.7 L/min? why is different than previously mentioned?

Compared to ambient PM, the concentration of POA is very high, and the filter area we use is 43.01 cm². Therefore, according to aerosol concentration, sampling conditions, filter area, small sampling flow rate of POA was selected.

3. How were the quartz filters pre-conditioned before sampling?

Before sampling, quartz filter is baked in muffle furnace at 450 °C for 4.5. Quartz filter is put it into a clean tin foil bag after baking.

4. Lines 120-133: verbs should be in the past tense. A further lack of detail, which is rather frustrating for the reader.

We have corrected it in the improved paper.

5. Line 122: specific which alcohol.

We have corrected “alcohol” to “ethanol”.

6. *Line 136: no need for acronyms here.*

We have deleted acronyms of limonene, α -pinene, toluene and naphthalene.

7. *Line 141: what was the concentration of cyclohexane added?*

We have corrected “cyclohexane” to “cyclohexene”. The concentration of cyclohexene is 2.9×10^{13} molecule \cdot cm⁻³.

- Liu, S. J., Jiang, X. T., Tsona, N. T., Lv, C., and Du, L.: Effects of NO_x, SO₂ and RH on the SOA Formation from Cyclohexene Photooxidation, *Chemosphere*, 216, 794-804, <https://doi.org/10.1016/j.chemosphere.2018.10.180>, 2018.

8. *Line 142: how was ozone produced? Which high voltage? Which instrument?*

We have corrected “Under pure oxygen flow conditions, O₃ is produced by a high voltage current, and VOCs are oxidized by O₃” to “and VOCs were oxidized by O₃”.

- Liu, S. J., Jiang, X. T., Tsona, N. T., Lv, C., and Du, L.: Effects of NO_x, SO₂ and RH on the SOA Formation from Cyclohexene Photooxidation, *Chemosphere*, 216, 794-804, <https://doi.org/10.1016/j.chemosphere.2018.10.180>, 2018.

9. *Line 150: discuss why the filters were ultrasonically extracted in light of the paper, “To Sonicate or Not to Sonicate PM Filters: Reactive Oxygen Species Generation Upon Ultrasonic Irradiation” (Miljevic et al., 2014)*

Our previous research has proved that the efficiency of ultrasonic extraction for water-soluble brown carbon is high (the extraction efficiency of fluorescent substances is 77%, and the extraction efficiency of light-absorbing substances is 89.3%).

- Chen, Q. C., Mu, Z., Song, W. H., Wang, Y. Q., Yang, Z. H., Zhang, L. X. & Zhang, Y. L.: Size-resolved characterization of the chromophores in atmospheric particulate matter from a typical coal-burning city in China, 124, <https://doi.org/10.1029/2019JD031149>, 2019.

Ultrasound may cause the production of reactive oxygen species. However, for example, the lifetime of hydroxyl radical is only 5-10 μ s. During the extraction process, we did not add any reactive oxygen species capture agent. Therefore, we do not think that the reactive

oxygen species generated by ultrasonic extraction will affect the study of the triplet driving active oxygen.

10. Lines 152-153: the filter was made of what material?

We have corrected “a 0.45- μm filter” to “a 0.45- μm quartz filter”.

11. Lines 153-154: add this background extraction data to the tables in the SI.

We have added it in SI.

Table S5. The background concentration of TOC analysis.

Background ID	Concentration/mg/L
1	10.83
2	10.15
3	22.78
4	3.55
5	0.84
6	4.81
7	2.23

12. Line 159: The Sievers M9 TOC analyzer, as far as I know is from Suez Technologies, not from General Electric.

When we bought the instrument, it was still owned by General Electric Company. We has referenced the mark marked on the instrument.

13. Lines 159-161: why was sample exposure to air and time a problem. Please show this data.

According to our previous analysis methods and research experience, the detection value of TOC of WSOC extraction sample will increase if it is placed or exposed to air for a long time.

14. Line 162: specific which background samples.

Each time a batch of TOC analysis is performed, the background sample is analyzed in the same way, Table S5 of SI.

15. Line 165: describe in detail the offline analysis method.

The off-line analysis mode is the internal program of the TOC analyzer. Each sample analysis requires a fresh start. Detailed is shown in section 2.3.

16. Line 166: why was the background subtracted? Please show the data.

Background is subtracted for more accurate concentration information of WSOC.

Background detail is shown in Table S5 of SI.

17. Line 174: define the background samples

The background samples are the sample without air sampling process, and the rest process is the same as the samples.

