Response to the Comments of the Reviewers

Dear Editor and Reviewers,

We acknowledge the comments and encouragement of two reviewers, and are also grateful to the efficient serving of the editor. Here we submit our revised manuscript **"Measurement report: VOC characteristics at different land-use types in Shanghai: spatio-temporal variation, source apportionment, and impact on secondary formations of ozone and aerosol" (Manuscript number: acp-2022-250)**, as well as a thorough, point-by-point response to each point raised from the reviewers. The revisions to the manuscript are highlighted in blue words in the provided "Response to the Comments of the Reviewers". Additionally, there is a clean revised manuscript as required.

We greatly appreciate those comments and valuable suggestions from the reviewers. The manuscript has been greatly improved. We do feel that we have demonstrated our efforts in the revised manuscript.

Yours sincerely,

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Response to the Reviewer #1

General Comments:

The manuscript presents a study on volatile organic compounds (VOCs) at three sites in a Chinese megacity, ~ Shanghai. The concentration, composition, sources and of VOCs have been extensively studied, especially in the major cities like Shanghai. The authors claim multiple-site comparison as the main selling point. However, it is not enough to provide new insights that the authors expect. Namely, I have reservations about the novelty of this study. Moreover, I have serious concerns on the rationality of the methods and quality of the results presentation. The lots of grammatical errors also make the manuscript very difficult to be reviewed. Overall, the manuscript is well below the average of papers published in Atmospheric Chemistry and Physics, so I do not think it is worth publishing in its current state. However, I do not mind providing some specific comments for the authors' reference and reviewing a resubmitted edition after it is substantially improved.

Response:

We would like to thank reviewer #1 for carefully reading our manuscript and for your valuable and constructive comments. All your suggestions are very important and they are of great significant to our scientific research. We carefully revised and improved each part according to the reviewer's suggestions.

The long-term VOC emission inventory highlighted that the VOC emission varied with the land-use types (Li et al., 2019). The observation campaign also showed that VOC concentrations were largely influenced by the land-use type (Tang et al., 2008; Kumar et al., 2018; Zhang et al., 2018). Besides, the land-use types not only influence the VOC concentrations but also the sources especially the anthropogenic sources (Yoo et al., 2015; Chen et al., 2017; Wang et al., 2017; Jookjantra et al., 2022). Additionally, the diversities of VOC concentrations among the different land-use types could cause the distinct ozone and SOA formation potentials (OFP and SOAFP), resulting in the variations of O_3 and SOA concentrations (Song et al., 2021; Zhan et al., 2021; Liu et al., 2022). Shanghai is regarded as an ideal area to perform atmospheric measurements with the different land-use types. However, many studies

were mainly focused on the single-site measurements, particularly conducted at the urban site in Shanghai, resulting that the impact of land-use type on VOC characteristics is still unclear to date. Moreover, they mainly concentrated on the O_3 characteristics, while the O_3 and SOA formations from VOCs and the relationship between VOCs together with $PM_{2.5}$ and O_3 were rarely analyzed. Additionally, the distinct land-use types among the sampling sites were observed. The JS site is located in the Second Jinshan Industrial Area of Shanghai as the industrial district, and surrounded by many chemical factories. The PD site is located in the Pudong New Area as the residential and commercial mixed districts, and surrounded by residences and administrative areas. The QP site is located at the southeast of Dianshan Lake as the background district, and surrounded by many farmlands and forests. Given the factors mentioned above, in this study, the concurrent multiple-site and high time-resolution measurement of the VOCs with three typical land-use types in Shanghai for their characteristics, sources and ozone and SOA formation potentials was performed. The results at the multiple-site measurement benefit the government to establish efficient and specific environmental control measures according to the specific land-use types. Lastly, we would like to thank reviewer for the positive comments again.

Comment 1: The term "secondary formation potentials" is not a common expression. It is not clear what you are referring to.

