Interactive comment on "Long-Range Transport Mechanisms in East and Southeast Asia and Impacts on Size-Resolved Aerosol Composition: Contrasting High and Low Aerosol Loading Events" *by* Rachel A. Braun et al.

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We thank the reviewers for thoughtful suggestions and constructive criticism that have helped us improve our manuscript. Below, we provide responses to comments from both referees.

Anonymous Referee #1

Through ground-based observations, the authors studied physical and chemical properties of aerosol particles for 14 selected cases over Metro Manila, Philippines. This

kind of ground-based data analysis is welcomed by both remote sensing folks as well as modelers for CTMs. The paper is well written. The data analysis part of the paper seems reasonable, as I am not a chemist. Thus, I rely on other reviewers who have a background in chemistry to fully evaluate the chemistry portion of the study. Still, I have a few concerns that I hope the authors can address.

First of all, for the methodology section, more details are needed. For example, the detection limits of ground-based observations are included in the supplement. Still, the authors need to mention uncertainties of other data and models used in the study. For example, precipitation amounts were obtained from PERSIANN-CCS data, but what is the uncertainty of the dataset? Also, details for some datasets such as CALIOP Level 2 VFM (version of the dataset, spatial resolution etc.) need to be provided for the benefit of the readers.

Response: While we are unable to provide specific numbers for the uncertainties of some of datasets, we have added the following text to clarify:

"Benefits of PERSIANN-CCS include the data availability at 0.04° x 0.04° spatial resolution, while uncertainties in the dataset arise from sources such as a lack of bias correction (Nguyen et al., 2014)."

"The CALIOP Level 2 Vertical Feature Mask (VFM) Version 4.20 was used to distinguish between clear air, clouds, and aerosol (Vaughan et al., 2004). For figures of CALIOP VFM data in this study, data are plotted at 30 m vertical resolution every 5 km along the satellite ground-track."

The temporal sampling window is different for different cases. While the temporal sampling window is approximately 2-day for most cases, the temporal sampling window is 5-day for MO4 and 1-day for MO1. Does the difference in temporal sampling window introduce a temporal-sampling related bias?

Response: The initial sets were taken at the very beginning of a year-long sampling

campaign. As these measurements were new to the metro Manila environment, we were unclear of how long to run the instrumentation for optimal sampling. Therefore, the variations in sample lengths represent different tests until we settled on a 2-day sampling window. However, for all sets, we attempted to sample in increments of 24 hrs in order to minimize diurnal influences. Furthermore, the length of sampling factors into the conversion of the aqueous concentrations to air-equivalent measurements. Therefore, we believe any differences in measurements due to varying sampling windows are minimized. We do not feel as though text revisions are needed in the draft to address this comment.

I understood that satellite aerosol retrievals have difficulties over the study region due to cloud coverage. But it is still useful to provide an aerosol optical depth (AOD) climatology for the study period (July-Oct. 2018) as well as the spatial distribution of AODs for the selected cases (e.g. MO7, 11, 12 and 14) using passive-based satellite data such as MODIS or MISR. Such an analysis will assist their modeling-based analysis (e.g. from NAAPS). This might also help the authors link their case studies with the aerosol climatology of the region.

Response: We have purposefully chosen not to include AOD from passive remote-sensors for the following reasons. As shown in Figure 1, very high average cloud fractions are found in all of the sample sets. Therefore, any AOD measurements from passive remote-sensors will either not have data available or have data with significant clear-sky bias. As an example, we have included plots for just the select case of MO7. As is seen for both the MODIS Terra and MODIS Aqua results, there are significant gaps in the data availability. Furthermore, one purpose of the paper is to demonstrate the variability in aerosol transport pathways. Therefore, any climatology of AOD in the region from July-October will remove these nuances and be subjected to the same clear-sky biasing as described above.

Speaking of which, I think the linkage between data analysis presented in this study and broader scientific questions is still plausible. For example, how representative

are those selected 12 events to different aerosol transport scenarios or to the general aerosol climatology of the region? What are the linkages between the data analysis presented in the study and some broader scientific questions? I hope the authors can add more related discussions.

