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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contributions of different anthropogenic volatile organic compound sources to ozone formation at a receptor site in the Pearl River Delta region and its policy implications" *by* Z. He et al.

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Response to Reviewers We appreciate the two anonymous reviewers for their constructive criticisms and valuable comments, which were of great help in improving the quality of the manuscript. We have revised the manuscript accordingly and our detailed responses are shown below. All the revision is highlighted in the revised manuscript.

Reviewer #3

General comments The manuscript enhances current understanding of ozone forma-

tion in the Pearl River Delta, China. The authors analyze one month of measurements made at a receptor site using positive matrix factorization (PMF) and a parameterization of photochemical age to identify and quantify contributions of four emission source types to VOC mass concentration. A box model is used to calculate relative incremental reactivities (RIR) and source contributions to ozone formation. The model is also used to develop ozone response surfaces as a function of VOC and NOx emission reductions.

The manuscript states that one important motivation for their study is the need to determine how photochemical processing affects the accuracy of PMF applications to VOC source apportionment in the study region (p 4, | 80 - 85). It would be informative to know how much difference the calculation of photochemical processing made to the PMF results (sections 3.2.1 and 3.2.2). A simple comparison of PMF results with and without the adjustment for photochemical losses would clarify how important it is to do the photochemical loss calculation and adjustment of species concentrations. Such a comparison might make the manuscript more general and of greater interest to a wider audience. Reply: The reviewer's suggestion is highly appreciated. Figure 4 was revised in order to present the difference on VOCs concentrations with and without the adjustment. Furthermore, in order to investigate the influence of photochemical processing on the source apportionment of VOCs, a comparison of PMF results with and without the adjustment of VOC concentration due to photochemical losses was conducted. Similar sources profiles and the same four anthropogenic sources (i.e., diesel vehicular emission, solvent usage, biomass burning, and gasoline vehicular emission were identified, however the sources made different contributions to VOC abundance compared to those with the adjustment of VOC concentrations (more details were provided in the supplementary). For example, higher contributions of solvent usage and biomass burning and lower contributions of diesel vehicular emission were found in the scenario without adjustments than that with adjustments, which were related to the relatively higher photochemical reactivity of main species in diesel vehicular emission than those in solvent usage and biomass burning. The above brief discussion

on the difference in source apportionment results with and without the adjustment of VOCs has provided in the revised manuscript (Lines 4-13, Page 17), while the detailed discussion was provided in the supplementary.

The authors correctly note that their "results could provide valuable information, facilitating local and regional policy-makers. . ." Their findings are indeed valuable for the specific region studied. The manuscript could have wider significance if the authors provided more comment about how their methods or their results would be of interest to researchers or policymakers in other geographical areas. Reply: The reviewer's suggestion is highly appreciated. To highlight the significance of this study and provide more statement on how the results of this study would be of interest to researcher or policymakers in other geographical areas, the conclusion section was combined with the policy implication section as follows: "The PRD region has long been facing severe photochemical air pollution, and VOCs has been the limiting factor of O3 formation in this region. To better understand the contribution of different anthropogenic VOCs to O3 formation in this region, we performed in-depth analyses on data obtained from intensive measurements of VOCs and related species conducted at a downwind rural site (Heshan site, HS) of the PRD region during October – November, 2014. Four anthropogenic sources were identified by the PMF model with the consideration of the influence of photochemical processing. The O3 formation at the HS was generally VOC-limited, with the vehicular emission (especially gasoline vehicular emission) as the most important anthropogenic VOC source contributing to O3 formation, followed by biomass burning. It indicated that priority should be given to controlling vehicular emission and biomass burning. Furthermore, with the current industries operating in the PRD region, particular attention should be given to toluene, xylenes, ethylbenzene, ethene and 1-pentene in efforts to control photochemical pollution. Indeed, many additional policies on VOCs have been and continue to be implemented and formulated in the PRD region. A series of policies regarding the control of vehicular emission have been conducted in the PRD region, the purposes of which can be mainly divided into two categories: 1) improve the environmental standards of the main air pollutants

and standards of emissions; and 2) improve the quality of the fuel used in vehicles. Policies on controlling biomass burning, however, are relatively limited. Nevertheless, some policies have been effective, and levels of NOx (another important O3 precursor), have decreased in the PRD region in recent years. On the other hand, for VOCs, most relevant policies only control the total mass and/or the total emissions of VOCs, and the level of O3 continues to increase in this region. To prevent net O3 increment, the VOCs and NOx should be controlled in an appropriate ratio since VOCs and NOx were frequently controlled simultaneously. Furthermore, long-term monitoring is still needed to evaluate the benefits and dis-benefits of the control measures on vehicular emissions and/or photochemical pollution in the PRD region. Overall, the results of this study will be valuable for facilitating local and regional policy-makers to propose appropriate strategies and effective control measures of VOCs and photochemical pollution in other regions of China, especially where O3 formation is VOC-limited. However, it is noteworthy that the above results were obtained based only on measurements taken over one month, and in a specific season (i.e., autumn), which may only represent the characteristics of photochemical pollution in autumn at a receptor site in the PRD region." For details, please refer to Line 2, Page 27 – Line 6, Page 28 in the revised manuscript.