18. Lines 176-177: define the “inner filter effect” and show the data

Inner filter effect: When the concentration of extraction is high, the fluorescence will be weakened because of the absorption of excitation or emission light by Light absorbing substance. During the experiment, we only reduce the concentration of the extraction liquid to avoid the internal filtration effect. It is a fundamental principle in EEM analysis. When the absorbance is 0.5 at most, the internal rate effect coefficient is about $3.16 \approx 10^{(Abs_Ex+Abs_Em)/2} = 10^{(0.5+0.5)/2}$ at most. The instrument (Aqualog, Horiba Science) matching data collection software also has the automatic correction function.

- Murphy, K. R.; Stedmon, C. A.; Graeber, D.; Bro, R., Fluorescence spectroscopy and multi-way techniques. PARAFAC. Anal. Methods 2013, 5 (23), 6557-6566.

19. Line 180: show dimensions of the customized reactor in Figure S1.

We have corrected it in Figure S1. “*The inside diameter of the reaction device is about 11 cm and the height of the reaction device is about 18 cm.*”

20. Line 186: why are the 25 °C and 50% RH conditions chosen? Are they relevant to Xi'an?

Our custom quartz plate is open, 50% humidity and temperature of 25 °C can not only ensure that the sample solution does not volatilize largely during the illumination process, but also provide a suitable reaction environment and reaction temperature.

21. Line 195: show the calculation (in the SI) to arrive at a factor 1.2-1.3.

We calculated the intensity of the illumination by *Tropospheric Visible Ultra-Violet (TUV) model web page*. Input parameters for the TUV model were: Longitude: E108°58'34.58", Latitude: N34°22'35.07", measurement altitude: 0.02 km, surface albedo: 0.1, aerosol optical depth: 0.235, cloud optical depth: 0.00.

22. Line 203: specific the "previous study" as there are no references.

We have added references.

- Bodhipaksha, L. C., Sharpless, C. M., Chin, Y. P., Sander, M., Langston, W. K., and MacKay, A. A.: Triplet Photochemistry of Effluent and Natural Organic Matter in Whole Water and Isolates from Effluent-Receiving Rivers, *Environ. Sci. Technol.*, 49, 3453-3463, <http://doi.org/10.1021/es505081w>, 2015.
- Zhou, H. X., Yan, S. W., Lian, L. S., and Song, W. H.: Triplet-State Photochemistry of Dissolved Organic Matter: Triplet- State Energy Distribution and Surface Electric Charge Conditions, *Environ. Sci. Tech.*, 53, 2482-2490, <https://doi.org/10.1021/acs.est.8b06574>, 2019.

23. Lines 208-209: support this claim with references

We have added a reference.

- Dogliotti, L & Hayon, E.:Flash photolysis of per[oxydi]sulfate ions in aqueous solutions. The sulfate and ozonide radical anions, *J. Phys. Chem.*, 71, 2511-2516, <https://doi-org/10.1021/j100867a019>, 1967.

24. Lines 209-219: this paragraph is vague and lacks details. Please specific which salts were investigated and why would the authors expect a salt effect on 3CDOM? Which literature are they building upon?*

In this study, NH_4^+ , SO_4^{2-} , NO_3^- and Ca^{2+} were the main components of water-soluble ions in $\text{PM}_{2.5}$ in Xi'an (Li et al. manuscript, in review), And sulfate ion can generate sulfate free radical. Sulfate was selected in order to demonstrate that free radicals formed by ions do not consume TMP.

- Dogliotti, L & Hayon, E.:Flash photolysis of per[oxydi]sulfate ions in aqueous solutions. The sulfate and ozonide radical anions, *J. Phys. Chem.*, 71, 2511-2516, <https://doi-org/10.1021/j100867a019>, 1967.

25. *Line 159 & 221: America is a continent not a country, and should be corrected.*

We have corrected “America” to “the US”.

26. *Lines 220-221: add information on the column used in the UPLC.*

We have added “Column type is C18 and length is 12 cm” in the improved paper.

27. *Lines 226-227: show the data for this statement in the SI.*

We have added the data in SI.

Table S7. Analysis results of parallel experiments of triplet state formation.

Parallel experiments	$k_{\text{TMP}}/\text{min}^{-1}$
1	0.041
2	0.042
3	0.043

28. *Lines 233-234: IO2 was quantified using EPR, how do these values compare with the FFA method (Appiani et al., 2017)? How was the signal quantified? Which positive control was used?*

It is a simple method to quantify the production of active oxygen by chemical probe FFA. The main quantitative method is the calculation of quantum yield. However, the EPR method is a direct method, and the main measurement method is the number of spin electrons. Our main purpose is to prove that the triplet state has a significant driving effect on reactive oxygen species. We have not compared the two methods and this is also the direction of our future research.