Response:

Thank you for your comments. The "secondary formation potentials" is referred to the secondary organic aerosol formation potential (SOAFP) and ozone formation potential (OFP). We revised the title to

> *"Measurement report: VOC characteristics at different land-use types in Shanghai: spatio-temporal variation, source apportionment, and impact on secondary formations of ozone and aerosol"*

Comment 2: Line 29: VOCs-S_{O3} has not been defined before. So is SOA in line 31.

Response:

Thank you for your comments. We added the definition in the revised manuscript.

Line 30:

"Alkenes and aromatics are both the key concerns in controlling the VOC-related pollution of O_3 and secondary organic aerosol (SOA) in Shanghai."

The calculation of VOC-O_3 sensitivity (VOCs-S_{O3}) was deleted after consideration.

Comment 3: Line 31: "new insights". I do not think the paper at its current state provides new insights into the accurate air quality management.

Response:

Thank you for your suggestion. We changed the words in the revised manuscript.

"These findings provide more information on the accurate air-quality control at a city level in China."

Comment 4: The whole section is rather simple and most of the contents (if they are right) can be found in text book. I do not think it is necessary to elaborate them in a research article. What's worse, I am confused by the introduction of some basic knowledge. For example, line 40: RO_2 is formed following oxidation of VOCs, but VOCs are oxidized by OH, O_3 and NO_3 (NOT RO_2). Also, the oxidation does not necessarily lead to formation of secondary VOCs, although some species, e.g., formaldehyde, can be formed through photochemical reactions. Then, not all secondary VOCs can be transformed to SOA. Lines 91-96: This is not an accurate summary of the roles of VOCs in SOA formation. Lines 98-100: I do not get the point why there are strong industrial, vehicular and power plant emissions in mountainous area. Moreover, motor vehicles and power plants are significant sources of NO_x. Then, how to explain the NO_x-sensitive regime for O_3 formation?

Response:

Thank you for your question. We have added professional knowledge into the new section to highlight the impact of land-use types on VOC concentrations, sources and O_3 and SOA formation potentials, specified in newly lines 39-103.

For example, lines 55-57:

"The long-term VOC emission inventory highlighted that there were significant spatial discrepancies of VOC emissions (Li et al., 2019a). The observation campaign also showed that VOC concentrations varied with the sampling sites.

These phenomena were attributed to the fact that VOC concentrations were closely correlated with the land-use types."

Lines 63-65:

"Besides, the land-use types not only influence the VOC concentrations but also the sources especially the anthropogenic sources (Yoo et al., 2015; Chen et al., 2017; Wang et al., 2017; Jookjantra et al., 2022)."

Lines 72-73:

"The diversities of VOC concentrations among the different land-use types could affect the ozone and SOA formation potentials (OFP and SOAFP), resulting in the variations of O_3 and SOA concentrations."

Lines 77-78:

"In detail, atmospheric VOCs undergoes degradation to produce oxidants (HO_2 and RO_2), which further oxidizes atmospheric NO, followed by producing NO_2 and the formation of O_3 finally via the photochemical pathways (Wang et al., 2017). "

Lines 81-85:

"As the key precursor of SOA, VOCs can be oxidized to produce the low VOCs, followed by the formation of SOA via homogeneous nucleation (Merikanto et al., 2009). Moreover, the partitioning of semi-volatile products from VOCs and oxidants gas-phase photochemical reactions to form SOA (Pankow, 1994; Lim et al., 2010). Additionally, low VOCs are produced via the aqueous-phase reactions in atmospheric waters e.g., clouds, fogs, and aerosol water which are largely retained in the particle-phase to generate SOA (Lim et al., 2010)."

Lines 87-94:

"The expanding urbanization and industrialization jointly aggravate the VOC pollution. Moreover, the O_3 concentration at the urban area in Shanghai increased by ~ 67 % from 2006 to 2015 with the growth rate of 1.1 ppbv pear year (Gao et al., 2017). The maximum 1-hour concentration of O_3 could exceed 380 $\mu g\ m^{-3}$ during polluted days (Shi et al., 2015; Gu et al., 2020). Such scenario suggested that O_3 played an important role in atmospheric pollution and

Shanghai was suffering from heavy O_3 pollution. Additionally, the large changes of land-use occurred in Shanghai due to the rapid development e.g., many cultivated areas became urban and/or industrial zones, resulting in the diverse land-use types (Tian et al., 2017). Therefore, Shanghai is regarded as an ideal area to perform atmospheric measurements with the different land-use types."

