

Interactive comment on “Size-resolved Composition and Morphology of Particulate Matter During the Southwest Monsoon in Metro Manila, Philippines” by Melliza Templonuevo Cruz et al.

Anonymous Referee #1

Received and published: 11 May 2019

The authors of this manuscript present a set of size-resolved aerosol measurements made in an urban location in Metro Manila, Philippines during the southwestern monsoon (SWM). Samples were analyzed using a variety of methods to produce average values for both gravimetric and speciated concentrations of particulate matter in twelve, roughly two-day time periods. Results are first compared with general meteorological conditions and transport pathways using a local weather station and the HYSPLIT backtrajectory model. The results were then classified into identified source types influencing the results using PMF.

The study represents a valuable addition to classification of aerosol type and sources

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impacting metro Manila during the SWM, in particular through the addition of size-resolved results. The authors' methodology was generally sound and produced findings that are generally consistent with reports of similar studies in other major urban areas. However, several of their primary findings and discussion were not fully supported by the results as presented. In addition, a more thorough description of certain aspects of their experimental setup and analysis are needed to fully understand their work. As a result, I recommend publishing of the manuscript after consideration of several major comments.

Major Comments: 1. The authors present a single short duration period with one measurement of size resolved black carbon aerosol in their dataset. While this is an interesting and useful finding, they over emphasize the extent to which they can claim general representativeness of this result on the wider Manilla urban aerosol environment during the SWM. If there is a different interpretation of their results, this needs to be clarified, as it is not apparent in their results as presented. Future work with additional such measurements would be worthwhile and support the current findings, but they should take additional care to not attribute the current BC results to wider claims or conclusions.

2. The authors conclude that the most common source type identified by PMF analysis, classified as "Aged/Transported aerosol," is evidence of the influence of a major non-local source. In the conclusions they further state that "the significant presence of Aged/Transported aerosols in Metro Manila indicates that PM in the region has the ability to travel long distances during the SWM season, despite the typical assumption that wet scavenging effectively removes most of the particles." Non-local sources dominating the Manila aerosol for considerable periods of time (the authors state this factor constituted 48% of PMF classified sources by mass) would indeed constitute a considerable change in the understanding of the sources of particulate matter in Manilla, but this conclusion is not supported by the analysis presented here.

Given the results as presented, it would seem this identified PMF type could just as

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easily be attributed to aged aerosol that includes mixing from various local and regional sources. At the very least the authors need to consider other possible interpretations of this PMF source. Is it possible this source is merely the result of mixing from local and regional sources in circumstances where the aerosol has not been impacted by recent precipitation, and therefore has aged more than other sources?

Several open questions need to be answered before this PMF type can be attributed to a non-local source.

- What constitutes a “transported” or non-local aerosol? Smoke transported from biomass burning regions in Borneo or Sumatra that have been transported thousands of kilometers would constitute a considerably different source from an anthropogenic source hundreds of km away in other parts of the Philippines, or from regional sources in cities neighboring Metro Manila.

- Address if markers for various sources constitute actual evidence of such sources dominating the PMF type and observed aerosol, or if they merely constitute mixing of various sources with local aerosol. Do the sources associated with ocean emissions (the authors note MSA and DMA), biomass burning (K is noted), or oxalate production in fact dominate this PMF type? That would not appear to be the case in the mass loadings from Figure 8. Further, NH_4^+ and SO_4^{2-} are observed in other urban areas, why should they constitute evidence of non-local transport here?

- Link the source with precipitation, transport pathways, and aging times. Could precipitation scrubbing or the lack thereof account for the differences between this source and others? The authors note that the source may require aging for production of various measured species, but do not account for what amount of time might be required or if that could be accounted for by local or regional sources, complex meteorology and transport pathways in the region, or differences in precipitation scrubbing of the airmass.

- Account for the high overall mass loading being transported from a non-local source.

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A consistent non-local accumulation mode aerosol plume with sufficient mass loading to account for the measured mass concentrations should be noted in other measurements (e.g. AOD).

- Validate that the aerosol is not merely aged and associated with complex mixing of local and regional sources already expected to be major sources of aerosol in the region (as correctly noted by the authors).

Classifying this aerosol type as aged aerosol would seem justified by the analysis in section 3.3.1, but it does not necessarily follow that it is therefore a non-local or non-regional source. As this is implied to be a major finding of this work, the authors may wish to revisit this analysis and potentially reframe this source as an aged aerosol with some evidence of mixing with non-local sources.

Minor comments:

1. Experimental setup and site information. Additional information is needed regarding the setup of the measurement system and weather station. In particular, the location and description of the sample inlet and any initial processing of the sample that occurred. The height of the weather station above ground level in addition to the sea level reference, and its location is needed as well (e.g. Is the third floor near the top of the building? Was the station located on the roof? Were any potential sources of bias considered such as wind steering around buildings considered?) From Figure 1 it appears the sampling location is some distance from the nearest major roadway. Has previous work using this sampling location indicated it is generally representative of the local aerosol?