29. *Line 244: which probe was used?*

We have corrected “the probe” to “TMP”.

30. *Line 254: specify which types of CDOM and give examples.*

We have explained in detail in section 3.3. Such as tryptophan may have more significant ability of driving triplet state.

Results and discussion:

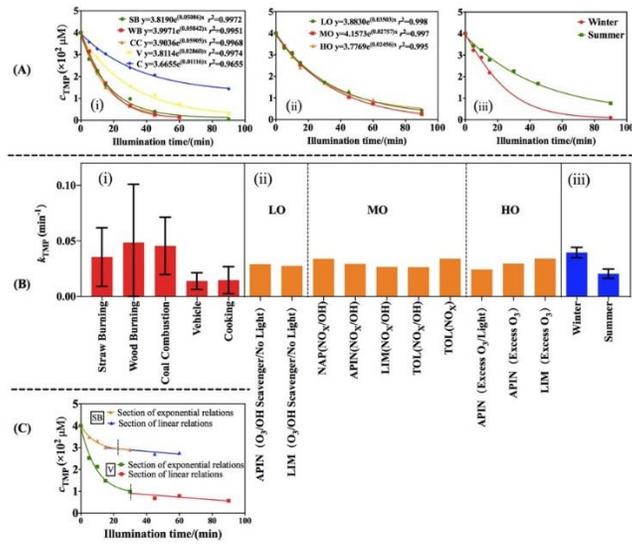
I did not find any evidence to support scheme 1 in the paper. The authors should clarify how their own experiments rule out or support a particular pathway. I am rather sceptical that the measurements done in this study can differentiate between a chemical reaction and an energy transfer. How do the authors know whether the product is directly from 3CDOM or from IO₂ + DOM? Also why did the authors chose to use the acronym BrC in this scheme when throughout the text, they use 3CDOM*? The scheme should also be made much larger and should have at least font size 12. All figures should be separated into individual panels. For example, Figure 1 should be 3 separate figures. Why do only a few of the panels in figure 1B have error bars? Where is the error in Figure 1A? Why are there so many significant figures reported in Figure 1A; I doubt they are all meaningful. The acronyms in the figures should all be described in the caption.*

The reaction mechanism of scheme 1 has been proven in previous study, which is mainly used to speculate on the reaction mechanism.

- Rosario-Ortiz, F. L., and Canonica, S.: Probe Compounds to Assess the Photochemical Activity of Dissolved Organic Matter, Environ. Sci. Tech., 50, 12532-12547, <https://doi.org/10.1021/acs.est.6b02776>, 2016.

We have corrected “BrC” to “CDOM”.

According to reviewer suggestions, we also corrected the Figure 1.



1. Line 288: what is means by “more $^3\text{CDOM}^*$ is formed in the initial stage”

We consider that there exists following-mentioned reaction process. In the early stage of illumination, the triplet state formed by CDOM is the main reaction. The degree of triplet reaction with TMP is relatively weak, so it has a certain amount of triplet accumulation. Therefore, we believe that “more $^3\text{CDOM}^*$ is formed in the initial stage”.



2. The average values reported in lines 307-308 represent which samples? What does the standard deviation represent? The authors should use IUPAC units and should report their values in seconds, rather than in minutes.

We have corrected “the values of k_{TMP} were 0.032 ± 0.032 , 0.030 ± 0.005 and $0.030 \pm 0.011 \text{ min}^{-1}$, respectively” to “which were 0.032 ± 0.032 , 0.030 ± 0.005 and $0.030 \pm 0.011 \text{ min}^{-1}$ ”.

The lighting stage and the curve fitting of TMP both are in minutes.

3. Line 308 contradicts line 307. The authors state no difference and then the authors state a significant difference. These statements need to be clarified that the seasonality of the ambient aerosols is now being discussed.

The average value of POA, SOA and Ambient PM from different sources is similar. For example, k_{TMP} of POA, SOA and Ambient PM were 0.032 ± 0.032 , 0.030 ± 0.005 and

0.030±0.011 min⁻¹, respectively. Coal burning, straw burning, and motor vehicles belong to POA, their values are significantly different.

4. Line 312: what is the chemical difference between straw and wood burning?

The straw we mentioned here is mainly agricultural crops. At the same time, there may be more phenols in the combustion products of straw, but wood does not have these characteristics.

