

Responses to Referee 2's new comments

General comment

The quality of the paper has improved significantly. However there are still a few issues which the authors want to address before publication in ACP. Here I only repeat my initial questions and include additional comments ("New"):

- 5 We highly appreciate the reviewer for reviewing our responses and the revised manuscript again. All comments have been addressed with proper revision in the manuscript. In the following section, the author's responses (in blue) are immediately after the reviewer's comments (in black for original and in green for new), with the changes in manuscript at the end (*in italic*).

10 Major issues:

Comment 2.1

- In most figures intra- and inter-annual variations are significantly larger than the 2005-2014 trend. For instance, the O₃ trend shows the highest increase from 2005-2014 (0.67 ppb/yr) in autumn (Fig. 3). However, this trend is only determined by 3 "outlier" months of the years 2012, 2013, 2014. These 3 months are just 10% of this specific data set. Another example is*
- 15 *Fig. 5 which shows very large scatter in O₃ data for the autumn season. Also, for instance Fig. S10 about the annual trends of VOC/NO_x ratios is largely determined by the last two years. The question is: how robust are all the trends shown in this paper?*

New:

- I want to state that the term "outliers" I used in my initial comment does not refer to any characteristics of this data, which*
- 20 *could be regarded as "bad" data, and I understand that the data shown in box-whisker plots consists of a larger data set. This is why I put the term "outlier" in brackets. My statement focused on the fact that these few years at the end of the reporting period 2005-2014 completely determine the statistical trend and one should be cautious to deduce a trend and this should be mentioned in the text. Looking into the VOCs/NO_x ratio time series (previously S10, now S17), for instance, it seems that there has been some significant variability. For instance the years 2009, 2011, and 2012 show significantly*

enhanced VOCs/NO_x ratio. Assuming the year 2015, just following the 2005-2014 time series, would show any values comparable to those seen in 2009, 2011 and 2012, the trend analysis would yield a completely different results. The similar comment would refer to O₃ time series (Figure 2). The authors should make the point why they believe that such variability could be excluded for future years and that their trend analysis is justified in this sense.

5 **Response:**

New:

Thanks for further clarifying the term “outlier” and giving the constructive suggestion on the issue. Indeed, the “outliers” in Figure 3 (i.e., the extremely high monthly O₃ mixing ratios observed in Octobers in 2012-2014) seemed to bias the O₃ trend in autumn. However, if we look at Figure S10 which showed the daily average O₃ values in autumn (i.e., more data points than in Figure 3), the values varied much more significantly in 2012-2014 than in previous years. The O₃ trend was determined by all the measured data points including both extremely high and low values in all the study years. We have made this point in the revised manuscript.

For VOCs/NO_x ratios, we fully agree that the significant variability cannot be excluded for future in Hong Kong given the changing contributions of local emission and regional transport to these O₃ precursors. Since the decreasing rate of the ratios was very small, -0.02 yr⁻¹ (Figure S17), over-interpretation of the result should be avoided despite the result is statistically significant ($p < 0.05$). To give the better image of the VOCs/NO_x ratios, we changed the plot from bar chart to scatter points. This point has also been added in the revised manuscript.

Revision in the manuscript:

20 *Page 11, line 16:*

“It is noteworthy that there were three extremely high O₃ data points in Octobers in 2012-2014 (Figure 3), which seemed to bias the O₃ trend in autumn. However, if we looked at Figure S10 which showed the daily average O₃ values in autumn (i.e., more data points than in Figure 3), the values varied much more significantly in 2012-2014 than in previous years. It is worth emphasizing that the overall O₃ trend was determined by all the measured data points including both extremely high and low values in all the study years.”

Page 20, line 32:

“Furthermore, the monthly variation of TVOC/NO_x ratios showed a statistically significant decreasing trend at a rate of -0.02 yr⁻¹ (*p*<0.05) (see Figure S17). The weak declining trend moderately supports that VOC reduction became more effective in reducing O₃ in the past 10 years, which is consistent with the conclusions from the above modelling results.”

Supplementary Figure S17:

To facilitate the discussion above, the format of Figure S17 has been changed from a bar chart to a time-series plot with dots.



Figure S17. Monthly trend of TVOCs/NO_x ratio at TC in 2005-2014.

Minor issues:

Comment 2.2

Page 4 L13-14:

How was the accuracy of 1-7% determined?

New:

- 5 *I assume that a "measured mixing ratio" would ultimately depend on the weekly span checks and calibration. What makes a "measured mixing ratio" different from an "actual mixing ratio"?*

Response:

New:

- The weekly span checks and calibration are part of the routine QA/QC to improve the accuracy of VOC measurements.
- 10 However, many other known or unknown sources, including at least but not limited to the preparation and storage of the standard gases, the variation of the instrumental performance, the operators and so on, making it impossible to eliminate the difference between "measured mixing ratio" and an "actual mixing ratio".
- As the description of measurement techniques and related QA/QC has been greatly enhanced in previous revision, no more revision has been made for this comment.

Comment 2.3

Page 5 L25-29:

The authors only measured 21 VOCs. What assumptions did the authors have on other VOCs not measured, but needed as an input for MCM?