Comment 5: It is not clear what the authors mean by pollution characteristics, which is too general. It is also not clear what the knowledge gap is. The authors must make it clear what the manuscript adds to the current understanding of VOCs in Shanghai.

Response:

Thank you for your comment. The VOC pollution characteristics involve the concentration variations, primary sources and the impact on O_3 and $PM_{2.5}$ formation. We rewrote some descriptions in the revised manuscript.

For example, line 56:

"The observation campaign also showed that VOC concentrations varied with the sampling sites."

Lines 62-63:

"However, the reported VOC concentrations were widely discussed by single-site measurements, the limited knowledge is available on the multi-site research at a city level."

Lines 653-655:

"Based on the observation data, this study carefully discussed the concentration variations, primary sources, ozone and SOA formation potentials of the atmospheric VOCs influenced by land-use types. "

The knowledge gap is how land-use types influences VOC concentrations, sources and ozone and SOA formation potentials in Shanghai, China. We added the discussion at the Sec. 4. The results herein could provide scientific-based information for policymakers to establish targeted strategies of alleviating VOC pollution. For example, the JS site exhibited higher fractions of aromatics and alkenes, particularly toluene and propylene, than those at the PD and QP sites. The VOC concentration in the early morning (5:00 LT) at the JS site was higher than those at the other two sites.

This result did necessarily correlate with the fact that the JS site is close to the industrial area with heavy industrial emissions, suggesting that industrial activities were key factors of VOC pollution at the JS site. Moreover, the industrial emission and biogenic source showed slight contributions to VOC concentrations at the QP and JS/PD sites, respectively. It was consistent with the regional characteristics of anthrogopenic activities dominated by land-use types. Additionally, the results of VOCs-$S_{PM2.5}$ varied with the land-use types. The aromatics at the JS and PD sites, as well as alkanes at the QP site played crucial roles in the VOC-induced haze pollution. The relevant emission sources, which are thought to be the industrial production at the JS/PD sites and vehicle exhaust at the QP site, should be controlled in priority. Therefore, these findings could provide more information on the accurate VOCs control in Shanghai, China. The results shown herein highlight that the simultaneous multiple-site measurements with the different land-use type in the megacity or city cluster could be more appropriate to fully understand the VOC characteristics relative to a single-site measurement performed normally.

Comment 6: In accurate expressions and grammatical errors are everywhere throughout the manuscript. I cannot list all of them, just give some examples here: line 93: "…that declines the vapor pressure reduction", line 94: pPM, gas-particular partition; line 95: a significantly decreased in the vapor pressure; line 98: transition ~ regime, line 100: strong emissions of industrial, vehicular, power and biogenic, line 101: NO_x transition regime (what is it? I never saw this kind of expression), same for the "VOCs transition area" in line 104; line 106: varied photochemical reactions; line 108: "VOCs are likely to response to the pollution of $PM_{2.5}$ and O_3" –I am not sure if I understand correctly because of language problem; if my understanding is correct, what is the point of studying the responses of VOCs to $PM_{2.5}$ and O_3, rather than the other way around?; line 112: pollution VOC characteristics.

Response:

Thank you for your suggestion. We have revised the statement in the revised manuscript.

For example, lines 62-63:

"However, the reported VOC concentrations were widely discussed by single-site measurements, the limited knowledge is available on the multi-site research at a city level."

Lines 76-78:

"In detail, atmospheric VOCs undergoes degradation to produce oxidants (HO_2 and RO_2), which further oxidizes atmospheric NO, followed by producing NO_2 and the formation of O_3 finally via the photochemical pathways (Wang et al., 2017)."

