

General Comments:

The revised manuscript by Ling et al. has improved greatly – the significant revisions have added substance to their analysis as well as improved the overall flow of the document. The principal points are now articulated more clearly, so the value of the measurements can be recognized. Additionally, the revised figures are now easier to assess and now strengthen the manuscript. However, I do feel that there still is a fair amount of editorial work to be done (word choice and sentence structure) on this version of the manuscript. Overall, the authors have done a good job responding to the comments and modifying the manuscript.

Specific Comments:

Abstract:

P2L2-3: You should include “Mt. Tai Mo Shan” to introduce TMS and “Tsuen Wan” for TW

P2L4-8: Revise to something like:

Although the levels of parent hydrocarbons were much lower at TMS ($p < 0.05$), similar alkyl nitrate levels were found at both sites regardless of the elevation difference, suggesting various source contributions of alkyl nitrates at the two sites. ~~which was proved by the analysis of photochemical evolution of alkyl nitrates.~~

P2L23: NO is not “nitrous oxide” – change to nitric oxide

P2L25: This same point is addressed later, but in terms of ozone production, you should refer to it as this, ozone production – if the rates are negative, it means that ozone is being destroyed, but it’s still technically the “production rate”. Moreover, if it’s a “reduction rate”, a negative value (as in the abstract) would imply production (reads as a double negative), so be mindful of the language and sign convention.

P6L11-20: I suggest revising this paragraph – not clear as written.

P6L12-4: Revise to something like: Further to the east (32 km) are the urban areas and to the north (the entire NE to NW corridor) is the polluted PRD region.

P7L4: replace duration with period

P8L2&4: Start sentences with “Ozone” and “Carbon monoxide”

P8L7: “2s” should be “2 σ ”

P8L8: Delete space after “2-”

P8L10-19: Revise to something like the following:

The O₃ analyzer was calibrated by using a transfer standard (Thermo Environmental Instruments (TEI) 11 49PS), while the other analyzers were calibrated daily by analyzing scrubbed ambient air (TEI, Model 111) and a span gas mixture weekly with a NIST (National

Institute of Standards and Technology) traceable standard which was diluted to representative mixing ratios using a dynamic calibrator (EnviroNics, Inc., Model 6100). The standard (Scott-Marrin, Inc.) contained 156.5 ppmv CO ($\pm 2\%$), 15.64 ppmv SO₂ ($\pm 2\%$), and 15.55 ppmv NO ($\pm 2\%$). For the O₃, CO, NO and NO_x analyzers, a data logger (Environmental Systems Corporation Model 8816) was used to control the calibrations and to collect 1-minute data.

P9L10-13: Add commas:

DMS was a typical tracer for marine emissions, while Ox (*i.e.*, O₃ + NO₂) was used as the tracer of secondary formation through photochemical reactions, including the formation of alkyl nitrates, because O₃ shares a common photochemical source with alkyl nitrates (Simpson et al., 2006).

P9L14: what about ethane? (...methane, propane...); additionally, it's more common to report the branched isomer before the normal isomer – that is, *i*-*n*-butanes

P9L16: Start a new paragraph; replace “Different” with “Various”

P9L16-29: While you have used PMF v3.0, there is a newer version (5.0) which includes two additional methods to estimate error. I'm just curious why you are not using the most recent version.

It seems odd that you give only one linear correlation value for each study site (L19). The PMF model gives an R² for each species (predicted vs. measured concentration). How did you calculate an average R² and is that meaningful?

For L22-23, could you clarify what you mean by the Q values being stable? They should be similar between runs, is this what is meant? Also, can you elaborate on comparing the Q values in the robust mode being approximately equal to the degrees of freedom – I was not able to find this in the Friend et al. manuscript. Also, it would be useful to report the Q/Q_{exp} value for the model run – the value should be close to 1, indicating all data points were fit well.

While the number of factors is reasonable in the paper, it would be useful to show/comment on how the number of factors was chosen. The number of factors should be based on what's physically meaningful, but there is a quantitative metric as well. The overall Q/Q_{exp} ratio will decrease as you increase factor number (because the residual decreases).

P11-12: Figure 2 – the Russo et al. values should also be adjusted to be meaningfully comparable as they were on the old UCI calibration scale.

P13 – would be useful to show the ozone distributions at both sites (even as SI) to enhance the discussion.

P13L12: Replace “weather” with “meteorological”

P13L15: Replace first “weather” with “meteorological” and delete the second “weather”

P14L10-11: Revise to: "...suggesting an important source of C3-C4 alkyl nitrates which was photo-oxidation of the parent hydrocarbons. For the C1-C2 alkyl nitrates, the temporal pattern..."

