Title: Effectiveness of replacing catalytic converters in LPG-fueled vehicles in Hong Kong

Author(s): X. P. Lyu et al. MS No.: acp-2015-790

Responses to the comments from Anonymous Referee #2

General comments:

This paper provides information on the impact of the replacement of catalytic converters in vehicles fuelled by LPG in Hong Kong. The policy was based on the need to maintain the effectiveness of the catalysts; it was extensive and conducted over a period of several months over 2013 – 2014. The paper reports roadside and background measurements of VOCs, NOx, O3 and CO and a set of analyses to investigate the effectiveness of the programme. The measurements and analysis are of considerable interest and of relevance to the development of effective urban air quality policies. The reduction in LPG-related VOCs was substantial over only a 9 month period. The paper, however, has a number of deficiencies related either to the clarity of the presentation or to the methodologies used in the analysis and interpretation.

We thank the reviewer very much for his/her valuable comments and suggestions, which helped improve the manuscript substantially. Apart from the technical/typographical comments, the referee's concerns mainly focused on three aspects: (i) the background of this study - more information about the program, the physico-chemical processes and previous O_3 modeling in Hong Kong. (ii) the methods, including the method to obtain the primary emissions, the photochemical simulation with the backgrounds being subtracted, and the subtraction method to obtain O_3 production by LPG and the atmospheric reactivity (i.e. OH, HO_2 and their formation/loss pathways). (iii) the results, mainly the source apportionment results and the simulation of OH, HO_2 and pathways of HO_x. To answer these questions, we have supplemented and revised the manuscript accordingly. The method removing the backgrounds and the subtraction method were abandoned in the source apportionment and in the simulation of O_3, OH, HO_2 and the pathways of OH and HO_2, which resulted in reasonable source apportionment results and the reactivity simulations. Our responses to the reviewer are as follows, along with indications of how the manuscript has been further revised for consideration by ACP. We hope that these changes will further strengthen the main points and make them clearer in the revised manuscript.

Specific comments:

1. More information should be given to aid the understanding of the general situation in Hong Kong and the basis of this intervention programme. (a) A brief review of the regional physico-chemical processes and atmospheric transport and their extent in influencing air quality in Hong Kong. There have been several such studies and a brief discussion would be helpful. (b) A tabulation of typical VOC concentrations at the roadside and background sites – at present it is difficult to assess the importance of the LPG-related VOCs – a number of values are given for the fractional contribution to the total atmospheric VOC burden, but real data on all significant VOCs measured, perhaps with the pseudo first order rate constant for OH removal (i.e. kOH + VOC [VOC]), would be of value.

Many thanks for the good suggestion. A brief review of the regional physico-chemical processes and atmospheric transport and their extent in influencing air quality in Hong Kong was added in the revised manuscript:

Apart from local emissions, air quality in Hong Kong is also affected by regional transport. In autumn and winter, prevailing northerly and northeasterly winds bring anthropogenic emissions from mainland China to Hong Kong, enhancing local pollution burden. In summer, clean oceanic air masses dilute the levels of air pollutants under the influence of southerly winds (Ling et al., 2013; Guo et al., 2009, Wang et al., 2005; So and Wang, 2003; Chan and Chan, 2000). Ling et al. (2013) quantified the contribution of regional transport as 0-61% for summer O_3 and 0-56% for autumn O_3 on high O_3 days in Hong Kong. Guo et al. (2006) found that over 45% of VOC samples collected from August 2001 to December 2002 at Tai O, Hong Kong, were influenced by regional transport from inland China. Based on numerical simulations, Wang et al. (2006) quantified regional contributions of 40-90% to O_3 formation in Hong Kong, and Huang et al. (2006) indicated that the regional contribution was <50% when a trough was over the South China Sea (SCS), and it increased to >50% in the presence of an anticyclone over mainland China and/or a tropical cyclone over SCS. Furthermore, Wang et al. (2006) revealed that the regional contribution was lower during daytime than that at night, while Lam et al. (2006) found that the contribution decreased from >50% during an O_3 episode to 30-50% after the episode. In addition, Jiang et al. (2008) reported that the contribution of horizontal transport to surface O_3 in Hong Kong was negligible during a typhoon-induced O_3 episode. However, all the studies focused on the regional contribution at non-roadside sites, and the O_3 chemistry at roadside sites in Hong Kong was seldom reported. Given high NO_x levels, tall buildings and low wind speeds along the main roads in Hong Kong, it is expected that the regional impact on air pollutants in roadside environment is relatively minor, especially during daytime hours on non-O_3 episode days.