Lines 82-83:

"Moreover, the partitioning of semi-volatile products from VOCs and oxidants gas-phase photochemical reactions to form SOA (Pankow, 1994; Lim et al., 2010)."

Comment 7: Lines 43-45: What's the point of emphasizing the 57 PAMS VOCs? There are a wide range of VOCs that can be the precursors of O_3 and SOA.

Response:

Thank you for your suggestion. The reason for emphasizing the 57 PAMS VOCs is that these VOCs contribute more on the O_3 formation compared with other VOC species. We revised the sentence in the new manuscript.

"Photochemical Assessment Monitoring Stations (PAMS) have confirmed that totally 57 VOCs, including C_2-C_{10} alkanes, alkenes, alkynes and aromatics are extremely contributed to the formation of O_3 (US EPA, 1990). "

Comment 8: Lines 121-122: Is there any evidence proving that VOC pollution in Shanghai is more serious than ever before? It is contradictory to the statement in lines 71-71 "the VOC concentrations of China have decreased in the recent years along with the effective control strategies".

Response:

Thank you for your comment. The original lines 71 and 121-122 have been deleted.

Comment 9: Lines 122-130: It reads like pollution characteristics just means concentration, which is not true.

Response:

Thank you for your suggestion. The VOC pollution characteristics include the VOC concentrations, sources, ozone and SOA formation potentials. We revised the sentence in the new manuscript.

For example, line 56:

"The observation campaign also showed that VOC concentrations varied with the sampling sites."

Lines 62-63:

"However, the reported VOC concentrations were widely discussed by single-site measurements, the limited knowledge is available on the multi-site research at a city level."

Comment 10: Lines 130-131: I cannot agree. In fact, sources and contributions of VOCs to O_3 and SOA have been well documented.

Response:

Thank you for your suggestion. The original lines 130-131 have been deleted. We added the effect of land-use types on O_3 and SOA formation potentials.

Comment 11: Lines 131-132: Which studies are you referring to when you say "ten years ago" - a specific time frame? At least, the studies you are referring to should be discussed.

Response:

Thank you for your comment. The original lines 131-132 have been deleted. We added the effect of land-use types on VOC concentrations.

Comment 12: Major comment: Different instruments, as well as analytical methods, were used for the analysis of VOCs at the three sites. How did the authors reconcile the data so that they can be compared? What is TD300 (line 177) that is not defined? In general, the small molecule and large VOCs are detected by FID and MSD, separately. Lines 182-184: What are the accuracies and detection limits for the minority of the species, i.e., those beyond the "95% and most VOC components", and what is the range of the precision? Line 188: What is the SEAS site? It has never been defined before.

Response:

Thank you for your comment. The three instruments (GC5000, GC580+TD300, GC866) all can be used to analyze VOCs. Their actual attainments were very similar in practice.

By using GC580+TD300, all PAMS substances meet the standard "curve correlation coefficient ≥ 0.995", all substances meet the standard "precision ≤ 10%", more than 95% of the target compounds meet the standard "accuracy ≤ ± 20%", all target compounds meet the standard "detection limit ≤ 0.15ppb", and more than 90% of the target compounds have blank response less than 0.1 ppb.

By using GC 5000 BTX/VOC, more than 90% of PAMS substances meet the standard "curve correlation coefficient ≥ 0.995", all substances meet the standard "precision ≤ 10%", more than 95% of the target compounds meet the standard "accuracy ≤ ± 20%", more than 98% of the target compounds meet the standard "detection limit ≤ 0.15ppb", and more than 95% of the target compounds have blank response less than 0.1 ppb.

By using GC866, more than 95% of PAMS substances meet the standard "curve correlation coefficient ≥ 0.995", all substances meet the standard "precision ≤ 10%", more than 95% of the target compounds meet the standard "accuracy ≤ ± 20%", more than 90% of the target compounds meet the standard "detection limit ≤ 0.15ppb", and more than 90% of the target compounds have blank response less than 0.1 ppb.

Because VOCs in three sites were collected and analyzed separately, we used the most suitable detection instruments for the three stations.