5. Line 318: the authors claim that N-alkyl, carboxylic acids and alkanols do not produce ³CDOM. Where is this evidence and/or this data?*

Because of the complex nature of the aerosol composition, current studies cannot accurately determine the precursors of triplet formation, but aromatic ketones, aldehydes and quinones may be typical of triplet state precursor. We have corrected to “*which do not produce ³CDOM**” to “*which may be not typical ³CDOM* precursor*”.

- Rosario-Ortiz, F. L., and Canonica, S.: Probe Compounds to Assess the Photochemical Activity of Dissolved Organic Matter, *Environ. Sci. Tech.*, 50, 12532-12547, <https://doi.org/10.1021/acs.est.6b02776>, 2016.
- Ma, J. H., Del Vecchio, R., Golanoski, K. S., Boyle, E. S., and Blough, N. V.: Optical Properties of Humic Substances and CDOM: Effects of Borohydride Reduction, *Environ. Sci. Tech.*, 44, 5395–5402, <https://doi.org/10.1021/es100880q>, 2010.
- Wenk, J., Aeschbacher, M., Sander, M., Gunten, U. V., and Canonica S.: Photosensitizing and Inhibitory Effects of Ozonated Dissolved Organic Matter on Triplet-Induced Contaminant Transformation, *Environ. Sci. Tech.*, 49, 8541-8549, <https://doi.org/10.1021/acs.est.5b02221>, 2015.

6. Line 323: explain why aliphatic compounds cannot form ³CDOM and why these specific compounds are attributed to the authors' result for vehicle exhaust.*

Similar to the previous question, there is currently no precise judgment on the precursors in the triplet state. We have made a reasonable guess based on the study results. We have corrected “*These aliphatic compounds cannot form ³CDOM*, which lead to the low k_{TMP}* ”

values of the vehicle exhaust and cooking samples” to “These substances do not contribute significantly to the triplet state”.

7. Line 345: which “types” of 3CDOM* are the authors referring to? Be specific.

What we mentioned in the paper are the types of CDOM. From the experimental results, we insist that the ability of different CDOM to form triplets is different. We have explained in detail in section 3.3.

8. Figure 2: avoid all the acronyms in the middle of the figure. What do the error bars signify and why are the + values shown but not the -values?

We have corrected Figure 2.

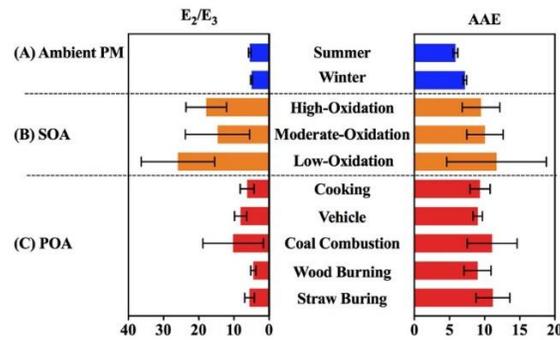


Figure 2. The characteristics of the AAE and E_2/E_3 ratio of different types of aerosols.

9. Lines 366-367: show the data.

The data are shown in Figure 3.

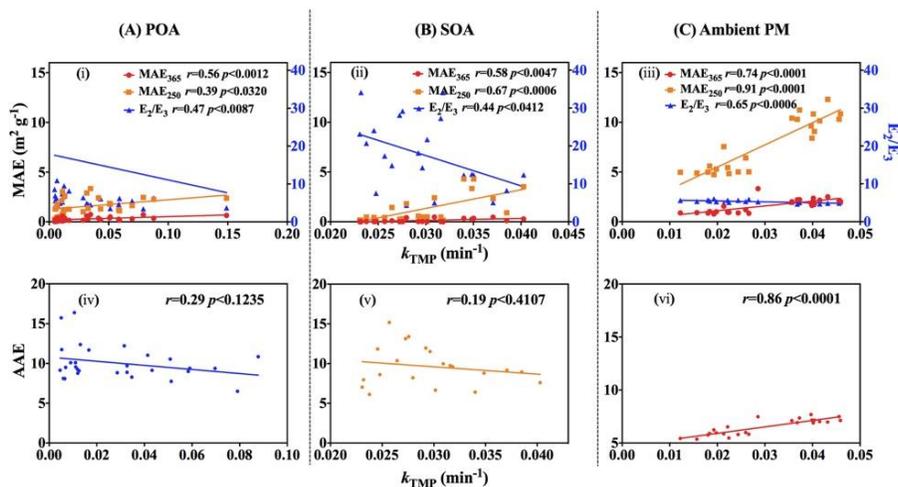


Figure 3. The characteristics of the correlation between k_{TMP} , the AAE, and the E_2/E_3 ratio. (i), (ii) and (iii) show the correlations between k_{TMP} , MAE and E_2/E_3 . (iv), (v) and (vi) show the correlations between k_{TMP} and the AAE. (A), (B) and (C) are POA, SOA and ambient PM sample results, respectively.

