Point-by-point response to editor comments:

Comments to the Author:

The manuscript has improved substantially after the revision based on reviewers' comments. I think the authors have adequately addressed the reviewers' concerns. I have some additional comments that I would like the authors to clarify/address prior to the publication of this manuscript:

1. Page 5 line 147. The recovery for the lowest mass is biased high while that for the higher mass is biased low. How could these potentially affect data analysis and conclusions of the manuscript?

Response: The results of the monthly multi-point calibrations and weekly single-point verifications show that the lower mass biased higher compared to the higher mass. We note that the measurement uncertainties become higher when the carbon concentrations are low. The carbon concentrations at MK AQMS, due to the site characteristics, were maintained at relatively higher levels during most of the time in a day except for the mid-night/early morning period. The (OC/EC) ratio for primary vehicular emissions was obtained mainly through examination of the traffic rush-hour data. Hence the conclusions of the manuscript would not be affected but the very low carbon concentration (mainly during midnight) would be overestimated.

2. Page 7 line 210. It seems like "OC" should actually be "organic vapor"?

Response: Corrected.

3. Page 7 line 223. If the field analyzer only collected data for ¾ of the 24 hour period, could this be a reason for the low OC values as compared to Partisol-OC and HV-OC as well?

Response: We would like to clarify that the comparison was made between the time-averaged concentrations over 3/4 of the 24 hr by the RT-OCEC analyzer and those over the entire 24 hr by the filter-based off-line measurements. The shorter sampling period does not necessarily translate to lower time-averaged EC and OC concentrations by the RT-ECOC analyzer. The difference in concentration resulted from the different sampling periods would be minimized when the concentrations are relatively stable. However it is not the case in MK AQMS which is a roadside station. The fluctuations of the concentration within the 1/4 of the 24-hr period which the RT-OCEC did not sample may partially contribute to the differences between the continuous and the filter-based measurements. It is difficult to conclude whether the fluctuations led to lower carbon concentrations since the carbon levels in the unsampled 1/4 period could be either higher or lower than those in the sampled period.

4. Page 8 line 250-251. The authors suggested that the higher OC in winter is due to pollutants transported into the MK area from elsewhere. The authors need to provide a reasonable as to how they can rule out any possible additional local OC sources in winter.

Response: The major local primary OC sources in the study area are believed to be from on-road vehicles and cooking-related activities. The little seasonal variations of EC concentration suggest that the contributions from vehicular emissions didn't vary much among seasons. Although we do not have measurement data to determine the seasonality of cooking emissions, it is reasonable to expect cooking activities do not have strong season-dependence. As such, more particle-phase cooking POC might be expected in the winter due to the lower ambient temperature favoring more partitioning in the particle phase. The "local" wind data (directions and speeds) at MK AQMS (Figure R1) indicates that the dispersion conditions of air pollutants at MK are similar in different seasons. On the other hand, the background wind for the study area and for the entire Hong Kong region (Figures R2&R3) indicate that

the prevailing winds in winter were northerly and northeasterly, which favored the pollutant transport from the mainland China. We further looked at spatial variations of OC observed in the PM10 monitoring network of nine general stations and the MK roadside station across Hong Kong, which provide consistent evidence for the important contribution of OC transported from outside Hong Kong (see the revised text given below).

Revised text (Lines 248-266):

"For OC, unlike the EC concentrations which maintained at a stable level during the study period, its concentrations were evidently higher in winter months. The OC increment in winter over summer was mainly attributed to air pollutants transported into the MK area from elsewhere if we consider relevant OC and EC measurement data in Hong Kong reported for a wider spatial coverage. A previous study examined PM_{10} EC and OC data in a monitoring network of nine general stations and the MK roadside station across Hong Kong from 1998 to 2001 (Yu et al., 2004). The winter average OC was found to be 5.7–10.5 $\mu g/m^3$ higher than the summer average OC across the monitoring network, with the highest OC seasonal increment associated with the station in the northernmost of the Hong Kong territory and the OC increment in MK (7.6 $\mu g/m^3$) similar to those recorded at a cluster of six general stations in the same airshed to the south of Tai Mo Shan (5.7–7.9 $\mu g/m^3$). Such spatial variation characteristics strongly suggest that the winter OC increment over the summer in Hong Kong was dominated by regional/super-regional sources. This is also consistent with the seasonality of prevailing background wind for Hong Kong, with northerly and northeasterly winds prevailing in winter that bring more polluted air masses from mainland China (Yu et al., 2004). Although additional local sources in winter, such as more of the semi-volatile cooking emissions partitioning to the particle phase, could not be ruled out, their contributions to the winter OC increment were most likely minor in comparison with outside sources."

