Dear editor:

Here we submit our revised manuscript for consideration to be published on **Atmospheric Chemistry and Physics**. The further information about our manuscript is as follows:

Topic: Measurement report: Ambient volatile organic compounds (VOCs) pollution at urban Beijing: characteristics, sources, and implications for pollution control

Type of Manuscript: article

Authors: Lulu Cui[1], Di Wu[1], Shuxiao Wang[1,2*], Qingcheng Xu[1], Ruolan Hu[1], Jiming Hao[1, 2]

Corresponding author:

Shuxiao Wang. Address: School of environment, Tsinghua University, Beijing 100084, China; Tel.: (+86)20- 62771466; E-mail: shxwang@tsinghua.edu.cn.

We acknowledge the suggestions of the editor, and are also grateful to your efficient serving. We have updated the manuscript on the basis of these valuable comments. Our responses were listed as following:

Main Comments

Comment 1: From the abstract and text (Sect. 2.1), it is not made clear to the reader that these are not continuous measurements. For instance, there are no measurements from mid-January to mid-April or from mid-July to mid-September. This point should be made more transparently in the manuscript. Additionally, the potential limitations/biases of the analysis, particularly for the "ozone polluted months" given

these measurement limitations should be briefly discussed.

Response (Lines 11-16, 74-76, 344-351): Thanks for the editor's suggestion. In the revised version, the observation periods have been clearly described in the abstract section as below:

"In this work, four intensive field measurements of VOCs during winter of 2018 (from 1 December of 2018 to 17 January of 2019), spring (15 April to 27 May), summer (17 June to 13 July) and autumn (22 September to 27 November) of 2019 were conducted at an urban site in Beijing to characterize VOCs sources and their contributions to air pollution."

The observation periods have also been clearly described in the last paragraph of the introduction section as below:

"In this work, ambient air samples were collected at an urban site in Beijing from December 2018 to mid-January 2019, mid-April to late May 2019, mid-June to mid-July 2019, and late September to late November 2019, respectively".

The potential limitations of the analysis due to the measurement limitations have been added in the revised version as below:

"3.4 Limitation

This study analyzed the VOC sources and their contributions to O_3 and SOA formation across different seasons. It should be pointed out that the sampling campaign for VOCs measurement was not conducted continuously during December 2018 and November 2019. For instance, the air samples were not collected in August and February-March of 2019, during which the pollution events of O_3 and $PM_{2.5}$ occurred, respectively. The

variations, sources and secondary transformation potentials of VOCs, particularly for O_3 and $PM_{2.5}$ pollution periods cannot be fully depicted. Despite the uncertainties that remained, the results obtained in this study provide useful information for VOCs emission control strategy and assist overcoming air pollution issue in Beijing."

Comment 2: More description about the RF model needs to be included in the manuscript (SI ok). For instance, how were the meteorological predictors included given the long sampling time for the VOCs and strong diel profiles of many of the meteorological variables? Further details about the training dataset would also be useful. For instance, is it representative across season, meteorological variables, weekday vs weekend, etc. or are some time periods overrepresented in the training vs test dataset? This is important given the relatively small number of time points.

Response (Lines 141, 145-146): Thanks for the editor's suggestion. In this study, the RF model relates the hourly variability of O_3 and $PM_{2.5}$ to that of meteorological variables. The sentence "The modelling relates the hourly variability of O_3 and $PM_{2.5}$ to that of meteorological variables." has been added in the revised version.

For the 10-fold CV approach, the training dataset in each round includes ~90% randomly selected data representing different seasons. In the revised version, the sentence "In each round, the training dataset includes ~90% randomly selected data representing different seasons" has been added.

Comment 3: Thank you for including the spreadsheet with the measurements. This is an important improvement in data availability. I think the data availability and the impact of the manuscript would be improved with some modifications. In particular, it

may be worth reporting the measurements in a more standardized format (for instance ICARTT format). At a minimum, please consider the following suggestions

a. Please standardize the names of the chemical compounds throughout the manuscript, the spreadsheet, and Table S2. For instance, the manuscript lists chloromethane whereas in the spreadsheet it is listed as methyl chloride. Including a CAS number for each compound would be helpful.

Response: The names of the chemical compounds in the text, Table S1, and Table S2 have been unified. Besides, the CAS number for each compound have been added in Table S1 and Table S2.

b. Is there a difference between the files "Table S1 – VOCs mixing ratios" and "Table S1 – VOCs mixing ratios (ppbv)"? Please include the units within the spreadsheet and not just in the file names.

Response: There is no difference between the files "Table S1 – VOCs mixing ratios" and "Table S1 – VOCs mixing ratios (ppbv)". The units within the spreadsheet have been added in the file names.

c. The mixing ratios reported in the files should have the correct number of significant figures.

Response: The mixing ratios of the VOCs species in Table S1 have been adjusted to two significant digits after the decimal point.

d. Please consider including an indication of the uncertainty and detection limit for each VOC in the spreadsheet. Please also consider indicating when a species was below detection limit with a standardized value (e.g., as per the ICARTT standard) rather than

reporting as zero.

Response: The uncertainty and detection limit for each VOC have been added in the spreadsheet. Besides, the VOCs species with concentrations below the detection limit were marked as "BDL" in the spreadsheet.

e. Add mention of the spreadsheet to the data availability section.

Response (376-377): The mention of the spreadsheet has been added in the data availability section: "The daily mixing ratio of individual VOCs species is given in Table S1 in the Supplement".

Technical

Comment 1: Line 101: "suitability" is probably a better word choice than "availability"

Response (Line 95): Thanks for the editor's suggestion. The word "availability" has been replaced by "suitability" in the revised version.