10. Figure 3: for optimal comparison, best to have all the same values for all the axes.

We have corrected Figure 3. As shown in the figure above.

11. Figure 4 should be split into 4 or 5 individual figures. Why were SOA values not included in the fit and which mechanism explains their deviation from the fit?

Figure 4 have been split into Figure 4 and 5 in the paper.

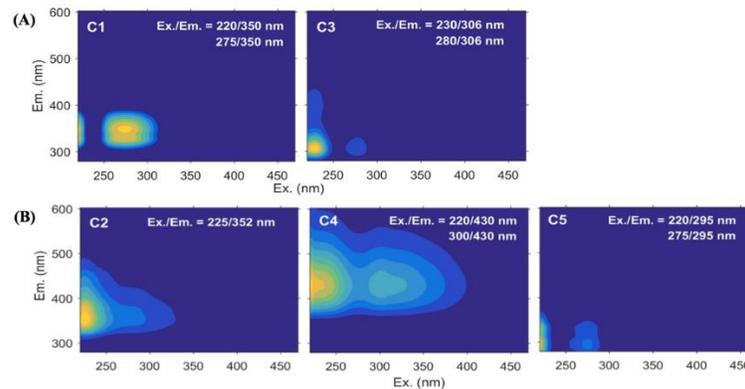


Figure 4. 5 types of CDOM. (A) CDOM contributing to triplet formation. (B) CDOM that do not contribute to the triplet state.

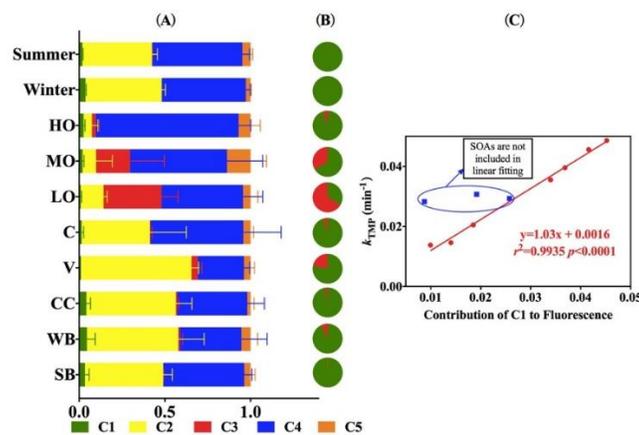


Figure 5. Types of CDOM and their contributions to $^3CDOM^*$. (A) Different CDOM types contributing to fluorescence. (B) Difference in CDOM contributions to $^3CDOM^*$. (C) Linear relationship between the contribution of C1 to fluorescence and k_{TMP} .

We found that the contribution of C1 to fluorescence did not significantly affect the k_{TMP} . As shown in the Figure 5, SOA do not include in the linear fitting.

Environmental implication:

1. Figures 5 & 6: why do the signals' noise look different in each figure?

When the reactive oxygen species were studied by EPRs, the same curves of the two groups could not be detected. According to the characteristic peaks, the generation characteristics of different reactive oxygen species are judged.

2. Where and how did the authors identify C1 and C3 chromophores in the study? And how did they measure N-containing substances. None of these experiments appear in this study.

C1 and C3 are tryptophan and the CDOM driven by the Maillard reaction, which does not mean that C1 and C3 are the two substances. It only means that the fluorescence characteristics of C1 and C3 are the same or similar to these two substances.

According to the EEMs characteristics of the detected chromophores compared with the previous studies, we found that the maximum excitation wavelength and maximum emission wavelength of C1 and C3 are similar to these of tryptophan and the CDOM driven by the Maillard reaction, so we state that C1 and C3 chromophores are tryptophan and the CDOM driven by the Maillard reaction, respectively. Both of these CDOM are nitrogen-containing substances.

3. Line 472-473: this sentence is confusing. The hypothesis should be reiterated here and the results stated with the implications of the work. The authors should be comparing their work with previous work on $^3\text{CDOM}^$ in the atmosphere in this section.*

We have corrected “However, the above do not mean that these substances do not have the ability to form $^3\text{CDOM}^*$. In this case, as shown in Scheme 1, $^3\text{CDOM}^*$ through self-quenching and energy transfer does not consume TMP, and low-energy $^3\text{CDOM}^*$ cannot react with TMP” to “because self-quenching, energy transfer (as shown in Scheme 1) and low-energy do not consume TMP”.

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