Response: The 12 events in the study represent the start of a campaign of weekly measurements at the study site that occurred for a full year. Here, we are presenting just the results from the first sets that occurred during the Southwest Monsoon season, into the transition to the Northeast Monsoon. During this time frame, we have identified a few high aerosol events and one low aerosol event, with the remaining cases forming what we are considering the "background conditions." As shown in the HYSPLIT for these cases in Figure S1, the predominant flow pattern is from the southwest. However, since these are only from one season of measurements and there is a lack of size-resolved aerosol measurements in the region, it is difficult to say with certainty whether this represents the general aerosol climatology for the time frame. Our objective is to highlight potential transport pathways for which we believe we can observe signals in Metro Manila.

In terms of broader scientific questions, we have amended that end of the conclusion to highlight some more relevant discussions and questions that can be addressed in future research:

"These results have important implications for better understanding the aerosol budget and influences in and around the Philippines and SE Asia. Transport of aerosol both into and out of Metro Manila can impact human health, cloud condensation nuclei (CCN) budgets, and radiative forcing in the region. Furthermore, the identification of various tracer species (e.g. K and Rb for biomass burning) and the impacts of different long-range transport mechanisms have worldwide applications. In addition, the mixing of different air mass types, resulting in changes in aerosol characteristics (e.g. enhanced oxalate in emissions from continental regions, enhanced MSA during periods of biomass burning influence), is a subject that requires more attention on a global ba-

sis. While this work has shown the influence of mixing biomass burning emissions and urban emissions, from both local and more distant urban centers, additional analysis at the study site has demonstrated the influences seen from the mixing of sea salt aerosol with other airmasses (AzadiAghdam et al., 2019). As remote-sensing measurements in this region are notoriously difficult (e.g. Reid et al., 2009, 2013), in situ and model results lend vital data to address the questions surrounding characteristics of aerosol that are transported into and out of this highly-populated region. Measurements from in situ airborne campaigns, such as CAMP2Ex, can further address the changes in aerosol physicochemical characteristics that occur during long-range transport and aging in the atmosphere in the region."

AzadiAghdam, M., Braun, R. A., Edwards, E.-L., Bañaga, P. A., Cruz, M. T., Betito, G., Cambaliza, M. O., Dadashazar, H., Lorenzo, G. R., Ma, L., MacDonald, A. B., Nguyen, P., Simpas, J. B., Stahl, C., and Sorooshian, A.: On the nature of sea salt aerosol at a coastal megacity: Insights from Manila, Philippines in Southeast Asia, Atmospheric Environment, 216, 116922, https://doi.org/10.1016/j.atmosenv.2019.116922, 2019.

Anonymous Referee #3 The authors presented their efforts in analyzing local observations collected at the MOUDI sample site, and tried to attribute the observed changes of water-soluble aerosol to long-range transport from MC and north East Asia. This is an interest and innovative research as the air quality in Philippines hasn't been well documented in the published studies. The observations collected through this study also provided detailed description of the aerosol size distribution, chemical composition, and temporal changes. The coupling of satellite product, surface observations, HYSPLIT trajectories, and NAAPS simulations is acceptable, but there are two major concerns regarding this method:

First, the sampling study period is relatively too narrow to justify the description of "high" and "low" aerosol loading periods, as the concentrate ranges from 2.7 to 13.7 ug/m3, I didn't see there is significant difference especially considering the wash-out effect of precipitation during the "low" loading period. It will be better if the authors can

present some data or cite from other studies to briefly describe the year-long trend of aerosol concentration at MOUDI site.

Response: We are not attempting to classify high and low loading events for a full annual basis; rather, our description of the "high" vs. "low" aerosol loading events is within the context of the sampling time frame (July – October 2018), as stated in the methodology: "For each MOUDI set (naming convention: MO#), the mass concentration sum of the water-soluble species was calculated; using this summation, the three high-aerosol loading events were identified (MO7, MO12, and MO14), as well as the lowest aerosol event (MO11)." Furthermore, we are only looking at the water-soluble portion of the aerosol, not the total mass, so the overall concentrations we are working with will be smaller. We have added to the text the following line to highlight the departure from the mean for the identified case studies within our sampling time frame:

"The average $\pm$ standard deviation of the total-water soluble species measured for the remaining 8 sets not identified in the high or low categories is 6.99 $\pm$ 2.71 μg m-3."