Comment 2: Lines 102-104: The description of the temperatures is confusing to me. Both -40 °C and 90 °C are listed as initial temperatures. I assume they are referring to different points, but it is not clear. Additionally, is the temperature ramped to 220 °C? If so, please include the rate.

Response (97-99): We are sorry for the wrong expression in the manuscript. The description of the oven temperature has been rewritten in the revised version: "The oven temperature was programmed at 40 °C for 3 minutes initially, then raised to 90 °C at 8°C per minute, and later raised 220 °C at 6°C per minute, holding for 9 minutes."

Comment 3: Lines 116: Acrolein is listed in the SI and excel spreadsheet, but

ethylacrolein is not. Should this be a reference to acrolein?

Response (Line 111): It's our overlook. The word "ethylacrolein" has been corrected into "acrolein" in the revised version.

Comment 4: Line 122: A word is missing after "meteorological." Variables could be an acceptable choice.

Response (Line 119): The word "variables" has been added after "meteorological".

Comment 5: Line 157: How was SO_2 measured? SO_2 measurements were not mentioned in Sect. 2.1.

Response (Lines 114 and 119): SO_2 was measured by a pulsed UV fluorescence (Thermo 43i, USA) with the detection limit of 0.5 ppbv. The description of SO_2 measurement has been added in the revised version.

Comment 6: Line 195/Figure 2: Please add a short sentence on the overlap of VOCs measured between this study and the ones shown in Figure 2 so that the reader can quickly judge that this trend is real and not an artifact of measuring different subsets of VOCs.

Response (Line 183): Thanks for the editor's suggestion. The overlap of VOCs groups measured between this study and the ones shown in Figure 2 has been described in the revised version as below:

"As shown in Fig. 2, the concentrations of TVOCs and major VOC groups including alkanes, alkenes, aromatics, halocarbons and OVOCs observed in this study were apparently lower than those during the sampling months in 2014 and 2016 in urban Beijing (An et al., 2012; Liu et al., 2020a; Li et al., 2015b), indicating the effectiveness

of control measures in most recent years on lowering VOCs emission.".

Comment 7: Lines 201-202: The dates listed for the ozone pollution days cover more than 14 days. Please clarify.

Response (Lines 190-191): We are sorry for the mistake. The date of 14 O_3 pollution days have been corrected in the revised version.

Comment 8: Line 207: Five, not four" ozone polluted months are listed earlier (line 203). Please clarify.

Response (Line 194): We are sorry for the mistake. The error has been corrected in the revised version.

Comment 9: Line 228: Typo. Should be "April"

Response (Line 216): We are sorry for the mistake. The error has been corrected in the revised version.

Comment 10: Line 235: I am not convinced that a correlation between RH and TVOCs really suggests that the "secondary transformation of VOCs was more conducive at higher RH." There could be co-varying factors that are not explored here. I suggest to either support this with more analysis or remove it.

Response (219-220): Thanks for the reviewer's suggestion. The sentence "Both the value of relative humidity (RH) and TVOCs increased significantly on $PM_{2.5}$ pollution days, suggesting that the secondary transformation of VOCs was more conducive at higher RH." has been deleted in the revised version.

Comment 11: Line 273: Please consider reversing the ordering of compliance and polluted days here since the reverse ordering was used earlier. It would make it easier

to read.

Response (Lines 256-260): The ordering of compliance and polluted days here has been reversed in the revised version.

Comment 12: Lines 298-300: I think this sentence is oversimplifying a complex situation. VOC mixing ratios can also exhibit "great change" due to meteorological factors (e.g., shallow boundary layers, low transport, etc.). I suggest focusing this first sentence on ratios rather than on absolute abundances.

Response (Lines 280-282): Thanks for the editor's suggestion. The sentence has been corrected into "The great changes in mixing ratios of different species are mainly affected by the photochemical processing and the emission inputs, and the ratios of VOCs species having similar atmospheric lifetimes can reflect the source features".

Comment 13: Line 403: I suggest changing the wording to "…from December 2018 to November 2019…"

Response (Lines 353-354): Thanks for the editor's suggestion. The revision has been made.

Comment 14: Figures: Please add letters to identify the subpanels to all figures and include this naming in the manuscript and figure captions.

Response: Letters for identifying the subpanels to all figures, and the naming in the manuscript and figure captions have been added in the revised version.

Comment 15: Figure 2: It would be easier for the reader to understand this figure if horizontal dashed lines (or some other separator) were included to separate out the different months (e.g., b/w Oct 2019 and July 2016, etc.).

Response: In the revised version, horizontal dashed lines have been in Figure 2 to separate out the different months.

Comment 16: Figure 3: Please clarify the caption with regards to which days are represented in the figures. For instance, do the ozone compliance and ozone pollution days only include the ozone polluted months or is all the data included? Please consider using the same colors for the VOCs as is used in other figures. For instance, alkenes are green here but pink in figure 5.

Response (Lines 660-667): The caption with regards to which days are presented in Figure 3 has been added in the revised version. Besides, the colors for the VOCs in Figure 3 and Figure 5 have been unified.

Comment 17: Figure 4: Why does the y-axis on the top panel go to -0.2?

Response: According to our results, the meteorological conditions are generally favorable for O_3 decrease during the non-O_3-polluted months, and the mean meterologically-driven O_3 concentration during the non-O_3-pollted months was -0.19 $\mu g\ m^{-3}$.

Comment 18: Figure 9: Please consider structuring Figures 5 and 9 with the same format rather than reversing the ordering.

Response: Figure 5 and 9 have been structured with the same format in the revised version.