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*** Corresponding Authors:**

Christian Mark G. Salvador
cmsgsalvador@pnri.dost.gov.ph
Preciosa Corazon B. Pabroa
pcbpbabroa@pnri.dost.gov.ph

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Variability and Source Characterization of Regional PM of Two Urban Areas Dominated by Biomass Burning and Anthropogenic Emission

Christian Mark G. Salvador ^{1*}, Jhon Robin dR. Yee²,
Iara Chantrelle V. Coronel², Angel T. Bautista VII², Raymond J. Sugcang²,
Mark Anthony M. Lavapie³, Rey Y. Capangpangan ⁴,
Preciosa Corazon B. Pabroa ^{2*}

¹ Balik Scientist Program, Department of Science and Technology - Philippine Council for Industry, Energy and Emerging Technology Research and Development, Bicutan, Taguig 1630, Philippines

² Department of Science and Technology (Philippines) - Philippine Nuclear Research Institute, Commonwealth Avenue, Diliman, Quezon City 1101, Philippines

³ Department of Chemistry, Caraga State University, Ampayon, Butuan City 8600, Philippines

⁴ Department of Physical Science and Mathematics, College of Science and Environment, Mindanao State University-Naawan, Naawan, Misamis Oriental 9023, Philippines

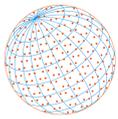
ABSTRACT

Identifying the sources and formation pathways of particulate matter (PM) and its components is important to determine the impact of atmospheric aerosols on human health and the warming of the global atmosphere. Here, we characterized the variability of the eleven-year concentration and the origin of PM_{2.5} in two urban regions (Metro Manila and Boracay) in the Philippines, a country in South East Asia (SEA) with high local anthropogenic emissions and frequently impacted by transboundary pollution. The surface concentration of PM_{2.5} was analyzed using simulated and reanalyzed satellite data (MERRAero), complemented, and validated with filter measurements for PM_{2.5} and black carbon (BC). The PM_{2.5} masses in Metro Manila ($12.3 \pm 2.4 \mu\text{g m}^{-3}$) and Boracay ($12.9 \pm 3.6 \mu\text{g m}^{-3}$) were comparable yet with varying concentrations of aerosol components. Anthropogenic related components of PM (organic carbon, sulfate, and BC) were more enhanced in the metropolis region ($4.9 \mu\text{g m}^{-3}$) than in the prime tourist spot ($2.72 \mu\text{g m}^{-3}$). Sulfate formation impacted the aerosol mass in Metro Manila, with the photochemical oxidation of SO₂ as the predominant production pathway of sulfates. Analysis of regional organic matter (OM) and BC in Boracay revealed extensive transboundary transport of biomass burning (BB) plumes from equatorial SEA countries. Also, the ratio of OM and SO₄ was utilized as a quick proxy for source characterization. Metro Manila and Boracay reported 0.77 ± 0.20 and 1.06 ± 0.55 , highlighting the evident impact of traffic emissions in the PM mass in the metropolis region. For Boracay, the mixed contributions of BB and vehicular activities influenced the formation of PM. Overall, the reanalysis of satellite data captured the long-term variability and origins of surface PM in two vital urban regions in the Philippines. The utilization of MERRAero will be an important procedure in making regulatory decisions on regional pollution control.

Keywords: Surface PM_{2.5}, MERRAero, Long term trends, South East Asia, Biomass burning

1 INTRODUCTION

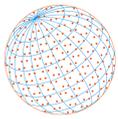
Atmospheric aerosols deteriorate air quality and influence human health at a varying degree depending on the size and composition (Hamanaka and Mutlu, 2018; Kim *et al.*, 2015). Heavy loadings of particulate matter (PM) can extensively impact the visibility and radiation fluxes, posing an environmental problem particularly in highly urbanized regions (Han *et al.*, 2012). An impact



analysis of air pollutants indicated that the reduction of PM_{2.5} (particles with diameter less than 2.5 μm) by 4 μg m⁻³ will prevent close to 8000 heart failure incidents, which translates to a third of a billion US dollars fewer expenses every year (Shah *et al.*, 2013). Unlike other typical atmospheric pollutants (e.g., CO), airborne PM poses multiple health hazards due to its heterogeneous components that can cause unique sickness depending on their concentration and distribution in the particle phase. The typical composition of atmospheric aerosols includes black carbon (BC), organic matter (OM), sulfates, and nitrates, which have different sources and formation pathways (Hallquist *et al.*, 2016; Kalisa *et al.*, 2019; Seinfeld and Pandis, 2016). Understanding the variability of the mass concentration of PM and its components is a crucial insight for the treatment and prevention of several respiratory and cardiovascular diseases related to exposure to PM. This is also vital in developing environmental policies for the long-term reduction of the emission of atmospheric aerosols.