New:

- 20 *The authors should include their comment specifically with respect to MCM in their manuscript.*

Response:

Thanks for the comment. We have included our comment specifically with respect to MCM in this version as follows.

Revision in the manuscript:

Page 6, Line 3:

“It was assumed that the measured VOCs contributed a dominant fraction to O₃ production, and that the initial concentrations of those VOCs not measured but needed by MCM were zero. We admit that the use of a limited number of VOCs would cause photochemical reactivity missing.”

Comment 2.4

5 *Page 7, L16:*

With regard to the precursors NO_x, total VOCs and CO did the authors calculate arithmetic means or medians? Would there be differences?

New:

10 *I primarily referred to O₃ precursors, not O₃ itself. I doubt that the number of data determines, whether data ensembles are distributed normally or not. Rather it is an intrinsic characteristic of the specific data ensemble. The authors should mention in the manuscript that they used arithmetic means in order to compare with other studies. Unfortunately, it does not make the use of arithmetic means more correct, the more studies have used this quantity.*

Response:

New:

15 *Thanks for the clarification. The authors fully understand the reviewer’s concern. The suggested information has been added in the text.*

Revision in the manuscript:

Page 8, Line 12:

“Please note that arithmetic means were used here in order to compare with other studies.”

20 **Comment 2.5**

Page 12 L11:

I was just wondering if the definition of daytime (0700-1900 LT) is valid regardless what season is concerned.

New:

The authors should include their answer in the manuscript.

Response:

The answers have been included in the revised manuscript.

Revision in the manuscript:

5 *Page 13, Line 17:*

“Although the actual duration of daytime in Hong Kong varies in 1-2 hours by seasons, the expected uncertainty from it would be limited if considering weak photochemical reactions in early morning and in late afternoon.”

Comment 2.6

Page 21, L17:

10 *“...increased emissions of alkenes from traffic related sources”. Is this due to enhanced alkene emissions from changes in the composition of the traffic fleet or from increased traffic volume? If it is the latter, then emissions of aromatic compounds would also increase.*

New:

The authors should include their answer in the manuscript.

15 **Response:**

The answers have been included in the revised manuscript.

Revision in the manuscript:

Page 22, Line 23:

20 *“In contrast, the contributions of AVOC (alkenes) to O₃ production in these years showed a significantly increasing trend with a rate of 0.14±0.01 ppbv/yr (p<0.05), perhaps attributed to the increased emissions of alkenes from changes in the composition of the traffic fleet and from increased traffic volume.”*

Comment 2.7

Page 21, L20-21:

Diesel driven vehicles emit significantly less VOCs than gasoline driven vehicles. In other words was the DCV program a significant contribution to the overall traffic related alkene emissions?

New:

The authors should include their answer in the manuscript.

5 **Response:**

The answers have been included in the revised manuscript.

Revision in the manuscript:

Page 22, Line 27:

10 *“Among the measures, gasoline and LPG vehicular emissions caused ambient alkenes to increase during the same period due to the increasing number of LPG/gasoline vehicles and some short-term/non-mandatory measures (Lyu et al., 2017). The diesel commercial vehicle (DCV) program (2007-2013) was shown to be effective in reducing the emission of alkenes from diesel vehicles (Lyu et al., 2017), however these vehicles emit significantly less VOCs than gasoline driven vehicles. In consequence, the overall emissions of alkenes from traffic related sources increased during 2005-2014, leading to the increased contribution of AVOC (alkenes) to O₃ formation (Lyu et al., 2017).”*

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Comment 2.8

Page 21, L26-28:

Why would the AVOC (alkanes) contribution to O₃ formation not increase with increasing alkane levels in 2005-2013?

New:

20 *The authors should include their answer in the manuscript.*

Response:

The answers have been included in the revised manuscript.

Revision in the manuscript:

Page 23, Line 1:

“Unlike AVOC (Aromatics/Alkenes), the contribution of AVOC (Alkanes) to O₃ formation during 2005-2014 showed no significant change ($p=0.23$) despite the increase of total alkane levels in the atmosphere in 2005-2013 (Ou et al., 2015). This is because alkanes include a bunch of compounds which have different but generally low reactivity with OH radicals. Hence, although the level of total alkanes increased over the years, it did not warrant the increase in its contribution to O₃ formation. For example, one possible case is that some alkanes with relatively high reactivity decreased with an increase of some low-reactivity alkanes.”

Comment 2.9

Page 22, L6-7:

The authors state that 90% of isoprene was emitted from biogenic sources, while traffic sources were less than 5%. From what sources did the remaining 5% isoprene come from?

New:

The authors should include their answer in the manuscript.

Response:

The answers have been included in the revised manuscript.

Revision in the manuscript:

Page 23, Line 15:

“The source of isoprene at TC site has been also investigated and confirmed by previous long-term source apportionment studies, which reported that during 2005-2013 about 90% of isoprene was emitted from biogenic emissions, with minor contribution from traffic emission, consumer products and printing processes (Ou et al., 2015).”