The absence of biogenic VOC (e.g., isoprene) contributions to ozone formation is of concern (please see specific comments). Since the results were determined for a one-month autumn sampling period, additional discussion of the applicability of the findings to other seasons is merited. Readers will need some additional information to evaluate the accuracy of the box model simulations (please see specific comments). Reply: The reviewer's comment is highly appreciated. We agree with the reviewer and consider that the result extracted from the PMF model might not applicable to all seasons because it was based on a one-month autumn measurement. Detailed responses to the individual specific comment/suggestion are as follows.

Specific comments Please note that the "hundreds" digits of the line numbers are not

visible in the PDF, so I refer to both page number and line number throughout this review. Reply: Thanks for the reviewer's consideration. The format of line numbers has been revised according to ACP's guideline in the revised manuscript.

p. 7, | 32 – 34. The exclusion of biogenic VOC measurements as inputs to PMF is a limitation. Because the measurement period was October 22 to November 20, 2014, it is possible that biogenic emissions were much lower than would be typical of spring or summer and the exclusion of biogenic VOC concentrations from the PMF inputs may not affect the conclusions for this study period. However, there should be some acknowledgment that the results pertain to a fall study and may not represent other seasons. Reply: The reviewer's suggestion is highly appreciated. The mean concentrations of biogenic species (i.e., isoprene) during the sampling period was 150 $\pm$ 17 pptv, much lower than those observed in summer at other rural sites in the PRD region ($\sim$250-1400 pptv) (Lau et al, 2010; Ding et al., 2012; Lu et al., 2012; Yuan et al., 2018). Furthermore, previous studies have reported that VOC abundance at the PRD region was mostly controlled by anthropogenic emissions, while controlling anthropogenic VOCs emissions seems to be more feasible than biogenic emissions (Tsui et al., 2009; Leung et al., 2010; HKEPD, 2012; Ling and Guo, 2014). As the aim of this study was to quantify the contributions of anthropogenic emissions to ambient VOCs and evaluate their contributions to photochemical O3 formation at a receptor site of the PRD region, here we only used the anthropogenic VOCs as input for the PMF model (Barletta et al., 2008; Liu et al., 2008; Zheng et al., 2010; Lu et al., 2012; Tan et al., 2012). Moreover, we agree with the reviewer and consider that the result extracted from the PMF model might not applicable to all seasons because it was based on a one-month autumn measurement. To clarify it, the following text has been added in the manuscript: ". . .. . .the results of this study will be valuable for facilitating local and regional policy-makers to propose appropriate strategies and effective control measures of VOCs and photochemical pollution in other regions of China, especially where O3 formation is VOC-limited. However, it is noteworthy that the above results were obtained based only on measurements taken over one month, and in a specific

season (i.e., autumn), which may only represent the characteristics of photochemical pollution in autumn at a receptor site in the PRD region." For details, please refer to Lines 1-6, Page 28 in the revised manuscript.

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p 8, | 52 – 54. Please clarify if biogenic VOCs were included among the inputs to the box model. The RIR values will not be correct if biogenic VOCs were excluded from the inputs to the box model. Reply: Thanks for the reviewer's comment. In this study, only one biogenic species (isoprene) was quantified at the Heshan site. We agreed with the reviewer that if the whole chunk of biogenic VOCs were excluded as inputs in photochemical box model, there will be bias for the model simulation. Actually, the simulation of PBM-MCM in this study included the observed concentrations of biogenic species (isoprene). To clarify that biogenic species, i.e., isoprene, was input into the PBM-MCM model, the text has been revised as follows: "In this study, the hourly data of VOCs, including both anthropogenic and biogenic species, five trace gases (i.e., O3, NO, NO2, CO, and SO2) and two meteorological parameters (i.e., temperature and relative humidity) measured during the campaign were used as the model input." For details, please refer to Lines 17-19, Page 9 in the revised manuscript.