For details, please refer to lines 71-92, pages 3-4 in the revised manuscript.

The average VOC concentrations and their OH-reactivity at the roadside and background sites were also provided:

Table S2 summarizes average mixing ratios of the main VOCs and trace gases, and their OH-reactivity at the roadside (MK) and background site (HT) before (October 2012-September 2013) and during (October 2013-May 2014) the program in Hong Kong. The much higher OH-reactivity at MK ("before": 86.6±6.0 s^{-1}; "during": 77.9±6.1 s^{-1}) than at HT ("before": 5.0±0.3 s^{-1}; "during": 4.9±0.1 s^{-1}) meant that the OH lifetime was much shorter at MK (~0.01 s) than at HT (0.2 s), and even shorter than that at an urban site in New York (0.05 s) (Ren et al., 2003), indicating that reactive species were extremely abundant in the roadside environment of Hong Kong. The LPG-related VOCs (*i.e.*, propane and *n*/*i*-butanes) contributed 56.5±2.7 % and 46.0±3.1% to total VOCs, and 18.7±1.2% and 15.1±1.1% to the total OH-reactivity of VOCs at MK, before and during the program, respectively. These fractional contributions suggest the importance of LPG-related VOCs in the budgets of VOCs and O_3 in Hong Kong. Consistent with the decreases of fractional contributions once the program started, the mixing ratios (29.4±3.3 and 21.0±2.2 ppbv for before and during the program, respectively) and OH-reactivity (1.12±0.04 and 0.79±0.02 s^{-1} before and during the program, respectively) of LPG-related VOCs significantly declined at MK ($p<0.05$), while at HT their mixing ratios increased from 1.5±0.1 ppbv to 1.9±0.04 ppbv, and the OH-reactivity increased from 0.06±0.004 s^{-1} to 0.07±0.003 s^{-1}. The minor variation of background LPG-related VOCs relative to the decreases at MK (4.8% in mixing ratios and 3.0% in OH-reactivity of VOCs) indicated that the influences of the background concentrations on the variations of LPG-related VOCs and O_3 at MK were minimal and can be neglected.

For details, please refer to lines 169-188, page 7 in the revised manuscript and Table S2 in the supplementary material.

2. Regional processes are excluded from the analysis on the basis of the propane /CO ratio at the main roadside site. The atmospheric lifetimes of the LPG VOCs are considerable (For [OH] 10^6 cm^{-3} and rate constants of 1-3 x 10^{-12} molecule cm^{-3} s^{-1} the lifetime is 100 h) Even for a windspeed of 2 m s^{-1}, ozone will be generated from such gases over 1000 km. So while the roadside concentrations of the VOCs are arguably dominated by local sources, chemistry and ozone production are necessarily regional, although titration and the establishment of the NO_x/O_3 stationary state are local. Since much of the emphasis of the paper is on ozone formation, some discussion of the area over which the new controls operate, and reference to previous Eulerian / Lagrangian analyses of ozone formation in the Hong Kong region, would be of value. Note that on p35949, line 27, the authors refer to the O_3 calculation as being “purely local”.