Lines 138-139:

> *"At the PD site, VOCs was measured by gas chromatography (GC580-FID, PE, USA) and TD300 (a transformer driver)."*

Lines 150-153:

> *"The meteorological variables including temperature, RH and wind speed were simultaneously acquired from a weather station about 10 km northwest of the Shanghai Academy of Environmental Sciences."*

Comment 13: Description about this method in section 2.3 is confusing. How can the spatial heterogeneity be determined for a single site, as stated in line 190? Line 193: Does j represent site or dataset? Contradictory descriptions.

Response:

Thank you for your comment. We revised the sentence in the new manuscript.

Lines 154-155:

"The spatial heterogeneity of VOC concentration between two different sites was determined by the coefficient of divergence (COD) (Wongphatarakul et al., 1998; Sawvel et al., 2015)."

Line 157:

"where x_{ij} presents the mass concentration in i time, j and k are two datasets, p presents the number of observations."

Comment 14: Lines 202- 203: the descriptions of g_{ik}, f_{kj} and e_{ij} are totally wrong. Lines 204-207: The function Q is introduced. However, it is not clear what the purpose of introducing it is and how the authors used it? Lines 210-212: "EF is the error faction and can be set to 0.05-0.2" -What was the EF the authors set in this study? How did the authors determine the solution with seven factors as the optimal one?

Response:

Thank you for your comment. We revised the sentence in the new manuscript.

Lines 166-167:

"g_{ik} represents the species contribution of the kth source to the ith sample, f_{kj} is the jth species fraction from the kth source, e_{ij} is the residual result for jth species in ith sample."

Lines 174-180:

"where MDL is the minimum detection limit, EF is the error fraction and can be set to 0.05-0.2 (Song et al., 2007). It was 0.1 in this study. In this study, four to eleven factors were utilized to determine the option solution. Q_{true}/Q_{robust} and and $Q_{true}/Q_{expected}$ are important parameters for characterizing the rationality of the PMF results (Brown et al., 2015). Seven factors were regarded as the optimal solution, comparing the ratios of Q_{true}/Q_{robust}, $Q_{true}/Q_{expected}$ and the PMF results. The Q_{true}/Q_{robust} values were set to 1.0 at the three sampling sites. The $Q_{true}/Q_{expected}$ values were 1.3, 1.1, and 1.0 at the JS, PD and QP sites, respectively."

Comment 15: Lines 223-225: How did you determine the number of polluted and all trajectories in a grid, and how was the weight function W_{ij} applied?

Response:

Thank you for your comment. In this study, the pollution trajectory was defined as the trajectories corresponding to the total VOC (TVOC) concentration that exceeded the 75th percentile concentration of TVOCs. The m_{ij} is the number of endpoints of the pollution trajectory passing through the grid (i, j), and n_{ij} is the number of endpoints of all the trajectories falling within the grid (i, j).The weight function W_{ij} was used to increase the model accuracy.

"Therefore, the $PSCF_{ij}$ can be calculated using the Eq. (7) as follows:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \times W_{ij} = \frac{m_{ij}}{n_{ij}} \times \begin{cases} 1.00 & 80 < n_{ij} \\ 0.70 & 20 < n_{ij} \le 80 \\ 0.42 & 10 < n_{ij} \le 20 \\ 0.05 & n_{ij} \le 10 \end{cases} \quad (7)"$$

Comment 16: The equation (9) calculates the responses of VOC to O_3, which is opposite to the statement in lines 245-246 that "The characteristic structure and reactivity could influence the contribution of VOCs to O_3 formation". Lines 249-252: The rationality of using 100 as a threshold of background O_3 should be justified. Why 100? Note that it is a quite high value, especially in cool seasons. It is also totally wrong to assign the VOC concentrations during the O_3-background time period as background VOC concentrations. In most cases, the patterns of O_3 and VOCs are inconsistent. For example, O_3 got lowest values at night and in early morning when VOCs are at high levels. Lines 251-252: What is the logic behind? Why is VOCs influenced by the variation of O_3, and not the other way around? Lines 254-255: The logarithmic conversion is also problematic. Equation 12 should be written as lny = lna + blnx.