Reference:

J. Z. Yu, J. W. T. Tung, A. W. M. Wu, A. K. H. Lau, P. K.-K. Louie, and J. C. H. Fung, Abundance and seasonal characteristics of elemental and organic carbon in Hong Kong, Atmos. Environ., 38 (10), 2004, 1511-1521.

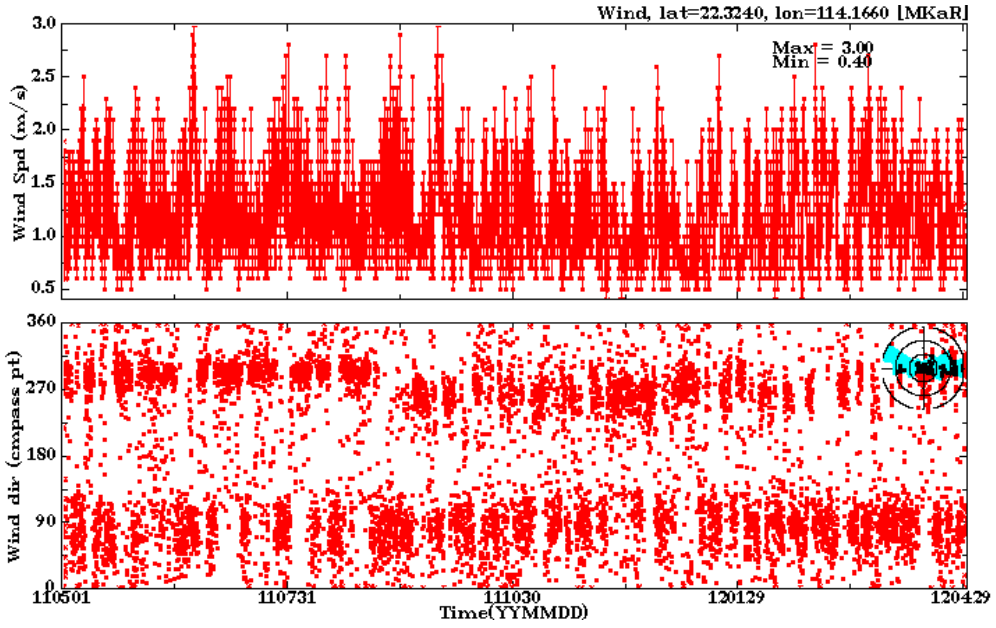

Figure R1. Wind speeds (upper) and wind directions (lower) at MK AQMS during the study period.

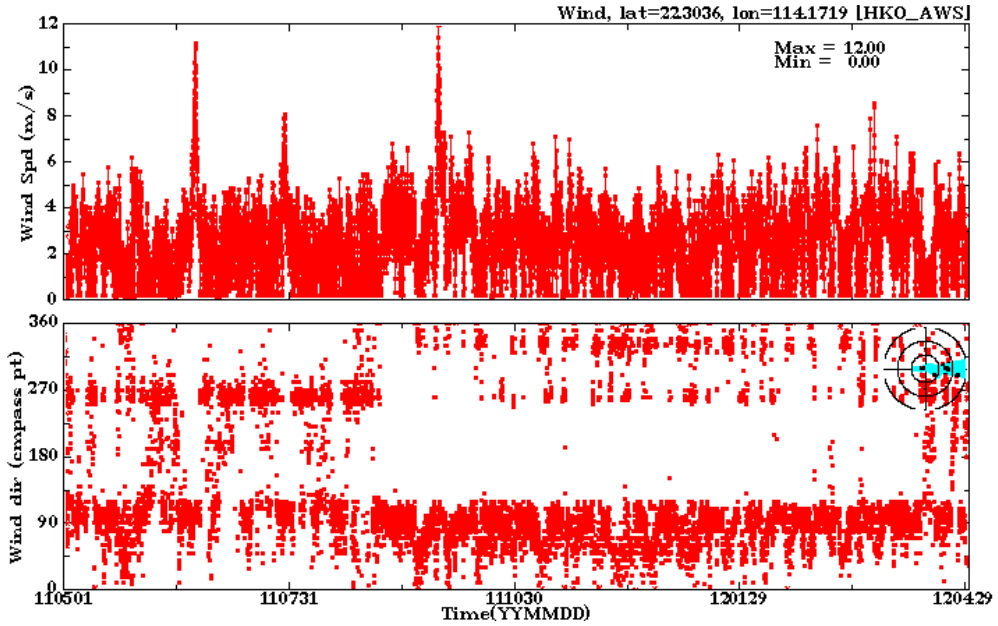

Figure R2. Wind speeds (upper) and wind directions (lower) at the Hong Kong Observatory during the study period.