Thanks very much for the excellent comment and suggestion. We agree that the regional transport of O_3 can occur, even at the very low wind speed (actual average=1.3 m/s at MK), because O_3 can be formed in the source region and then transport to Hong Kong, and/or during the transport of O_3 precursors. As suggested, the regional impacts on O_3 levels in Hong Kong were discussed based on previous numerical simulation study. In addition, we also provided more information about the consideration of regional transport influence in the present PBM-MCM model used in this study. Since the reviewer considered that the subtraction method used in previous version was inappropriate and led to strange results, we checked both “with” and “without” input of background concentrations of air pollutants, and found that the impact of the background O_3 precursors on the O_3 simulation at the roadside site was negligible. Hence, in the revised manuscript, we used the whole-air ambient concentrations of VOCs and trace gases at roadside site to simulate O_3 formation, rather than the subtraction method. All the revisions were shown as follows. The inappropriate wording such as “purely local” was deleted in the revised manuscript.

Based on numerical simulations, Wang et al. (2006) quantified regional contributions of 40-90% to O_3 formation in Hong Kong, and Huang et al. (2006) indicated that the regional contribution was <50% when a trough was over the South China Sea (SCS), and it increased to >50% in the presence of an anticyclone over mainland China and/or a tropical cyclone over SCS. Furthermore, Wang et al. (2006) revealed that the regional contribution was lower during daytime than that at night, while Lam et al. (2006) found that the contribution decreased from >50% during an O_3 episode to 30-50% after the episode. In addition, Jiang et al. (2008) reported that the contribution of horizontal transport to surface O_3 in Hong Kong was negligible during a typhoon-induced O_3 episode. However, all the studies focused on the regional contribution at non-roadside sites, and the O_3 chemistry at roadside sites in Hong Kong was seldom reported. Given high NO_x levels, tall buildings and low wind speeds along the main roads in Hong Kong, it is expected that the regional impact on air pollutants in roadside environment is relatively minor, especially during daytime hours on non-O_3 episode days.

Please refer to lines 79-92, pages 3-4 in the revised manuscript for details.

A base case was established to simulate O_3 and the photochemical reactivity (*i.e.*, the formation and loss pathways of OH and HO_2 listed in Table 2). The observed VOCs and trace gases were input to construct the base case, and the simulated O_3 was compared with the observed levels to validate the model and check the influence of regional transport during daytime hours. At low wind speed (1.3±0.01 m/s) and high NO_x (223.2±1.8 ppbv) at MK, the majority of regional O_3 would be completely consumed before being detected by measurement instruments. In fact, the PBM-MCM model considered the regionally-transported concentrations of O_3 precursors, as the observed concentrations input into the model included the regional fractions. Moreover, since O_3 levels were higher at night (8.1±0.2 ppbv) than that during daytime (5.9±0.1 ppbv) at MK, it suggests the existence of nocturnal regional transport. Hence, the observed O_3 at 07:00 was input into the model to initiate the simulation for daytime hours, *i.e.*, the nocturnal regional transport of O_3 was also considered.

For details, please refer to lines 303-314, pages 12 and 13 and Table 2 in the revised manuscript.

3. The arguments deriving from the PMF analysis, that NOx concentrations can be ascribed to emissions from gasoline vehicles is unconvincing. Diesel Euro5 NOx emissions are still substantial and the controls on gasoline vehicles would have to be very weak if they were to dominate. This problem emphasises the need for more data –we are not told about the magnitude

of the total VOC concentrations in the four factor areas, nor about the numbers and types of vehicles in the gasoline, LPG and diesel categories.

