<u>**Reviews for acp-2017-288-version2**</u>

Second review of the manuscript entitled “Long-term (2001-2012) trends of carbonaceous aerosols from remote island in the western North Pacific: an outflow region of Asian pollutants and dust”. The revision is with significant improvement, but still some corrections are required before accepted for publication and I'm terming this again a minor revision.

Some technical edits are offered as following and listed below with other comments.

Specific comments:

1. Line 26: were associated
2. Line 40: This point was
3. Line 78: (IPCC, 2013)
4. Line 140: were low or
5. Line 141: which were analyzed….
6. Line 161: were corrected..
7. Line 162: were less than..
8. Line 167: equation was used
9. Line 167–168: all trends were assessed by using
10. Line 170: analyses are
11. Line 180: “over South and Southeast Asia” Also add “East Asia”. Biomass burning is also more frequent in continental East Asia in winter. From BT and fire count analysis you are getting the intensity of biomass burning over all regions.
12. Line 181: air masses were
13. Line 183: air masses were
14. Line 190: There was…
15. Line 198: All measured species (EC, OC, and WSOC) clearly showed…………
16. Line 200: The seasonal variation in carbonaceous aerosols observed in this study was found
17. Line 204: In this version you’re not discussing “synoptic wind circulation”. Replace it with “air mass back trajectories”.
18. Line 206: Relatively higher
19. Line 207: were lower

20. Line 210: air masses were
21. Line 234–236: "The OC/EC ratios > 2.0 have been used to point out the presence of secondary organic aerosols (SOA) (Cao et al., 2003; Chow et al., 1996; Kunwar and Kawamura, 2014)". It is suggested to add one recent work on impact of Asian outflow over East Asia here as reference.

    S. K. Pani, C. T. Lee, C. C. K. Chou, K. Shimada, S. Hatakeyama, A. Takami, S. H. Wang, and N. H. Lin (2017), Chemical Characterization of Wintertime Aerosols over Islands and Mountains in East Asia: Impacts of the Continental Asian Outflow, Aerosol and Air Quality Research, doi: 10.4209/aaqr.2017.03.0097.
22. Line 245: are the tracers
23. Line 254–256: which clearly showed that air masses were occasionally coming from Southeast Asia (e.g., Indonesia, Malaysia, and New Guinea etc.)…
24. Line 277: WSOC/OC ratios were….
25. Line 280: SOA formation was enhanced
26. Line 289: VOCs (Gilardoni et al., 2016; Youn et al., 2013) over continental East Asia…….
27. Line 305: It was seen
28. Line 376–377: The RF of aerosol is generally estimated by using the aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetry parameter (Pani et al., 2016).

    Pani, S.K., Wang, S.H., Lin, N.H., Tsay, S.C., Lolli, S., Chuang, M.T., Lee, C.T., Chantara, S. and Yu, J.Y. (2016). Assessment of aerosol optical property and radiative effect for the layer decoupling cases over the northern South China Sea during the 7-SEAS/Dongsha Experiment. J. Geophys. Res. 121: 4894–4906. doi:10.1002/2015JD024601.
29. Line 377–379: OC (except for brown carbon) and SO_4^{2-} mainly scatter the short-wave incoming solar radiation whereas EC strongly absorb the short-wave solar radiation as well as the long-wave outgoing terrestrial radiation in the atmosphere (Charlson et al., 1992; Ramanathan et al., 2001).

    Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. D. Coakley, J. E. Hansen, and D. J. Hofmann (1992), Climate forcing by anthropogenic aerosols, Science, 255, 423–430.

Ramanathan, V., P. J. Crutzen, J. T. Kiehl, and D. Rosenfeld (2001), Aerosols, climate and the hydrological cycle, Science, 294, 2119–2124.

30. Line 918–920: Figure 3. Box-whisker plots of monthly variations of carbonaceous aerosol components (μg m^{-3}) and some specific mass ratios at Chichijima Island in the western North Pacific during 2001-2012.
31. Line 977–980: Figure 4. Annual trends (time series) in the concentrations (μg m^{-3}) of carbonaceous aerosol components, water-soluble ionic tracer compound (MSA^-), and some specific mass ratios during 2001-2012 over the western North Pacific. The liner trend equation ($y = mx + c$) is also shown for the each annual trend.
32. Line 999: Figure 5. Regression analysis between WSOC and MODIS-derived cloud condensation nuclei (CCN) concentrations over the western North Pacific.
33. The abscissa range of Fig. S1 and S3 should be consistent. Reposition the text (year) in X-axis to the middle and flip it to horizontal format as in Fig. S2. For Figure S3, please do the same.
34. Figure S2. Annual mean variations (μg m^{-3}) of carbonaceous species, water-soluble ionic tracer compound (MSA^-), and some specific mass ratios during 2001-2012 over the western North Pacific.
35. As biomass burning aerosol in SE and E Asia is concerned, a special issue (Nov., 2016) of the Seven South East Asian Studies (7-SEAS) on the journal of Aerosol and Air Quality Research gives the most updated information that can be included for comparison and discussion.