Response:

Thank you for your comment. The calculation of VOC-O_3 sensitivity (VOCs-S_{O3}) was deleted after consideration.

The equation 11 was rewritten.

"$\ln y = \ln a + b \cdot \ln x$"

Comment 17: Inaccurate expressions and grammatical errors in this section include but are not limited to the followings. Lines 180-181: "The samples were condensed low-carbon (C_2-C_6) compounds and high-carbon (C_6-C_{12}) compounds …"; Line 185: "trace instruments"; Lines 211-212: "option solution", "greatest solution"; Line 214: "observe the back trajectories, source and direction of pollutants"; Line 218: "This study was determined the 24-h back trajectory".

Response:

Thank you for your comment. We have revised the statement in the new manuscript.

For example, lines 142-143:

"The samples were condensed for low-carbon (C_2-C_5) compounds at 15℃ and high-carbon (C_6-C_{12}) compounds at 30℃."

Lines 148-149:

"The O_3, NO-NO_2-NO_x were characterized by trace gas instruments (49i ozone analyzer and 42i nitrogen oxide analyzer, produced by Thermo Environmental Instruments Inc., USA) with the detection limits of 0.50 and 0.40 ppb, respectively."

Lines 175-177:

"In this study, four to eleven factors were utilized to determine the option solution. Q_{true}/Q_{robust} and and $Q_{true}/Q_{expected}$ are important parameters for characterizing the rationality of the PMF results (Brown et al., 2015). Seven factors were regarded as the optimal solution, comparing the ratios of Q_{true}/Q_{robust}, $Q_{true}/Q_{expected}$ and PMF results."

Lines 182-184:

"PSCF and Cluster were widely used to determine the back trajectories, source and direction of pollutants (Draxier and Hess, 1998; Hong et al., 2019; Liu et al., 2019), and designed to measure the potential VOC source and primary transport pathway of trace elements (Ashbaugh et al., 1985; Xie et al., 2007; Zheng et al., 2018; Liu et al., 2020)."

Lines 185-186:

"This study was determined by the 24-h back trajectories (one hour interval) at the height of 500 m via the MeteoInfoMap software."

Comment 18: Line 266: "60 VOC species" is contradictory to the statement that "Totally 43 species of VOCs were observed" in line 183.

Response:

Thank you for your comments. We have revised the statement in the new manuscript.

Lines 263-265:

"During the observation campaign, 43 VOC species including 16 alkanes, 11 alkenes, 16 aromatics and 1 alkyne were measured and the contributions of total VOCs (TVOCs) > 1 % were marked."

Comment 19: Lines 271-273: What's the point of comparing the wind speed that is very spatially uneven?

Response:

Thank you for your comment. We want to highlight that the wind speed in our study is higher than those in the other studies. We revised the sentence in the new manuscript.

Lines 238-240:

"The wind speed at the QP site (4.37 ± 1.47 m s^{-1}) was 2.29 and 1.36 times higher than those at the JS (1.91 ± 0.49 m s^{-1}) and PD (1.30 ± 0.62 m s^{-1}) sites, respectively, indicating the decreased dilution and diffusion conditions at the latter two sites."

Comment 20: Lines 274-288: The comparisons are rather simple. Are there same number of species, same species, same sampling season and etc.? Without discussion on these factors, the comparisons are meaningless.

Response:

Thank you for your comment. We rewrote the description in the revised manuscript.

"Compared with the relevant measurements performed previously in Shanghai at the same sampling sites, this study generally presented lower VOC concentrations (Cai et al., 2010b; Zhang et al., 2018; Zhang et al., 2020a). In

detail, at the JS site, the VOC concentration was approximately 4 times lower than the measurement of Zhang et al., (2018) (94.14 ppb). At the PD and QP sites, the results in this study were slightly lower than those reported by Cai et al. (2010b) (24.3 ppb) and Zhang et al. (2020a) (15.41 ppb). A variety of control strategies, such as prohibiting of fireworks in the open air, improving VOC detection standards and strengthening control technology were implemented, thus resulting in the low VOC concentrations herein. Particularly, the policy of "one factory, one strategy", targeted at mitigating VOC emissions, was published by Shanghai government in 2018."