p 8, | 52 – 54. Please clarify that the model was applied to each individual day in the study for the RIR calculations (rather than that hourly data were averaged across days

and then input as averages to the model as was later done to generate the ozone isopleths). The citations to Lyu et al. (2016) and Wang et al. (2017) imply that the model was applied to each individual day (as done in those studies) for the RIR calculations but it would be helpful to be explicit in this paragraph. Reply: Thanks for the reviewer's great suggestion. Indeed, similar to Lyu et al. (2016), the PBM-MCM model was applied to the observed data collected in each individual day for the RIR calculation during the whole sampling period. To highlight the above model configuration, the following text has been added in the revised manuscript: ".....Similar to Lyu et al. (2016), the PBM-MCM model was applied to the observed data collected on each individual day for the RIR calculation during the whole sampling period, while the hourly data during the whole sampling period were averaged across sampling days to provide mean diurnal variation as a base-case input for the PBM-MCM model to generate the O3 isopleths." For details, please refer to Lines 19-23, Page 9 in the revised manuscript.

Reference Lyu, X., Guo, H., Simpson, I. J., Meinardi, S., Louie, P. K. K., Ling, Z., Wang, Y., Liu, M., Luk, C. W. Y., Wang, N., and Blake, D. R.: Effectiveness of replacing catalytic converters in LPG-fueled vehicles in Hong Kong, Atmos. Chem. Phys., 16, 6609-6626, https://doi.org/10.5194/acp-16-6609-2016, 2016.

p 8, | 64. It might be helpful to say "net O3 production plus NO consumed" rather than "net O3 production and NO consumed" (the equations are correct). Reply: Thanks for the reviewer's suggestion. The text has been revised accordingly. For details, please refer to Line 7, Page 9 in the revised manuscript.

P 9, | 95 – 96. If O3 formation at the site is VOC-limited as previously discussed, it isn't likely that photochemical processing of the arriving air masses is truly complete. Even if NOx mixing ratios are too low to sustain further O3 production in the arriving air, injection of fresh NOx emissions into aged air masses would permit further photochemical processing. The next section (3.2.1) indicates that photochemical aging was relatively incomplete (high correlations between more reactive and less reactive species; similarities between initial and observed VOC species concentrations except

for highly reactive species such as some alkenes and aromatics). Reply: Sorry for the mistake and confusion it caused. We agree with the reviewer that the air mass arrived at the site was not through complete photochemical processing. Furthermore, we also agreed with the reviewer #1's comment that variations of VOCs were related to the variations on sources other than photochemical processing. Therefore, the text has been deleted accordingly in the revised manuscript. For details, please refer to Line 24, Page 10 in the revised manuscript.

P 11, Table 1. It would be helpful to include summary statistics for NO and NO2. Reply: Thanks for reviewer's great suggestion. The summary statistics for NO and NO2 have been added in Table 1, and the following text has been added in the revised manuscript: "The mean mixing ratios of NO and NO2 were 4.2 $\pm$ 0.4 ppbv and 39.9 $\pm$ 1.2 ppbv, respectively, at the HS during the measurement, and the relatively low ratio of NO/NOx indicated that the site was distant from the source areas (Qin and Zhao, 2003; Melkonyan and Kuttler, 2012; Hagenbjörk et al., 2017)." For details, please refer to Lines 12-15, Page 10 in the revised manuscript.

Table 1. Average, range, and standard deviation of concentrations for NOx (i.e., NO and NO2) and the eight most abundant volatile organic compounds (VOCs) measured at the Heshan site, together with a sum of the mixing ratios for each hydrocarbon category (i.e., alkanes, aromatics, and alkenes). Species Average $\pm$ standard deviation (ppbv) Range (ppbv) NO 4.22 $\pm$ 5.58 0.50 - 35.35 NO2 39.92 $\pm$ 16.12 8.02 - 130.77 Ethane 3.86 $\pm$ 1.34 1.08 - 10.44 Toluene 3.74 $\pm$ 2.89 0.56 - 15.80 acetylene 3.42 $\pm$ 2.33 0.11 - 28.22 Propane 3.01 $\pm$ 1.82 0.45 - 10.70 Ethene 2.94 $\pm$ 3.34 0.37 - 64.56 i-Pentane 1.90 $\pm$ 2.20 0.22 - 16.16 n-Butane 1.85 $\pm$ 1.28 0.20 - 10.10 m/p-Xylene 1.62 $\pm$ 1.44 0.17 - 12.74 Alkanes 17.04 $\pm$ 10.64 2.04 - 61.01 Aromatics 9.07 $\pm$ 7.01 1.46 - 40.12 Alkenes 5.29 $\pm$ 5.01 0.67 - 77.39