We have also added the following text to the introduction:

"Previous research conducted at the Manila Observatory (MO) in Quezon City, Metro Manila characterized PM2.5 (particulate matter (PM) with aerodynamic diameter less than 2.5 μm) and sources of measured particles, with traffic emissions being the major source at MO (Simpas et al., 2014). Interestingly, levels of measured PM2.5 at MO showed little variance between the wet (June-October) and dry seasons (Simpas et al., 2014)."

Second, the title indicates the long-range transport mechanism will be discussed, but in the manuscript really only describes the influence of long-range transport in Quezon City. Please consider rephrase the title or include more discussion of the transport mechanism.

Response: While our in situ ground-based measurements were taken in Quezon City,

we believe that other datasets and discussions included in the manuscript broaden the scope of the transport beyond just Quezon City. For example, we include NAAPS model results at the larger scale and the discussions in Section 3.1 include descriptions of smoke transport from the maritime continent and transport of emissions from continental Asia during the typhoon.

Following are some detailed comments. Detailed comments: (1). The "abstract" section was poorly organized, and it contained too many details about method and dataset while the innovative findings and conclusions were described in a style too general for scientific publication. It begins with a very clear statement of the research objective, as "analyzes mechanisms of long-range transport and chemical characteristics", but is followed by various types of information piece by piece. For example, line#28-29, what is the long-range transport mechanism found through this study? Is it driven by synoptic weather event, or large scale jet, or other typical or abnormal conditions? Line#30-31, the impacts of continental EA transport was identified, so what are they? Please reorganize the whole section.

Response: We have attempted to address the specific points mentioned by the reviewer. However, we have tried to write the abstract in as straightforward a manner as possible, so we are unclear of the best method to reorganize the current form of the abstract. The revised abstract is provided here:

"This study analyzes mechanisms of long-range transport of aerosol and aerosol chemical characteristics in and around East and Southeast Asia. Ground-based size-resolved aerosol measurements collected at the Manila Observatory in Metro Manila, Philippines from July - October 2018 were used to identify and contrast high and low aerosol loading events. Multiple data sources, including models, remote-sensing, and in situ measurements, are used to analyze the impacts of long-range aerosol transport on Metro Manila and the conditions at the local and synoptic scales facilitating this transport. Through the use of case studies, evidence of long-range transport of biomass burning aerosol and continental emissions is identified in Metro Manila. Long-

range transport of biomass burning aerosol from the Maritime Continent, bolstered by southwesterly flow and permitted by low rainfall, was identified through model results and the presence of biomass burning tracers (e.g. K, Rb) in the ground-based measurements. The impacts of emissions transported from continental East Asia on the aerosol characteristics in Metro Manila are also identified; for one of the events analyzed, this transport was facilitated by the nearby passage of a typhoon. Changes in the aerosol size distributions, water-soluble chemical composition, and contributions of various organic aerosol species to the total water-soluble organic aerosol were examined for the different cases. The events impacted by biomass burning transport had the overall highest concentration of water-soluble organic acids, while the events impacted by long-range transport from continental East Asia, showed high percent contributions from shorter chain dicarboxylic acids (i.e. oxalate) that are often representative of photochemical and aqueous processing in the atmosphere. The low aerosol loading event was subject to a larger precipitation accumulation than the high aerosol events, indicative of wet scavenging as an aerosol sink in the study region. This low aerosol event was characterized by a larger relative contribution from supermicrometer aerosols and had a higher percent contribution from longer-chain dicarboxylic acids (i.e. maleate) to the water-soluble organic aerosol fraction, indicating the importance of both primary aerosol emissions and local emissions. Results of this study have implications for better understanding of the transport and chemical characteristics of aerosol in a highly-populated region that has thus far been difficult to measure through remote-sensing methods. Furthermore, findings associated with the effects of air mass mixing on aerosol physiochemical properties are applicable to other global regions impacted by both natural and anthropogenic sources."

(2) Line#58-60: this sentence is confusing, do you want to distinguish long-range transport from synoptic scale transport?