Thanks for the useful comments and suggestions. The source apportionment is a critical part of this study to evaluate the effectiveness of the program and its impact on O_3 production. We fully agreed with the reviewer's comments, and made some substantial revisions in this part. Firstly, considering this comment and other comments (points 4-6 below) of the reviewer, the full (or whole-air) ambient concentrations of VOCs and trace gases rather than those with the backgrounds being deducted were used, in order to keep consistency with the simulations of whole-air ambient O_3 and photochemical reactivity. Secondly, because of the change of input data for the PMF apportionment simulation, the sources of O_3 precursors were re-identified and re-quantified. Four sources including "Gasoline/diesel vehicle exhaust", "LPG-fueled vehicle exhaust", "Aged air masses", and "Solvent usage" were identified. The majority of NO_x was assigned to the "Gasoline/diesel vehicle exhaust". Thirdly, the actual influence of the background deduction on the reduction of LPG-related VOCs and NO_x was evaluated to ensure the appropriateness of the use of whole-air ambient concentrations as model input, which would not interfere with the assessment of the replacement program. Lastly, the magnitude of the total VOC concentrations in the four sources, and the number and type of vehicles in the gasoline, LPG and diesel categories were provided.

For details, please refer to lines 483-549, pages 21-25 (section 3.2) in the revised manuscript and Table S4 in the supplementary material.

4. The method of establishing VOC concentration from primary processes, discussed in section 2.3.1-3, which are then used extensively in the rest of the paper, is questionable. The background concentrations are subtracted to determine the base case, and the LPG-related concentrations, presumably based on the PMF analysis, are subtracted to form a set of scenarios. We need to be given information on what these sets of concentrations are.

Thanks for the comments and suggestions. In the revised manuscript, the whole-air ambient concentrations (not the subtracted ones) were used for the source apportionment and the simulation of O_3 and photochemical reactivity in the base case. To evaluate the possible impact with and without the background subtraction on the assessment of the interventional program, we have conducted sensitivity analysis for both PMF and PBM-MCM results. For the source apportionment section, the following revision was made:

During the study period, the LPG-related VOCs and NO increased by ~1.4 and ~2.9 μg/m^3, respectively, at the background site, only respectively accounting for 3.0% and 1.3% of the decreased LPG-related VOCs (~46.1 μg/m^3) and NO (~226.8 μg/m^3) in LPG-fueled vehicle exhaust. Therefore, it is believed that the reductions of VOCs and NO in LPG-fueled vehicle exhaust benefited from the intervention program.

For details, please refer to lines 537-542, page 23 in the revised manuscript.

For the PBM-MCM results with and without the background subtraction, the following revision was made.

The simulation results for the scenarios with and without the input of background concentrations indicated minor contribution of background concentrations to O_3 for both periods of "before" (0.24 ppbv, accounting for ~3.5% of measured average) and "during" (0.27 ppbv; ~3.7%) the program, suggesting the use of whole-air ambient concentrations without background subtraction for O_3 simulation was appropriate. The slight increase of O_3 (~0.03 ppbv) caused by the background variations of O_3 precursors from before to during the program constituted only ~7.5% of the O_3 enhancement (0.40 ± 0.03 ppbv) due to the replacement program, further confirming a negligible impact of the background on the assessment of the program.

For details, please refer to lines 575-583, page 26 in the revised manuscript.

In addition, the simulation method for the estimation of the contribution of LPG source to O_3 formation was revised to avoid possible confusion. At last, the configurations of the model input for the base case and the three constrained cases were provided in Table S3 in the supplementary material.

To simulate the contribution of LPG source to O_3 formation, the following approach was adopted to avoid altering the real ambient environment for atmospheric chemistry. Briefly, the concentration of species X in the whole air and in the assigned LPG source was defined as $X_{whole\text{-}air}$ and X_{LPG}, respectively. Hence, the concentration of "$X_{whole\text{-}air}$ - X_{LPG}" was the input for the constrained cases while $X_{\text{whole-air}}$ was the input for the base case. The three constrained cases were i) only VOCs in the LPG source were excluded from the whole-air; ii) only NO_x in the LPG source was excluded; and iii) both VOCs and NO_x were removed simultaneously. As such, the differences of the outputs between the base case and the three constrained cases were the contributions of i) VOCs, ii) NO_x and iii) VOCs + NO_x in the LPG source to the O_3 formation, respectively. The changes of these contributions from before to during the program represented the impact of the intervention program on O_3 formation. Table S3 in the Supplementary material lists the configurations of the model input for the base case and the three constrained cases.

