Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2017-288-RC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term (2001–2012) trends of carbonaceous aerosols from remote island in the western North Pacific: an outflow region of Asian pollutants and dust" *by* Suresh K. R. Boreddy et al.

Anonymous Referee #2

Received and published: 19 May 2017

This paper looks at measurements of organic carbon (OC), elemental carbon (EC), and water soluble OC (WSOC) from 2001-2013 collected on a remote island off the coast of Japan. While the timescale of the measurements is impressive, the paper lacks any suitable interpretation beyond the idea that polluted air masses come from Asia in the winter. The main conclusion is that seasonal variations in organic aerosol are based on the seasonal wind patterns. There is some discussion of ratios of the three main measurements, but their interpretations are not well explained. Some conclusions are not supported by the results of analysis. This paper could be interesting to the community, but it needs major revisions before publication.

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Discussion paper

Major Comments. The use of OC and EC and OC/EC to attribute air masses and emissions to local sources needs more explanation. For example, Lines 232-234 are not explained well. It is saying that because the EC concentrations are low, there is a negligible contribution of local emissions at the sampling site? Couldn't the concentrations of local emissions just be smaller than the concentrations from other sources? Low concentrations alone do not rule out contributions. Similarly in the following sentences, there needs to be more explanation on why a higher ratio of OC/EC in the summer indicates less local anthropogenic sources. Again, this could just mean that local sources have high OC/EC ratios. Other things like correlations with tracers and changes in time or correlations with wind direction, wind speed, or air mass back trajectories could be useful. Could the concentration of OC be controlling the trend in the ratio of OC/EC, since there is little variation in the total concentration of EC throughout the year (0.3 μg/m3 for EC and more than 1 μg/m3 change for OC)? It would be useful to have a table with literature values of OC/EC for fossil fuel combustion, biomass burning, aged biomass burning, etc. for comparison to the results shown here. The authors could also show a map of forest fires or discuss anthropogenic fossil fuel burning sources in specific regions where the air masses originate.

More explanation is needed for the tracers and their sources. For example, why is WSOC a tracer for SOA? It is stated that as the organics are oxidized, they become more water soluble, but that does not necessarily make WSOC directly linked to SOA. There is plenty of primary OA that is water soluble. This argument needs to be more clearly laid out with references and explanations for other possibilities. The aging of organic aerosols and SOA are two different things, which are both represented by WSOC in this paper. Additionally, the attribution of low WSOC to OC ratios to primary marine OC is not justified (Lines 273 – 277). This is only one sentence with no discussion of emissions correlated to wind speed or OC correlated with any sea salt tracers. This should be removed or discussed in more detail.

The discussion of the time series in Figure 4 is confusing. It is unclear why an increase

in OC/EC and OC/TC (which are not shown) suggest an “enhanced formation of SOA” over the measurement time. An increase in OC/EC may suggest different sources, but not all OC is SOA. This needs a clearer explanation. Additionally, it may be clearer if the data was binned by year and/or just the first and last years were shown. With all of the variability in the time series, even the trends that are statistically significant do not appear to be actual trends.

OC is not purely “scattering” or “cooling” as suggested here. Brown carbon is OC that can absorb solar radiation. More explanation is needed on how the ratio of OC/EC can be used to understand the relative contributions of scattering and absorbing of aerosols. Additionally, at line 341, there is no mention of radiative forcing by other types of aerosols. The ratio of OC/EC only applies to the organic fractions. And, the size of the particles plays a role as well as their morphology and composition. This needs to be included in the discussion.

Along these lines, the “Atmospheric implications” section is far reaching for the data shown and needs to be revised to reflect the actual measurements and analyses. The correlation between WSOC and CCN is very short and not fully explained. There needs to be much more discussion, if that is included. Figure 5 just shows a correlation between total particles activated (CCN) and the organic concentration. There is no description of initial particle sizes or compositions. It is possible that WSOC also correlates with NaCl in the particles, which are driving the CCN activity. No direct link can be shown with this correlation alone.

Specific Comments. Lines 56-49: References are necessary for this statement. While it is true that OC may warm less than EC, it does not necessarily provide a “cooling effect” as written here. This is still a major topic of study and should be written to reflect that, along with the necessary references.