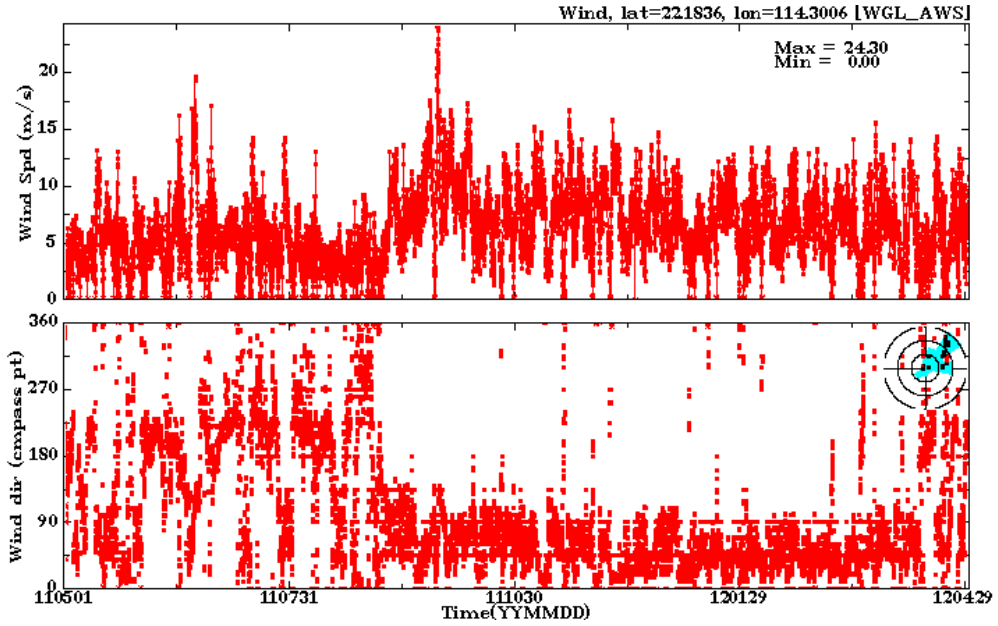

Figure R3. Wind speeds (upper) and wind directions (lower) at Waglan Island during the study period.

5. Page 8, line 255. The authors wrote: “The difference of OC concentrations between weekdays and holidays were more significant in summer than the other seasons”. What is the basis for this statement? Are the authors just referring to an average value? From the data in Figure 5 I don’t think this statement is well-justified. It seems that the difference is the largest (or “consistent”) for “Jan 2012”. This would affect subsequent discussions in this paragraph. Please explain.

6. Page 9 line 257-258. Please provide citation (s) for this statement.

Response: We agree with the editor that the statement was not carefully thought-out. The paragraph is now rephrased as follows,

Lines 267-280

"The diurnal variations of carbon concentrations for weekdays (Mon–Fri), Saturdays, and holidays (Sunday and public holidays) were examined for individual months (Fig. S1 & Fig. S2) and four months were selected to represent the different seasons (Figure 3, August for summer, October for fall, January for winter, and March for spring). The EC concentrations on holidays, especially during daytime, were consistently lower in individual months, indicating the on-road diesel-powered vehicles as its major sources (i.e. reduced bus schedule on holidays) and the "local" characteristics. The difference of OC concentrations between weekdays and holidays was less significant in all seasons. The potential reasons include: (1) more gasoline-powered vehicles (e.g. private cars) would offset the OC concentration reduction due to fewer diesel-powered vehicles; (2) cooking-related activities might make greater contributions during holidays, and (3) polluted air masses transported from elsewhere outside Hong Kong make a more sizable contribution to OC, especially in winter and the two transitional seasons (Yu et al., 2004), obscuring the weekday-holiday variation in primary OC from vehicles."

7. Page 9 line 280. What is the source of the large ozone peak at night (4am)? I understand that it can decrease in early morning because of titration by NO. But it is not clear why there is an increase from midnight to 4am. Please explain. I agree with Reviewer 2 that comparing OC to ozone here might not be as informative. Do the authors have sulfate data? That might be a better indication of secondary chemistry.