For details, please refer to lines 317-328, page 13 in the revised manuscript and Table S3 in the supplementary material.

5. These concentration sets are then used in box models of the local processes, using the detailed chemistry from the MCM. Is this approach justifiable? The chemistry occurring is unable to distinguish between the sources of the species present in the atmosphere. LPG VOCs are affected by the background concentrations, and those derived from other sources. Subtracting out species concentrations is highly questionable.

Sorry for the confusion. As replied in questions 3&4 above, in the revised version, the full (or whole-air) ambient concentrations of VOCs and trace gases rather than those with the background concentrations being deducted were used, in order to keep consistency with the simulations of whole-air ambient O_3 and photochemical reactivity, and to avoid altering the real ambient environment for atmospheric chemistry. The concentration of "$X_{\text{whole-air}}$ - X_{LPG}" was the input for the constrained cases while $X_{\text{whole-air}}$ was the input for the base case. Through this approach, the interference of other sources on the evaluation of the effectiveness of the interventional program was eliminated. This method also guaranteed that the background concentrations and those derived from other sources still existed in the base case and the constrained cases, which would not alter the chemistry of the atmosphere. Indeed, the influence of the background concentrations to the O_3 production was minor (~3.5%-~3.7%), as estimated in question 4.

The corresponding revisions were made in the "Methodology" as follows:

A base case was established to simulate O_3 and the photochemical reactivity (*i.e.*, the formation and loss pathways of OH and HO_2 listed in Table 2). The observed VOCs and trace gases were input to construct the base case, and the simulated O_3 was compared with the observed levels to validate the model and check the influence of regional transport during daytime hours.

To simulate the contribution of LPG source to O_3 formation, the following approach was adopted to avoid altering the real ambient environment for atmospheric chemistry. Briefly, the concentration of species X in the whole air and in the assigned LPG source was defined as $X_{whole-air}$ and X_{LPG}, respectively. Hence, the concentration of "$X_{whole-air}$ - X_{LPG}" was the input for the constrained cases while $X_{whole-air}$ was the input for the base case. The three constrained cases were i) only VOCs in the LPG source were excluded from the whole-air; ii) only NO_x in the LPG source was excluded; and iii) both VOCs and NO_x were removed simultaneously. As such, the differences of the outputs between the base case and the three constrained cases were the contributions of i) VOCs, ii) NO_x and iii) VOCs + NO_x in the LPG source to the O_3 formation, respectively. The changes of these contributions from before to during the program represented the impact of the intervention program on O_3 formation. Table S3 in the Supplementary material lists the configurations of the model input for the base case and the three constrained cases.

For details, please refer to lines 303-306, page 12, lines 317-328, page 13 and Table 2 in the revised manuscript, and Table S3 in the supplementary material.

Corresponding revisions were also made in the "Results and discussion".

Please refer to lines 550-592, pages 25-27 (section 3.3) in the revised manuscript for details.

6. I presume that this approach is responsible for the rather strange results of the HOx calculations that are given: a. The calculated [OH] and especially [HO2] are very low. A rationale is given, but it is highly qualititative. b. What is meant by "net O3 photolysis", which is found to consume OH. I presume this arises from the subtraction method used and clearly demonstrates the problems such an approach generates. The production and destruction rates are, I presume, calculated from the ambient concentrations multiplied by the rate constant or J value. O3 photolysis as a sink is nonsensical.

Thank you very much for the excellent comments and suggestions.

(a) Indeed, the strange results of OH, HO_2 and the formation/loss pathways of HO_x were caused by the subtraction of the background concentrations. In the revised manuscript, the whole-air ambient concentrations were used to simulate the OH and HO_2, and reasonable results were obtained.

For details, please refer to lines 594-634, pages 27-29 (section 3.4.1) in the revised manuscript and Figure S3 in the supplementary material.