2. Usage of HYSPLIT. Were there any other HYSPLIT receptor heights considered. Vertical wind shear can be considerable in the maritime continent, and can alter transport pathways considerably. Table 1 indicates primarily easterly winds. Was there any comparison between backtrajectories and local wind measurements? Were expected transport pathways or HYSPLIT trajectories related in any way to sample results? Local

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measurements of wind direction, which are more indicative of local transport pathways, were correlated to mass concentrations, but not backtrajectories that are more indicative of long range transport pathways. Does this relate in any way to expected source type or location?

3. Description of data collection and usage in various analyses. The experimental methods section 2 would benefit from more clarity regarding how many data points were utilized in the various analyses, and which measurements they came from. It appears that there were 12 sampling periods during which data was collected for PMF analysis, which each consisted of 11 size bins. How was the data prepared for use in the PMF analysis?

Specific comments and technical corrections:

26. Need to specify that particle size is defined on a diameter basis. Also would help to briefly describe the complete range of sizes measurements were made across before describing results in specific size ranges.

28. The authors need to clarify that the Greenfield gap term is in reference to the reduced efficiency of precipitation scavenging of accumulation mode aerosol particles as compared to particles of other sizes. The wording of this sentence seems to imply that either accumulation mode particles exist because of the lack of precipitation scavenging, or that the Greenfield gap describes accumulation mode particles. At the very least “(the so-called Greenfield gap)” should be moved to the end of the sentence.

105/113: Manila Observatory is defined after the first use of MO as an acronym.

150: Additional detail about the typical duration and strategy for collection of sample sets would be helpful in this paragraph.

156: State the specific height of the HYSPLIT backtrajectory receptor.

Section 2.2: Again, information about the inlet and sample collection strategy is needed. Is there a general methodology that was followed for collection of the aerosol

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that has been reported elsewhere? If so, this should be cited. If not, more information is needed (e.g. was rotation utilized, was sample RH monitored or expected to affect samples, were there any other expected sources of potential bias in the measurements)? More information or methodology references are also needed to fully describe methodologies for the gravimetric and optical absorption analyses.

Sections 2.3 and 2.4: Methodologies described in these section are somewhat lacking in references. The authors may want to consider including additional references with more description of both the methods and any caveats to their use for interested readers.

Section 2.5: Which measurements specifically were included in the PMF and correlation analyses, and how was the size distribution of various species included in these analyses? Also specify how many data points were used for these analyses.

334: May be useful to remind readers that jeepneys are local vehicles in common usage in Manila.

349: Need to consider the relatively few BC measurements in this study and moderate speculation about the representativeness of this single measurement unless otherwise justified.

354: The authors seem to be claiming that most BC was measured in the accumulation mode primarily as a result of the lack of precipitation scrubbing in the Greenfield gap (“95% of the BC mass is concentrated in the Greenfield gap, and thus the removal of BC due to precipitation is inefficient”). While the claim that BC is not being efficiently removed by precipitation makes conceptual sense, the emphasis on this arising due to precipitation processes is not supported by the arguments presented here. Precipitation scavenging is not the only process that affects the size distribution of particles. Coagulation, growth, and aging tend to move particles from nucleation and Aitken modes into the accumulation mode, while smaller particles typically already have minimal contributions to mass distributions. Further, there were not enough measurements of BC

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size distributions (only one it would seem, sample MO13) to compare relative differences between distributions in periods of more and less precipitation. In this sentence specifically, and section 3.2.1 more generally, the authors make a number of claims about the size distribution of BC that appears to be based on only one sample. While the authors may comment on the nature of the BC mass distribution data point, they should refrain from undue speculation on its cause in this case, unless it is merely to mention potential relevant processes.

445: The sentence beginning on this line is not completely clear, and the authors may wish to reword to better clarify their intent. Additional explanation of why they consider the PMF solution valid may be helpful.

447: I assume the authors meant to refer to the coefficient of determination (i.e. r^2) rather than the coefficient of variation (i.e. σ/μ) here.

467: I do not understand how the authors came to this conclusion based on one measured BC data point. What is the interrelationship with water soluble ions that justifies concluding they necessarily vary in concert with each other? How do you correlate BC to 15 other species? Is this just based on correlation of the size distribution? Many species exhibit a similar distribution in the accumulation mode, so that alone would not seem to justify this statement.

508: In this paragraph the authors state they expect PM in Metro Manila to be dominated by local sources of aerosol, and that the “Aged/Transported” pollution PMF type is in fact the largest source. Is it not reasonable to associate this source with precisely what was expected for a typical background aerosol in Manila that features complex mixing between local, regional, and distant sources that have experienced some aging before being either advected to other areas or impacted by precipitation? If mixing with distant sources is in fact detectable, that does not necessarily imply that local sources are not still the dominant source.

560: How are BC correlations being conducted? Following on the earlier comment

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regarding valid data points for various analyses, how many data points and of what type are used in this correlation? Is this intended to highlight BC found in the same size bins as As and Ni?

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2019-270>, 2019.

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