Response: (1) Explanation for the early morning ozone peak: Integrated Process Analysis using chemical transport modeling (private communication, Dr. Ying Li at HKUST) shows that, during nighttime, vertical transport (advection and diffusion) and the chemical destruction (NO_x titration) are the major processes controlling O_3 abundance in Hong Kong. The drop of Ozone concentration in the early part of the night (before mid-night) is due to NO_x titration. This process is prominent at the roadside station and the urban stations as a result of abundant NO_x emissions. An ozone concentration gradient is thus formed horizontally decreasing from the outer background areas to the urban areas as well as vertically decreasing from the upper level to the ground. The NO_x titration effect is significantly weakened as the night progresses and NO_x emissions from vehicles are reduced. With wind blowing, air masses from the outer background areas and from the upper levels (that contain higher O_3 concentrations) are mixed with the urban air (depleted with O_3) and consequently increase O_3 concentrations in the urban center. The concerted result of reduced NO_x titration and mixing-in of outside and upper air masses is thought to account for the early morning O_3 peak around 3–5 am local time that reaches the level of background ambient air (< 20 ppb). O_3 declines again around 6 am as NO_x emissions pick up.

(2) We do not have online sulfate data for the MK site. Sulfate in Hong Kong is dominated by formation process at the regional scale, as its spatial distribution in Hong Kong is fairly uniform (Yuan et al., 2006). We feel the diurnal variation of sulfate probably does not provide directly relevant information for understanding of diurnal variation of ozone, as the NO_x titration process greatly affects O_3 dynamics while not so for sulfate.

The text below is added to the manuscript to explain the early morning O_3 peak:

Lines 303-311:

"We note that there was the consistent presence of an early morning O_3 peak around 3–5 am local time in all months. This nighttime ozone peak is also observed across all the urban monitoring sites in Hong Kong. Integrated process analysis using chemical transport modeling (private communication, Dr. Ying Li at HKUST) shows that vertical transport (advection and diffusion) and NO_x titration are the major processes controlling nighttime O_3 abundance in Hong Kong. The joint result of reduced NO_x titration and mixing-in of outside and upper air masses, which contain higher O_3 concentrations, is thought to account for the early morning O_3 peak that elevates to the level of background ambient air (< 20 ppb)."

Figure 5 caption is revised to:

"Diurnal variations of OC ($\mu gC/m^3$) and O_3 ($\mu g/m^3$) at MK AQMS during different seasons. See text for the explanation of the early morning O_3 peak."

Reference:

Yuan, Z. B., J. Z. Yu, A. K. H. Lau, P. K. K. Louie, and J. C. H. Fung: Application of positive matrix factorization in estimating aerosol secondary organic carbon in Hong Kong and its relationship with secondary sulfate, Atmos. Chem. Phys., 6, 25-34, 2006.

8. Page 9 line 284. As photochemical activities should be stronger in summer, if the first OC peak is from SOA, one would expect the peak to be larger in summer than winter. But from Figure 5, it seems that the OC peak is stronger in winter than summer?

Response: We note that in Hong Kong it is not necessary that more local SOC would be formed in summer than in winter. While the stronger solar irradiation would speed up reactions leading to secondary OC formation, there are at least two counteracting factors: (1) the higher ambient temperature would favor more of the semi-volatile SOC to partition in the gas-phase in the summertime; (2) the VOC precursors are more abundant in winter due to contribution from regional transport (Yuan et al., 2006, Lau et al., 2010). As a subtropical location, winter solar irradiation in Hong Kong is probably not a strong limiting factor for SOC formation. Higher SOC in winter than in summer for Hong Kong has been discussed in details in the paper by Yuan et al (2006).

References:

A. K. H. Lau, Z. B. Yuan, J. Z. Yu, P. K. K. Louie, Source apportionment of ambient volatile organic compounds in Hong Kong, Sci. Total Environ., 408, 4138-4149, 2010.

Z. B. Yuan, J. Z. Yu, A. K. H. Lau, P. K. K. Louie, J. C. H. Fung, Application of Positive Matrix Factorization in Estimating Aerosol Secondary Organic Carbon in Hong Kong and Its Relationship with Secondary Sulfate, Atmos. Chem. Phys., 6, 25-34, 2006.

Technical comments

1. Page 2 line 55-56. Sentence not clear.

Response: The sentence was rephrased as follows,

"A significant fraction of $PM_{2.5}$ mass, ranging from 16% in rural areas to around 40% in urban/roadside areas, was identified as carbonaceous aerosols in Hong Kong (DRI, 2010; HKUST, 2013)."

2. Page 5 line 143, need a space between 0.60 and ug/m3.

Response: Corrected.

3. Page 7 line 241-243. Sentence not clear.

Response: The sentence was rephrased as follows,

"This can be explained by the quite comparable EC concentrations throughout the year while $PM_{2.5}$ concentrations were much lower during summertime than wintertime."

4. Page 30. Need to add "EC" to y-axis label.

Response: Corrected.

5. Page 31. Please use a different color for NOx (so that it's not similar to OC to avoid confusion).

Response: Corrected.