Overall, this paper has been improved by the revisions. The paper still contains grammatical mistakes and should be read through carefully and edited. The language should be toned down in some cases to be less definitive and more suggestive. The main conclusion is still that seasonal variations in wind patterns change the organic aerosol concentration. There is some discussion of decreasing particles from fossil fuel emissions. There is a small section on the contribution of marine aerosol, which is not complete, and the discussion of CCN at the end is still overreaching.

General Comments:
The OC/EC description has been improved, and Table 2 helps a lot in explaining the ratios measured in different sources. Based on Table 2, the split at OC/EC higher or lower than 2 may not be the best indicator (i.e. the first line with fossil fuel combustion has OC/EC of 4.0, 4.1, and 1.1). The authors could point out this range and then state that their values are much larger in the summer and still greater than the cutoff in the winter to spring, as shown.

The part about the lower WSOC/OC ratio in the summer suggesting an ocean-derived source of organic carbon is still speculation. It is fine to include this paragraph with these references, but there is no concrete evidence here. This just says that other studies have measured OC in ocean-derived aerosol. Additionally, there is a wind-speed threshold required to produce breaking waves that in turn produce primary marine aerosol. The "low speed easterly winds" mentioned should include an actual wind speed and a reference to the speed needed for breaking waves, if the authors keep this discussion. The most that could be inferred from this data set, not including any correlations to sea salt, would be that air masses that originate over the ocean have lower concentrations of OC and are mixing and decreasing the total OC.

The authors need to include references if they are going to state that OC particles "majorly scatter" solar radiation to the same degree that sulfate or other salts do. At the very least, this should be rephrased to state that EC scatters less than OC and that EC is more absorbing. That is well known, whereas the scattering efficiency of OC alone is not.

The original comment on the "Atmospheric Implications" was not fully addressed. Figure 5 with one correlation between CCN and WSOC is not enough to show that "water-soluble organic matter also plays an important role in CCN formation." At the very least, this needs to be rephrased to state the uncertainty. There are many more factors that contribute to CCN activation, so a single correlation (and not showing the possible correlations between WSOC and salts or particle size, etc.) is not enough. Figure 5 does not show a direct link, and the new text is not enough.

Specific Comments:
Figure 2: The new figure is much better, but it still runs into the problem of 12 years of data overlapping in the same plot. The back trajectories show the general trends, and the fire data is interesting. Could both of those be colored by the year? Here, it is unclear if one year had a lot of fires and others had none or if there are always fires in the same area. Using a color bar for the years would be also useful since the back trajectories don't

perfectly overlap. The MODIS data also stops at 80E – mention that in the caption to be clear.

Line 248: Add numbers to the ratios to describe "higher" and "lower", especially since the higher and lower ratios are indicative of different sources.

Line 338: Why would there have been higher ocean-derived OC emissions during 2007-2008? There is no evidence presented here supporting a one-year difference in marine aerosol emissions. This should be removed.

Technical Corrections:
These are examples of technical corrections in the abstract only. The whole paper should be checked for grammatical errors and corrected.
Line 32: Remove "that"
Line 35: Add "a" after "found"
Line 36: Change to: "that the concentration of biomass-burning-derived carbonaceous aerosols has increased"
Line 37: Change "are" to "has"
Line 40: Change "source" to "sources"
Line 41: Add "a" after "found"
Lines 42-44: This is unclear.

More examples of technical corrections:
Line 54: Remove "invisible"
Line 57: Remove "hence"
Line 59: This implies that EC is volatile - rephrase
Line 60: Change "while" to "and"
Line 61: Add ", which" after "BC)"
Line 63: Change "some" to "a" and remove "so called"
Line 67: Remove "about"; change "are" to "is"
Line 83: Change "exist" to "existing"
Line 91: Change to "dominating"
Line 96: Change "increased" to "increase in"
Line 119: Change "is" to "are"
Line 181: Fix "the air masses are stronger to transport"
Line 184: Add "and" before "mostly"
Line 226: Change to "EC particles are primary and predominately come"
Line 375: Add "and" before "thus"