Response: In this sentence, we try to highlight that long-range aerosol transport occurs all over the world, yet the conditions and types of aerosols vary by region due to various

factors, including regional conditions. We have re-written the sentence as follows:

"Although impacts and processes of long-range aerosol transport have worldwide applicability, the variety of meteorological conditions and emission sources that can contribute to aerosol transport necessitate detailed analyses of transport events at the regional level."

(3) Line#61: there are many these kind of general common sense statements that are not really helpful in this manuscript, please consider exclude them.

Response: We have re-written lines 61-62 to address this comment and the following comment #4:

"The plethora of both natural and anthropogenic emissions in and around the Southeast (SE) Asia, the proximity of islands and continental regions in SE and East Asia, and the large, growing population makes SE Asia a prime candidate for the study of long-range transport of atmospheric aerosol."

(4) Line#62: I didn't see any reason for starting with "however"

Response: See response above to comment #3.

(5) Line#70: "urban mega-cities" emission? Do you mean residential emission?

Response: While residential emissions are one factor in the emissions from urban mega-cities, as stated in Reid et al. (2013), differences in emissions amongst cities arise from a variety of factors, such as traffic, cooking, heating, industrial activity, etc.

(6) Line#82-86: These sentences commented the satellite-derived biomass burning emission inventory was underestimated, so the readers would expect to see further discussion about the underestimation, or how to improve it. But line#87-94 started to claim that transport mechanism of biomass burning is important. I didn't see any logistic connection between these two sections. As the importance of this study is described in line#87-94, line#82-86 is not helpful to demonstrate this importance.

Response: We believe that this description is applicable in that ground-based measurements and verifications of biomass burning can help explain and better account for the under-estimation, including both amount and frequency, of satellite-derived biomass burning emissions in the region. We have clarified this by amending the text as follows:

"In order to better understand the frequency, amount, and fate of biomass burning emissions in the MC and SE Asia, both in situ measurements and modeling studies are needed."

(7) Line#119: when is the northeast monsoon season?

Response: While variability exists in the start dates of the different seasons, Cruz et al. (2013) indicate that the transition to the northeast monsoon generally occurs in October. Bagtasa (2011) defined the northeast monsoon season from October – February. We have clarified this in the introduction by adding the following text:

"While variability exists in the start dates of the different seasons, the northeast monsoon transition generally occurs in October (Cruz et al., 2013), and previous research has defined this season as occurring from October – February (Bagtasa, 2011)."

Bagtasa, G.: Effect of Synoptic Scale Weather Disturbance to Philippine Transboundary Oxone Pollution using WRF-CHEM. Int. J. Environ. Sci. Dev., 2, 402-405, 10.7763/IJESD.2011.V2.159, 2011.

(8) Line#110-111: varies by season: smoke in Aug-Oct, dust in Feb-Apr, SWM for Jun-Sep, what about the other months? These introductions are important to justify your studying period Jul-Oct 2018 mentioned at line#138-139.

Response: The full sentence that the reviewer cites reads: "Another study of the aerosol over the South China Sea (SCS), which is bordered to the east by the Philippines, found seasonal changes in aerosol emission sources, with year-round anthropogenic pollution, smoke from the MC between August – October, and dust from northern continental Asia between February – April (Lin et al., 2007)." As stated, "year-round

anthropogenic pollution" was measured; therefore, we believe that the other months are already addressed. Furthermore, the other studies cited discuss additional sampling periods from previous studies, biomass burning during the SWM, and aerosol from East Asia during the northeast monsoon. Therefore, we do not feel as though text revisions are needed in the draft to address this comment.

(9) Line#139-150: I like the way that objectives and aims are clearly listed, please consider reorganize the manuscript in such a straightforward manner.

Response: Our organization of the manuscript attempts to follow the same order in which the objectives are listed in these lines. In the results section, we begin by discussing the atmospheric conditions for each case study (objective i), followed by discussions of the composition of the size-resolved aerosol samples (objective ii). Within this discussion, we have added descriptions of transformational processes that we observe, especially with regards to the organic aerosol (objective iii). Therefore, we believe that the organization of the manuscript already mirrors that of the objectives.