(b) Also, the subtraction method generated wrong reaction pathways of HO_x. With the use of whole-air ambient concentrations for the PBM-MCM simulations, the problems were solved.

For details, please refer to lines 621-634, pages 28-29 in the revised manuscript.

7. The HOx calculations are, in principle, of value in assessing the influence of the replacement strategy on the photochemistry but the approach must be revised. The calculations must be conducted with the full ambient concentrations, rather than using the subtraction approach. One way to examine the effects of the replacement programme would then be to compare total rates and concentrations (of ozone, HOx etc) before and during the programme. Of course ambient conditions can change, presenting comparison problems. The best way would be to conduct a proper sensitivity analysis, examining the sensitivity of specific objectives, e.g. the ozone formation rate or [OH] on the concentration of LPG VOCs and NOx, under conditions prior to and during the replacement process.

Excellent comments. As stated earlier**,** the subtraction method was not used anymore, *i.e.*, the background concentrations were not subtracted in the revised manuscript. Using the whole-air ambient concentrations, the sensitivity analysis of [OH] and [HO_2] on the concentration of LPG VOCs and NO_x, under conditions prior to and during the replacement process was conducted.

For details, please refer to lines 635-658, pages 29-31 (section 3.4.2) in the revised manuscript.

8. It would also be useful to see a more complete analysis of production and destruction, with separate analyses for OH and HO2. I presume that Figure 11 refers to the overall HOx radical formation and removal rates and this is why OH + VOC reactions do not figure in Figure 11. It

explains why overall production and destruction rates do not balance, the differences relating to other radical formation / destruction. This partial analysis leaves more questions than it answers. Either this table should relate to total radical formation and destruction – a substantial problem in a region with a complex VOC species range. This would allow assessment of the total initiation and termination rates for the reaction system. Or it should concentrate on OH and HO2, as at present, but reporting their total production and removal rates, including the propagation reactions (e.g. reaction with VOCs). Such an analysis is straightforward with the MCM. It would provide a much more useful analysis of the impact of the LPG reductions than does the present analysis, whose objective is obscure.

The comments and suggestions are appreciated. We fully agreed with the reviewer, and accepted the valuable suggestions. In the revised manuscript, the formation and loss pathways of OH and HO_2 were calculated separately, and they were balanced for both OH and HO_2.

The corresponding revisions in methodology are as follows:

A base case was established to simulate O_3 and the photochemical reactivity (*i.e.*, the formation and loss pathways of OH and HO_2 listed in Table 2).

For details, please refer to lines 303-304, page 12, and Table 2 in the revised manuscript.

For the revised results and discussion on the complete analysis of production and destruction, with separate analyses for OH and HO_2, please refer to lines 594-658, pages 27-31 in the revised manuscript.

Technical / typographical comments

35942; 16: change since to over.

Accepted with thanks. Please refer to line 104, page 4 in the revised manuscript.

35942; 21: initiate rather than initialize. Also elsewhere in the paper.

Accepted with thanks. Please refer to line 109, page 4 in the revised manuscript.

35942; 23. Reword – the LPG vehicles weren't replaced with catalytic converters – the converters on them were replaced.

Replaced by "….and the catalytic converters in ~75% of LPG-fueled vehicles were renewed….."

Please refer to lines 110-111, page 4 in the revised manuscript.

35943, 8. Insert the before South.

Accepted with thanks. Please refer to line 123, page 5 in the revised manuscript.

35943, 12. Insert A before marine

Accepted with thanks. Please refer to line 127, page 5 in the revised manuscript.

35945, 20 et seq. There is confusion of emissions / formation and concentrations. The wording should be changed. Concentrations indicate emissions or formation, but aren't synonymous– they have different units.