P 17, section 3.3. For readers to understand the accuracy of the box model, it would be helpful to provide model performance statistics for the base case at the beginning of this section. Reply: Thanks for reviewer's helpful suggestion. The following text has been

added to evaluate the model performance: "To quantitatively evaluate the performance of the O3 simulation, the index of agreement (IOA), which was widely used for evaluation of PBM-MCM model (Wang et al., 2015; Wang et al., 2017; Liu et al, 2019) was introduced. The IOA was calculated by Eq. (7) (Huang et al., 2005): IOA=1-($\sum$_(i = 1)n($O\_i - S\_i$)2)/($\sum$_(i = 1)n($|O\_i - O| + |S\_i - O|$)2), (7) $where O i and S i represented obse$ suggesting the abundance and variation of O3 were reasonably reproduced and could be used for further calculation." For details, please refer to Line 8, Page 19 – Line 5, Page 20 in the revised manuscript.

P 18, Figure 8 and related text. The RIR calculation for each species depends on the concentrations of all other species. If biogenic VOCs (e.g., isoprene) were not included as inputs to the box model, the reported RIR values will not represent the "real-world" condition. If biogenic VOCs were included as inputs to the box model, Figure 8 should show their RIRs and relative contributions. Please note in the caption what the error bars represent (e.g., one or two standard errors of the means or a different measure of variation?) Reply: The reviewer's comment is highly appreciated. We agreed with the reviewer that if the whole chunk of biogenic VOCs were excluded as inputs in photochemical box model, there will be bias for the model simulation due to it may not reflect the real atmospheric environment. In this study, only the most important biogenic species in this region, i.e., isoprene, was quantified at the Heshan site, which were actually included in the PBM-MCM model. In the revised manuscript, a discussion on the RIRs and relative contributions of VOC species have been added as follows, together with the description for the error bars: "Furthermore, Fig. 8c-f also showed the mean RIR values and the contributions to photochemical O3 formation for the top 10 VOC species and groups at the HS. Aromatics had the highest RIR value, with the average contribution of $\sim$82% to the sum RIR of all VOCs, followed by alkenes ($\sim$11%) and alkanes ($\sim$7%). Among the individual VOC species, toluene and m/p-xylene made most significant contribution (with a relative contribution of $\sim$40% and $\sim$34%, respectively) to O3 formation at the site when both the reactivity and abundance of VOC species were considered. The PMF results suggested that aromatics (including

toluene, xylenes, and ethylbenzene) were mainly from gasoline vehicular emission and solvent usage, while alkenes were mainly related to diesel vehicular emission. Overall, gasoline vehicular emission was the dominant contributor to O3 formation at the HS, and greater efforts should be devoted to toluene, xylenes, ethylbenzene, ethene, and 1-pentene for effectively controlling photochemical pollution." On the other hand, the error bars in Fig. 8 represented one standard errors of the mean RIR values. For details, please refer to Line 17, Page 20 – Line 7, Page 20 in the revised manuscript.

P 18, | 37 - 38. The phrase "based on the average diurnal variation" suggests that hourly data were averaged across days to provide a single base-case input for the box model's generation of ozone isopleths, which would be consistent with the approach described by Lyu et al. (2016). Please expand this description slightly. Do the 58 VOCs include isoprene and alpha- or beta-pinene? Reply: Thanks for the reviewer's comment. The description on PBM-MCM model running has been expanded in the revised manuscript. The simulation of PBM-MCM in this study indeed included the concentration of biogenic species (i.e., isoprene). If the whole chunk of biogenic VOCs were excluded as inputs in photochemical box model, there will be bias due to it cannot reflect the real atmospheric environment. It has been added or advised as follows: "In this study, the hourly data of VOCs, including both anthropogenic and biogenic species, five trace gases (i.e., O3, NO, NO2, CO, and SO2) and two meteorological parameters (i.e., temperature and relative humidity) measured during the campaign were used as the model input. Similar to Lyu et al. (2016), the PBM-MCM model was applied to the observed data collected on each individual day for the RIR calculation during the whole sampling period, while the hourly data during the whole sampling period were averaged across sampling days to provide mean diurnal variation as a base-case input for the PBM-MCM model to generate the O3 isopleths." ".....The PBM-MCM model was employed based on the average hourly observed data across days to provide a single base-case input for the box model's generation of O3 isopleths. ....." For details, please refer to Lines 17-23, Page 9 and Lines 11-13, Page 21 in the revised manuscript.