Comment 21: Lines 295-312: I do not see the necessity of discussing such simple facts with too many words.

Response:

Thank you for your comment. The original lines 295-312 have been deleted.

Comment 22: I am surprised to see such high levels of O_3 in the sampling period. Without any doubt, the authors made mistakes in calculation or unit conversion.

Response:

Thank you for your comment. We revised the unit in the new manuscript.

Lines 249-251:

"During the observation period, the average $PM_{2.5}$ values were 45.57 ± 27.59, 48.51 ± 27.22 and 40.27 ± 27.78 μg m^{-3}, and the mean O_3 concentrations were averaged to be 73.59 ± 23.59, 57.48 ± 20.49 and 99.30 ± 24.00 μg m^{-3} at the JS, PD and QP sites, respectively."

Comment 23: Lines 315-317: Readers would have no idea what the point of this discussion is. Are the dates special?

Response:

Thank you for your comment. We wanted to illustrate when the minimum hourly $PM_{2.5}$ levels occurred, and highlight that VOCs was positively correlated with $PM_{2.5}$ while was negatively correlated with O_3. The original lines 315-317 have been deleted.

Comment 24: Lines 317-318: I do not think this was the reason for the correlation. Otherwise, did you see correlation between VOCs and O_3, where the former was also precursors of the later?

Response:

Thank you for your comment. We revised the sentence in the new manuscript.

Lines 269-270:

"VOCs was found to be positively correlated with $PM_{2.5}$, and the pearson correlation coefficients ($R_{Pearson}$) were 0.58, 0.71 and 0.25 at the JS, PD and QP sites, respectively."

Comment 25: Lines 320-324: Why not refer to sources of $PM_{2.5}$ and VOCs in Shanghai. Transportation as the main source of $PM_{2.5}$ and VOCs in different cities does not necessarily mean the homology $PM_{2.5}$ and VOCs in Shanghai.

Response:

Thank you for your comment. We added the references documenting the sources of $PM_{2.5}$ and VOCs in Shanghai.

Lines 272-275:

"Moreover, $PM_{2.5}$ and VOCs present similar emission sources. For example, traffic exhaust was proven as the predominant contributor for both of them (Li et al., 2009; Cai et al., 2010a; Cai et al., 2010b; Wang et al., 2013; Kuo et al., 2014; Liu et al., 2019)."

Comment 26: Lines 325-331: First, I do not think the correlation is worth discussing. In most cases, the diurnal patterns of VOCs and O_3 are opposite. Second, the opposite patterns are mainly due to inconsistent patterns of VOC emission (e.g., emissions in morning and evening rush hours) and O_3 formation (e.g., daytime). The discussions are far-fetched and I do not understand "and counteraction was imposed by uncertain factors during the formation of O_3."

Response:

Thank you for your comment. We rewrote the description in the revised manuscript.

"However, the VOC concentrations were negatively correlated with O_3 ($R_{Pearson}$ = -0.24 at the JS site, $R_{Pearson}$ = -0.48 at the PD site and $R_{Pearson}$ = -0.25 at the QP

site, respectively). The termination and titration ($NO + O_3 \rightarrow NO_2 + O_2$) were more efficient and lots of factors including sunshine duration, temperature and relative humidity rather than the emission of precursors, impacted on the surface O_3. Li et al. (2019b) emphasized that the absolute concentration of precursor was not the only factor during the O_3 formation in Zhengzhou, China."

The other sections are revised in the new manuscript. Please review our revised manuscript. We greatly appreciate any comments and valuable suggestions from the reviewer. Thank you for your time in handling our manuscript.

Lastly, we would again express our appreciation to the reviewers and editor for their warmhearted help. Thank you very much!

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