Thanks for the comment. We revised the expression as follows:

The measured concentrations of VOCs and trace gases were composed of the backgrounds, primary emissions and secondary formation (applicable to O_3 and NO_2). To obtain the concentrations of LPG-related VOCs and NO_x emitted from primary sources at the roadside sites, the background concentrations and the concentrations elevated by the secondary formation (if applicable) in this study were excluded using the equations (1) - (5) (Takekawa et al., 2013):

$$[VOC]_{prim.} = [VOC]_{obs.} - [VOC]_{bg.} \quad \text{(Equation (1))}$$

$$[NO_2]_{sec.} = [O_3]_{bg.} - [O_3]_{obs.} \quad \text{(Equation (2))}$$

$$[NO_2]_{prim.} = [NO_2]_{obs.} - [NO_2]_{sec.} - [NO_2]_{bg.} \quad \text{(Equation (3))}$$

$$[NO_x]_{prim.} = [NO_x]_{obs.} - [NO_x]_{bg.} \quad \text{(Equation (4))}$$

$$[NO]_{prim.} = [NO_x]_{prim.} - [NO_2]_{prim.} \quad \text{(Equation (5))}$$

where $[xx]_{obs.}$, $[xx]_{prim.}$, $[xx]_{sec.}$ and $[xx]_{bg.}$ represent the observed concentrations, the concentrations emitted from primary sources, secondary formation and the backgrounds, respectively.

For details, please refer to lines 212-224, pages 8-9 in the revised manuscript.

35946, 18: omit the before fresh

Accepted with thanks. Please refer to line 234, page 9 in the revised manuscript.

35949, 14: omit in the (or omit the whole phrase and change to the concentrations of some species in the free troposphere

Accepted with thanks. Please refer to line 300, page 12 in the revised manuscript.

35950, equation (9). The dash before free troposphere looks like a minus sign – I couldn't understand what was going on. Make identical to that on line 7 or change to RIR (weighted concentrations).

Accepted with thanks. It was revised as "RIR (weighted concentration)". For details, please refer to line 334, page 14 and Equation (9) in the revised manuscript.

35956, 22. The current use of "well" is clumsy and rather colloquial. ? Change well to accurately.

Thanks for the suggestion. Since this part was revised substantially, the word "well" was deleted in the revised manuscript. For details, please refer to lines 551-566, pages 24-25 in the revised manuscript.

35956, 25. What is meant by 'background O3 : : :..reaction'?

Thanks for the question. In the original manuscript, O_3 simulation was based on the concentrations with the backgrounds being subtracted. Therefore, the difference between the simulated and observed O_3 was partially due to the backgrounds, which was defined as background O_3. In the revised manuscript, O_3 was simulated with the full ambient concentrations, and the expression of "background O_3" was deleted. For details, please refer to lines 551-566, pages 24-25 in the revised manuscript.

35959, 25 change to 'constitutes the major source of..'

Thanks for the comment. Since this part was revised substantially, the original description was changed. For details, please refer to lines 621-632, pages 28-29 in the revised manuscript.

Figure 4. Explain the variability during Jun-Aug 2013 – the variations exceed the subsequent downward trend. Define x.

Thanks for the comment. We noticed that the mixing ratios of VOCs were far from the regression line in June and July, 2013. The explanations were provided in the revised manuscript.

The abnormally low alkane levels in late June 2013 might be caused by the anti-cyclone over southern China and a tropical storm over the SCS, leading to extremely high temperature on June 18-20 (*i.e.*, favorable for photochemical reactions), and stronger winds (*i.e.*, conducive to atmospheric dispersion) on the following days, respectively. In contrast, Hong Kong was strongly influenced by an active ocean flow and a low pressure trough over the SCS in late July 2013. The consequently heavy rain and low temperature suppressed the photochemical reactions and caused the unusually high levels of VOCs (Figure S2 in the Supplementary material illustrates these events).

For details, please refer to lines 401-408, page 17 in the revised manuscript and Figure S2 in the supplementary material.

x in Figure 4 was defined.

Figure 4 Variation trends of the daily average propane and *n/i*-butanes at MK from June 2013 to May 2014. In the equation label x has units of days.

For details, please refer to lines 434-435, page 18 in the revised manuscript.