P 19, | 61. Even in the NOx-limited regime, VOC reductions never increase ozone formation. Consistent with past studies, the isopleths are nearly vertical on the left side of Figure 9b and indicate a very small ozone decrease in response to VOC reductions at fixed NOx. Reply: Sorry for the mistake. It has been revised as follows: "On the other side of the ridge, for the reduced NOx to 7.5-15% of original mixing ratio, O3 concentration decreases with NOx concentration, but decreased VOCs would lead to minimal O3 variation." For details, please refer to Lines 17-19, Page 22 in the revised manuscript.

Pages 21 – 22. Many readers will not be able to follow this discussion as written. Reply: Thanks for the reviewer's patience and suggestion. It has been rewritten in the revised manuscript. For details, please refer to Line 7, Page 23 – Line 10, Page 26.

p. 23, | 34 – 35. Does this conclusion apply to other seasons? Reply: Thanks for the reviewer's comment. We consider that the results might not applicable to all seasons because it was based on a one-month autumn measurement. To clearly clarify that the results of this study was obtained based on the one-month measurement at autumn at a receptor site of the PRD region, the following text has been added in the manuscript: ".....the results of this study will be valuable for facilitating local and regional policy-makers to propose appropriate strategies and effective control measures of VOCs and photochemical pollution in other regions of China, especially where O3 formation is VOC-limited. However, it is noteworthy that the above results were obtained based only on measurements taken over one month, and in a specific season (i.e., autumn), which may only represent the characteristics of photochemical pollution in autumn at a receptor site in the PRD region." For details, please refer to Lines 1-6, Page 28 in the manuscript.

Minor comments Abstract. The abstract uses a term ("abatement ratio") that is defined in the text (page 21), requiring a full page of explanation there. Another full page (p 22) is needed to define and discuss the abatement ratios of the individual source types. To aide readers of the abstract who will not have already read pages 21 - 22, I suggest

revising the following sentence: “Sensitivity analysis indicated that in order to prevent the increment of O3 concentration, the abatement ratios of the individual VOC source vs. NOx should be higher than 3.8, 4.6, 4.6, and 3.3, respectively, for diesel vehicular emission, solvent usage, biomass burning, and gasoline vehicular emission, respectively.” A sentence such as the following appears to me to better convey the intended meaning: “Sensitivity analysis indicated that combined VOC and NOx emission controls would effectively reduce incremental O3 formation when the ratios of VOC-to-NOx emission reductions were higher than 3.8 for diesel vehicular emission, 4.6 for solvent usage, 4.6 for biomass burning, and 3.3 for gasoline vehicular emission.” Reply: We thank the reviewer’s helpful suggestion. It has been revised accordingly as follows: “Sensitivity analysis indicated that combined VOC and NOx emission controls would effectively reduce incremental O3 formation when the ratios of VOC-to-NOx emission reductions were > 3.8 for diesel vehicular emission, > 4.6 for solvent usage, > 4.6 for biomass burning, and 3.3 for gasoline vehicular emission.” For details, please refer to Line 25, Page 1 – Line 3, Page 2 in the revised manuscript.

P 3, | 71-73. Please clarify if the percentages refer to percent of VOC mass or percent of VOC reactivity. Reply: Sorry for the mistake and confusion it caused. It has been revised as follows: “Results from source apportionment using the PMF model demonstrated the important roles of vehicular emissions in ambient VOCs in urban and suburban environments of Hong Kong, accounting for 48-54% and 31-40% of the concentrations of VOCs, respectively (Lau et al., 2010; Guo et al., 2011a).” For details, please refer to Line 24, Page 3 – Line 1, Page 4 in the revised manuscript.

P 5, | 99-100. The map shows that the site is southwest of Guangzhou and Foshan, not northeast (alternatively, the cities are northeast of the site). Reply: Sorry for the mistake. It has been revised accordingly in the revised manuscript (Line 3, Page 5).

P 13, eq. 6 and | 53 – 54. For consistency, please use either kVOC or kNMHC in both eq. 6 and the text. Reply: Thanks for pointing this out. It has been uniformly expressed as “kVOC” in the revised manuscript (Line 13, Page 14).

p. 20. Figure 9 is confusing for two reasons. First, it would be easier to understand the figure if the two panels were swapped (right replacing left). Second, the axes are also reversed from the more common presentation format. Reversing the vertical and horizontal axes and then placing the current left panel above the current right panel would be consistent with customary presentations (NOx reductions on the vertical, VOC reductions on the horizontal axis). Reply: Thanks for the great suggestion. It has been revised accordingly in the revised manuscript (Fig. 9, Page 23).

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-1293/acp-2018-1293-AC1-supplement.pdf

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