Line 68: Check this reference.

Line 71: The particles are what act as CCN. Change “act” to “aid in particles acting”

Line 85: Contribution to what?

Line 92: Some general statements are made about increasing trends over time periods throughout the paper. These need to be rewritten or better explained. For example, at Line 92, it states “an increasing trend of biogenic emissions in northern China during 1982-2010.” It is unclear from this statement if the emissions were simply higher in 2010 than 1982 or if emissions increased every year during that period.

Line 98: Is marine aerosol the main focus of this paper? It needs to be defined here. Just sampling at a remote island does not necessarily mean that the sampled air masses are marine in origin and can include polluted air masses, as the authors discuss.

Figure 1: Why does this figure show the concentration of chlorophyll? Why is the winter 2008 chlorophyll map shown? That is not representative of the summer season. Is it representative of all of the years included in the study? That seems unnecessary to include. Also, the color bar is too small.

Line 133: How good of an assumption is it to assume that the carbonate carbon is insignificant. The IMPROVE network may be observing different sources and particle types that those in this study. Provide a reference to show that carbonate carbon is expected to be low in this region or with these sources.

Line 154: The whole section on statistical analysis should be condensed or moved to the supplement. Also, at line 163, this equation is unnecessary to list here.

Figure 2: The text says 2001 to 2013, and the caption says 2001 to 2012. And an average over 11 years is not that interesting without some sort of error bars or something to show the lack of change with time. If this is the general trend during these four times of year, that is fine, but it seems unnecessary to show in a figure then. Additionally, there is no label on the color bar. The main point of this figure is to show that in the winter the air masses measured at the sampling site are from East Asia while the weaker winds in

the summer transport air masses from the central Pacific. These figures and the wind show the general patterns, but it would be more relevant for this study to show the actual air mass backtrajectories for the samples. The wind pattern does not show origin or destination but rather a general pattern. Additionally, the arrows in many regions are too small to actually determine their direction, and there is no scale for their size.

Line 205: This is not new. Change "we found" to "there is" to be consistent. Figure 3: What are the small open, grey boxes (means?) and the X grey markers (outliers?)? So the EC is pollution and the OC is not?

Line 268: Needs a reference.

Lines 283-284: Panels d and e show the opposite trend.

Line 289: Why are OC/EC and OC/TC not shown in Figure 4, especially since their trends are significant?

Lines 292-293: How do these results show that "the contribution of combustion-derived sources to enhanced SOA seems to be decreased significantly"? This sentence is unclear. Is it possible that the EC concentration did not change?

Lines 297-300: Biomass burning is combustion. This sentence should be clarified if combustion is supposed to be anthropogenic or fossil fuel burning only.

Lines 301-303: This conclusion is unwarranted for the results shown.

Line 305: Need more references.

Line 307: There is also a significant decrease in WSOC/OC that is not discussed here. What is that cause of that decrease?

Line 310: What is the evidence that this is photochemical oxidation? These conclusions are all overreaching the data that is presented.

Line 317: MSA can also be used as a tracer for ocean biogenic emissions. Is this

referring to continental or oceanic biogenic emissions? Also, the trend line shows a slight increase, but the figure is not that convincing.

Line 323: This is a correlation between land temperature and isoprene emitted from land. It is unclear how much on an influence this would have on the data presented. This needs more explanation and some analysis if it is included.

Line 329: Where is this data? It could be included in the supplement, if it is useful.

Line 330: An value of r = 0.40 is not a significant correlation.

Line 336: OC can also absorb solar radiation (i.e. brown carbon).

Technical Comments. This paper should be read through and edited for grammar. Some examples of necessary edits are below. The authors should be consistent with terminology (i.e. organic vs. carbonaceous, etc.) and write out all acronyms the first time they are used (i.e. MEGAN, MOHYCAN, etc.)

Line 62: Remove “ever”

Line 80: Awkward phrasing – “because of knowledge gap on”

Line 82: Change “worst” to “worse”

Line 91: Remove “that”

Line 104: Remove “from the” and “away”

Lines 156-157: This sentence is a fragment.

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