(10) Line#141: Confusing, please rephrase this sentence: isolate characteristic aerosol physiochemical properties indicative of long-range transport

Response: We have re-worded as follows:

"(ii) characterize aerosol physicochemical properties associated with long-range transport."

(11) Table1: please explain why the sampling period was 2-day, and why the starting time was different, some in the morning and some in the afternoon

Response: We received a very similar comment from Referee #1 and have listed our response to their comment here:

"The initial sets were taken at the very beginning of a year-long sampling campaign. As these measurements were new to the metro Manila environment, we were unclear of how long to run the instrumentation for optimal sampling. Therefore, the variations

in sample lengths represent different tests until we settled on a 2-day sampling window. However, for all sets, we attempted to sample in increments of 24 hrs in order to minimize diurnal influences. Furthermore, the length of sampling factors into the conversion of the aqueous concentrations to air-equivalent measurements. Therefore, we believe any differences in measurements due to varying sampling windows are minimized. We do not feel as though text revisions are needed in the draft to address this comment."

(12) Line#245: water-soluble aerosol refers to the species shown in Fig.6? are these measurements for ambient air concentrations?

Response: Our method for analyzing the chemical composition of the ambient aerosol was to extract the filter samples in water. Therefore, all concentrations described are for the water-soluble component of the aerosol only.

(13) Fig.2: why there are multiple blue lines for back-trajectories, did y ou trace back at different altitude?

Response: As stated in the methodology section, "The model was run for back-trajectories terminating at the MOUDI inlet (~85 m above sea level) every 6 h during each sample set." This results in multiple back-trajectories for each sample set, since each set was run for at least 24 hrs, with most run for 48 hrs. We have clarified this by amending the text as follows:

"The model was run for back-trajectories terminating at the MOUDI inlet (~85 m above sea level) starting at the beginning of the sample set and every 6 h thereafter during each sample set, resulting in (1 + N/6) trajectories for each set, where N is the total number of sampling hours."

(14) Line255-257: the NAAPS only shows the surface concentration, the MC smoke may not necessarily transport all the way to Quezon, this is why the altitude of HYSPLIT also need to be demonstrated to correlate the source and receptor

Response: As stated in the methodology section, "The model was run for back-trajectories terminating at the MOUDI inlet (~85 m above sea level)." Since we were trying to correlate our surface measurements with the transport patterns, we wanted to use only HYSPLIT back-trajectories that ended at our surface site altitude. While we ran the HYSPLIT model for this altitude only, we believe that this is representative of the region, due to a previous work that has shown the significance of boundary layer transport in the region. Furthermore, as shown in the CALIPSO overpasses, it appears that the aerosol layer extends from the surface.

(15) Section3.2.3: the concentration of organic aerosol seems very low, can you show the total ambient air concentration of OA, other than the water-soluble aerosol?

Response: Unfortunately, we do not have concentrations of the water insoluble components. However, as shown in the analysis by Cruz et al. (2019), black carbon measurements were conducted for one sample set and the filters were weighed for two other sets. The results show that there are significant fractions of black carbon and unresolved aerosol mass, which could be insoluble organic and inorganic components.
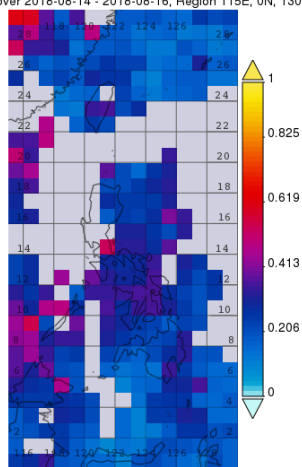
Fig. 1. MODIS-AQUA AOD for Sample Set MO7
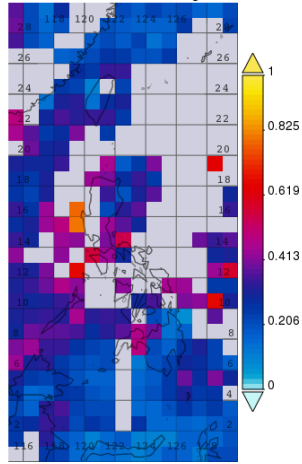
Fig. 2. MODIS-TERRA AOD for Sample Set MO7