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Point by Point Response to Review Comments (ACPD-14-57-2014)
By Anonymous Referee #3
Comment
Huang et al. reported seasonal observations of carbonaceous observations using the Sunset OCEC monitor in Hong Kong. The modified NIOSH method protocol was used for particle analysis and the EC tracer method was used to decipher the contribution of vehicular carbon contributions. The authors demonstrated a thorough effort to find the suitable determination of the OC/ECpri using the EC tracer method. These discussions are of interest to the general audience with interest in air quality and atmospheric processes. A list of revision suggestions is provided for the authors to strengthen the paper before publication.
Page 85, abstract, line 27, PM was used not PM2.5. Need to be consistent in the paper. Suggest spell out particular matter or aerosol, as PM2.5 could not represent PM10 or ultrafine particles.

Response: Suggestion taken. We have replaced PM with $PM_{2.5}$ throughout the manuscript to be clear on what we refer in this work is $PM_{2.5}$.

Comment
Page 59, line 13, "Since EC undergoes little chemical transformation…", not sure I agree with this statement, as more and more observational evidence has suggested that black carbon is not as simple as what we had thought. Would suggest the authors modify this by saying "EC has been considered to undergo little chemical transformation, and thus it has been used as an indicator for primary combustion emissions."

Response: Suggestion taken and revision made accordingly.

Comment
Page 59, second paragraph, & page 60 2nd paragraph, mixed usage of PM and PM2.5.

Response: Revised and thanks the reviewer for pointing it out.

Comment
Page 59, line 26, with respect to not in respect to
Response: Revised

Comment
Page 60, line 14, please add "…processes that happen at a faster time scale". One could argue that the semi-continuous OCEC measurements could not capture fast chemical transformation as well. I would suggest the authors comment on this.

Response: Advice taken and the sentence is rephrased as "…and they are inherently incapable of capturing the dynamics of pollutant emissions and atmospheric chemical conversion processes that happen on a faster time scale".

We agree with the reviewer's comment that the semi-continuous OCEC measurements may not be able to capture very fast chemical transformation (e.g, at a scale of minutes or shorter). However, the instrument with 1-hour time resolution does a better job in obtaining diurnal profiles of OC and EC than 24-hr integrated filter measurements. For example, as we demonstrate in the paper, the hourly ECOC data allows us to examine data obtained during rush hours to extract $(OC/EC)_{vehicle}$.

Comment
Page 60, line 23, could you describe how high the time-resolution is? Generally, Sunset OCEC measurements provide hourly measurements. It is faster than daily, but not really that high compared to other techniques. Also see my previous comment.

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Response: We specified the time resolution of the measurements in the previous sentence as "Measurements of hourly OC and EC concentrations were conducted for a year from May 2011 to April 2012".

Comment
Page 61, line 3, instead of "one set of" just use "A".

Response: Revised.

Comment
Page 61, line 7, the flow rate for Sunset instrument is normally 8 lpm, could you comment on why you chose 8.2 lpm? Did you do any corrections of your data considering the flow rate may affect the sample mass calculation?

Response: The sample collection flow rate was pre-set by the instrument controlling software. It is found in the user manual that a more accurate statement of the sampling flow rate should be "around 8 LPM". The revisions were accordingly made in the manuscript. We thank the reviewer for pointing it out.

In the data treatment, no corrections were done since the instrument monitored the flow rates during sampling so as to calculate the sampled volume for each sample. The hourly OC and EC concentrations were then calculated by the total amount of carbon measured divided by the sampled volume.

Comment
Page 64, line 10-14, could you explain more clearly the rationale to compare two different protocols? What new insights are you trying to bring? Also, for Fig. 1 on Page 86, it will be easier to see the plots by putting them into two rows, i.e., the first row for RT vs. Partisol comparisons and the second row for RT vs. HV comparisons.

Response: The comparisons between semi-continuous measurements with two independent datasets obtained from 24-hour integrated filter measurements served as a higher level of data validation in order to identify any atypical data or systematic bias [*USEPA*, Quality Assurance Guidance Document, EPA-454/R-01-001, 2000].
Figure 1 has been revised accordingly and we thank the reviewer for this constructive suggestion.

Comment
Page 64, line 15, why do you decide to use zero-intercept linear regression analysis?

Response: The instrument blanks for both bench-top aerosol carbon analyzer and the semi-continuous OC-EC field analyzer were estimated to be zero (by comparing the absolute blank values, the analytical uncertainties of the blank and the instrumental uncertainties of the blank). Hence, the zero-intercept linear regression analysis was applied to the comparisons of the datasets. This information is added to the revised manuscript.

Comment
Page 64, line 17 to Page 65, line 4, these equations are very simple, they should not take the main content of the paper. They belong to supplemental materials.

Response: Advice taken and the equations are moved to the supplemental materials.

Comment

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Page 65, line 5-28, suggest the author refer to Fig. 1 when discussing the results here. Also suggest the authors bring out the key point of these comparisons. Why are you doing these measurement comparisons? What new insights are we getting from this exercises?

Response: Please see the response to the previous comment related to "Page 64, line 10-14."

Comment

Page 66, line 7, the number of significant numbers for reporting OC and EC concentrations. Considering the detection limit of Sunset OCEC is on the order of 0.1-0.2 mg/m3 (see Bauer et al., 2009 and ref therein), it is pointless to report more than 1 digit after the decimal point. I suggest the authors revise these throughout the manuscript.

Ref: Characterization of the Sunset Semi-Continuous carbon Aerosol Analyzer, J Air & Waste Management, 2009, 59(7), DOI:10.3155/1047-3289.59.7.826, Jace J. Bauer, Xiao-Ying Yu, Rober Cary, Nels Laulainen & Berkowitz.