Additionally, "peaks and troughs" is used several times throughout the manuscript – I would recommend using more appropriate wording corresponding to the key point to be made (i.e., max & min, temporal, etc.)

P15: Figure 4 – convert the NMHC mixing ratios to ppbv for clarity.

P15L10-13: Revise to something like:

Although the levels of the parent hydrocarbons were lower at TMS, similar values of alkyl nitrates were observed at both sites, regardless of the elevation, suggesting the contributions of different sources and/or the influences of different air masses.

P16L1: Change "in-depth studied" to "analyzed"

P16L16: Change "destruction" to something like "removal"

P17L20: Change "could be" with "were"

P17L29: Change "laid" to "were positioned"

P19-20L20-3: Revise to something like the following:

For example, the average MeONO₂ and EtONO₂ mixing ratios at Hok Tsui, a PRD regional background site, were 10.4 ± 0.7 and 9.6 ± 0.7 pptv (unpublished data, 2001-2002), respectively.

P21L15-23: Revise to something like the following:

The average yields of 1- and 2-PrONO₂ were 0.032 ± 0.004 and 0.22 ± 0.02 , respectively, higher than the laboratory kinetic values by factors of 4–9 (Kwok and Atkinson, 1995). This confirms the presence of additional emissions of C3 alkyl nitrates at TW, including locally-emitted C3 alkyl nitrates and/or secondary formation other than the production pathway from propane to proxyl radical and PrONO₂ (Reeves et al., 2007; Worton et al., 2010). The slope of 1-PrONO₂ to 2-PrONO₂ at TW was 0.15 ($R^2 = 0.80$, $p < 0.05$), lower than the theoretical ratio of 0.21, further demonstrating the influence of other significant sources on ambient mixing ratios of C3 alkyl nitrates at TW.

P21L29-30: What do you mean by the signatures were damaged? "...the source signatures of alkyl nitrates and their parent hydrocarbons **were damaged** at this mountain site."

P23L11: Change to: "By summing up the mass of the alkyl nitrates in each source category,"

P23L11&15: You aren't reporting "concentrations" here, these are mixing ratios or mole fractions. Especially for line 15, you say "absolute concentration" – this is a mole fraction/mixing ratio – concentration would be in molecules/cm³.

P23L23: Delete “On the other hand”, and say “For the...”

P23L28: What are “stronger” photochemical reactions?

P24 – Figure 9 – not absolute concentration – what is presented is the summed mixing ratio

P24L14-20: Is the point that you are trying to make that local sources are also influencing the mesoscale transport?

Also – peaks and troughs: do you mean max and min?

P24L22: add “for biomass burning” after “1 pptv”

P25L2: add “emissions were” after “biomass burning”

P25L5: Change “(data not shown here)” to “(not shown)”

P26L11: Change to: “Except for MeONO₂,”

P26L22: What do you mean by “gradually undertaken”? Revise accordingly

P26L23: add “levels of” after ambient

P26L24-27: Revise this sentence. What do you mean by “the air masses flew down the mountain”?

P26L30-31: should read: “...when the valley breeze occurred.”

P27L4: end the sentence after respectively; revise the following three lines.

P27L17: Delete “Indeed”

P27L19: Delete “firmly” and change “confirmed” to “corroborated”

P27L20-21: Change to something like the following:

By excluding the locally-formed alkyl nitrates from their overall levels, the contribution of regional sources to alkyl nitrates was determined for TMS.

P27L25-27: Change to something like the following:

It is noteworthy that the regional alkyl nitrates included influences from all source categories (photochemical, biomass burning and oceanic) in the inland PRD region.

P28L8: Change “of help to evaluate” to “useful for evaluating”

P28L12: Explain what is meant by “recover the loss of O₃ due to the NO titration.”? Revise accordingly.

P28L23: Delete “It was obvious that”

P29L3: Again, as stated previously, a negative ozone reduction rate would imply production. I would refer to this as the ozone production rate, with a negative value meaning that there is ozone loss, or have it be an ozone loss rate, with the value being positive.

P29L11: Delete “i.e.,”

P29L13: simulation should be plural (simulations)

P30L10: “remarkable” is not an appropriate word choice, please revise

P30L14-18: Reads awkwardly – please revise

P30L22: Yet again, change to ozone production rate or ozone loss rate with the appropriate sign convention.

P30L23-26: Change to something such as the following:

The findings of this study will aid in understanding the source contributions and photochemical formation pathways of alkyl nitrates in Hong Kong’s mountainous areas.