Response: Advice taken and the revisions made accordingly.

Comment

Page 66, no previous description of trace gas measurements, i.e., NOx, O3, etc., please add.

Response: The hourly data including PM2.5 mass, NO, NO2, and O3 at the sampling site are provided by the Hong Kong Environmental Protection Department (HKEPD). This information is now added to Section 2.1 Sampling equipment and method.

Comment

Page 68-69, the EC tracer method discussions, suggest the author consider adding additional references of more recent results using the EC tracer method and how to arrive the OC/EC pri and enrich the discussions. Additional references may include the following:

Ref: Primary and secondary organic carbon downwind of Mexico City, ACP, 2009, 9, 6793-6814. doi: 10.5194/acp-9-6793-2009, X.-Y. Yu, R.A. Cary, and N. S. Laulainen

Linear regression techniques for use in the EC tracer method of secondary organic aerosol estimation, Atm. Env., 2006, 7546-7556, R.D. Saylor, ES Edgerton, BE Hartsell, doi: http://dx.doi.org/10.1016/j.atmosenv.2006.07.018

Response: The additional recent references are now added.

Comment

Page 70, paragraph 1, since wind seems to play a role in the OC/EC min, it is useful to show representative wind speed and wind direction data in the paper. For example, wind roses of the seasonal pattern could be useful in elucidating your points.

Response: We totally agree with the reviewer that wind information is important in source identification. Unfortunately, the wind data at the MK AQMS is less meaningful since the monitoring station is located at the junction of two busy roads in downtown Hong Kong and surrounded by many tall buildings along the road. The street-canyon effect is significant. As a result, the wind observed at this site is either easterly or westerly (along the road) and at fairly constant speeds (see example wind data in the figure below). The wind data at locations in HK with open surroundings better represent transport of regional pollutants. The seasonal wind variations in Hong Kong are well understood and have been described in details in Yu et al (2004). Therefore, we decide not to include the wind information local to Mong Kok for discussion in this manuscript. Instead we now add the Yu et al (2004) as a reference for further information on wind.

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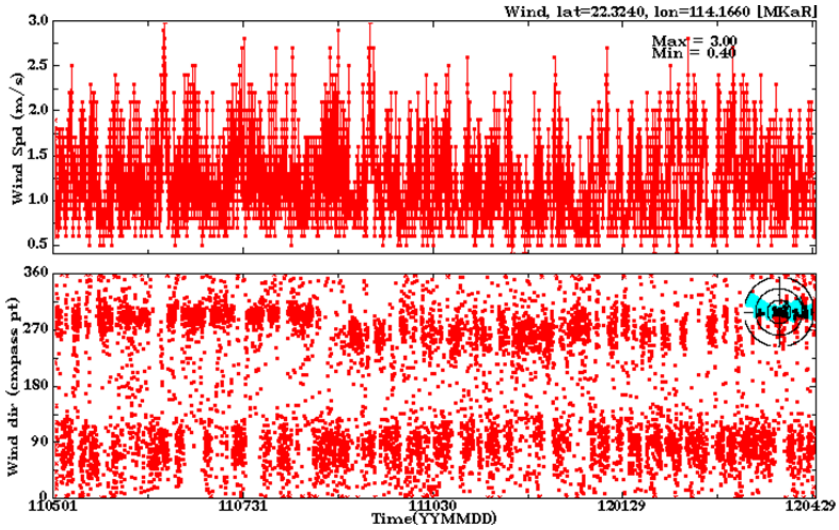

(Source: ENVF database)

Comment

Page 77, lines 6-11, the conclusion is somewhat unsupported by the data presented in the paper. No POC or SOC data were presented after the determination of OC/EC pri. Maybe this will go to a second paper? These should be moved to discussions or implications of your results or deleted, although the words all sound good.

I also would like to ask the authors to think about the most important take-home message after the rigorous exercise to determine OC/EC pri and compare with different methods. How would the community utilize your results in analyzing the Sunset observations in the future?

I recommend revisions in the conclusion section.

Response: We would like to clarify that our analysis of hourly OCEC data and knowledge about this site have led us to conclude that the data is more suitable for deriving a $(OC/EC)_{vehicle}$ ratio instead of $(OC/EC)_{pri}$, unlike other ECOC measurement studies in the literature. Consequently, the focus of this work is on estimating the carbonaceous aerosol contribution from vehicular sources to $PM_{2.5}$, not the relative contributions of POC and SOC.

The diurnal variation of OC shows a nighttime OC peak around 19:0022:00 LT. This observation combined with the knowledge of the sampling site characteristics (e.g, busy commercial district with numerous restaurants in the neighborhood and more private cars on the road in the early evening hours) suggest that there are a variety of time-varying primary emission sources in the sampling area. Hence, using a single value as the OC-to-EC ratio (derived from the minimum OC/EC ratio approach) for primary emissions to estimate POC and SOC may cause sizable biases especially when certain primary sources which emitted little EC were dominant (e.g. cooking). For these reasons, we analyzed data subsets which were obtained from a specific period of time and derived an OC-to-EC ratio that could reasonably represent the vehicular emission sources. With the derived $(OC/EC)_{vehicle}$, the OC generated by vehicle-related sources can subsequently be estimated.

In the same time, we agree with the reviewer that continuous efforts should be taken to investigate a more accurate way to differentiate POC and SOC. Specific to our sampling site, other tracers, such as C_{16} and C_{18} fatty acids for cooking-related sources, are needed. Alternative approaches, such as SOA tracers in conjunction with the bulk OC and EC measurements, may be tried to get a better handle on SOA estimation. We feel these implications for future work are important to merit mentioning in the conclusion section instead of being buried in the discussion section.