Two of the most prominent PM sources are anthropogenic and biomass burning (BB) activities. Human-related endeavors primarily include the transportation sector (i.e., vehicle exhausts) and industrial emissions that significantly impact the PM mass concentration and composition in highly urbanized regions (Duarte and Duarte, 2020). Traffic emissions heavily influenced several developed regions across the globe (Martínez-Cinco *et al.*, 2016; Pabroa *et al.*, 2011; Sowlat *et al.*, 2016; Tolis *et al.*, 2014) through the emission primary PM. Vehicular activities also emit volatile organic compounds (e.g., benzene, toluene, trimethylbenzene, and xylene) and NO_x that serves as precursors for aerosol nucleation/formation events (Guo *et al.*, 2020; Tsiligiannis *et al.*, 2019). On the other hand, biomass burning events, which include domestic cooking, agricultural and residue burning, and forest fires, drastically contribute to PM mass observed at local, regional, and global scales (Chen *et al.*, 2017). During the combustion process, a suite of organic compounds partitioned in both the gas and particle phase are also emitted that can modify both the physical (e.g., size) and chemical properties (e.g., hygroscopicity and radiation absorption) of atmospheric aerosols (Kong *et al.*, 2021; Salvador *et al.*, 2021; Vidovic *et al.*, 2020; Xie *et al.*, 2019). Biomass burning events, particularly the large-scale combustion processes, are typically characterized by seasonal variation, which sometimes follow the trends of meteorology (e.g., humidity and temperature) and agricultural practices (e.g., burning of residues). Distinguishing the dominant source of PM at certain period, whether anthropogenic or biomass burning, is necessary to implement the proper environmental policies targeted on curbing the immediate enhancements of the PM. Typically, particle-phase tracers such as levoglucosan (Simoneit *et al.*, 1999; Zhang *et al.*, 2012) and di-acids such as phthalic acid (He *et al.*, 2018; Salvador *et al.*, 2016) are used to differentiate pyrolysis events or the urban emissions from traffic and industries. A readily available metric that can immediately provide insights into prevailing emissions should be developed and utilized, particularly at a regional scale.

In this work, we explore the eleven-year profile of the regional PM and the major aerosol components (BC, OC, SO₄) of two important tropical areas in the Philippines using simulated ground concentration output from the Modern-Era Retrospective Analysis for Research and Applications Aerosol Reanalysis (MERRAero) developed by the Global Modeling Assimilation Office (GMAO). MERRAero provides an estimate of the surface level PM_{2.5} through constraining Aerosol Optical Depth (AOD) and Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) aerosol module (Buchard *et al.*, 2016). The applicability of MERRAero in providing PM_{2.5} concentrations was previously validated over India (Mahesh *et al.*, 2019; Navinya *et al.*, 2020), Europe (Provençal *et al.*, 2017a), Israel and Taiwan (Provençal *et al.*, 2017b), southern Philippines (Salvador *et al.*, 2022), and United States (Buchard *et al.*, 2016; Saunders and Waugh, 2015). The ratio of sulfate (SO₄²⁻) and OM in the atmospheric aerosols estimate the relative contribution of anthropogenic and biomass burning sources to account for the high PM mass events (Saunders and Waugh, 2015). An evident increase in OM corresponds to regional combustion processes while industrial sources induce the enhancement of sulfate in the particle phase. A value of more than 1.0 corresponded to wildfire emissions while ratios below 0.7 were typically observed in areas with strong anthropogenic impact. Here, we assess the variation of the OM–SO₄ ratio in the two regions, particularly during episodes of strong influence of transported biomass burning plumes. This was complemented with our integrated filter-based black carbon speciation analysis that can efficiently separate biomass burning and vehicular emissions, thus validating the proxy ratio from MERRAero that can immediately provide source apportionment information.



2 METHODOLOGY

2.1 Site Description of Metro Manila and Boracay

Metro Manila and Boracay, two of the most known urban regions in the Philippines, are the focus areas of this study (see Fig. 1). Metro Manila (14.6091°N, 121.0223°E) is the country's national capital region (NCR), which is comprised of 16 cities. The total area of NCR is 619.57 sq. kilometers with a dense population of 13,484,462 in 2020. Heavy traffic is one of the urban dilemmas of Metro Manila due to increasing vehicle volume in the region. In 2015, the traffic analysis zone indicated that Metro Manila had a population density above 20,000 persons km⁻² which caused the average travel time per person of 1.17 hours in 2014 and will escalate up to 1.33 hours in 2030 (Rith *et al.*, 2020). Gaseous emission of CO₂ from the transportation sector in Metro Manila was estimated to be 13.78 million tonnes and is expected to increase to 27.90 tonnes in 2040 (Ahanchian and Biona, 2014). Between 2010 and 2020, the average surface temperature and wind speed were 26.2 ± 1.3°C and 4.8 ± 1.1 m s⁻¹, respectively.

Boracay (11.9674°N, 121.9248°E) is a small island in the mid-Philippines and a famous tourist destination of both local and foreign citizens. In 2015, the number of tourists increased to 1.5 million, contributing to unprecedented anthropogenic activities in this area (Limates *et al.*, 2016). It has a total area of 10.32 sq. kilometers, with significantly fewer residents than Metro Manila (32,267 in 2015). Similar to Metro Manila, the climate in Boracay is influenced by cool northeast wind (Amihan) and southwest wind (Habagat) that both modify the variability of ambient atmospheric pollutants. Between 2010 and 2020, the average surface temperature and wind speed in Boracay were 28.3 ± 1.1 °C and 6.7 ± 1.7 m s⁻¹, respectively.

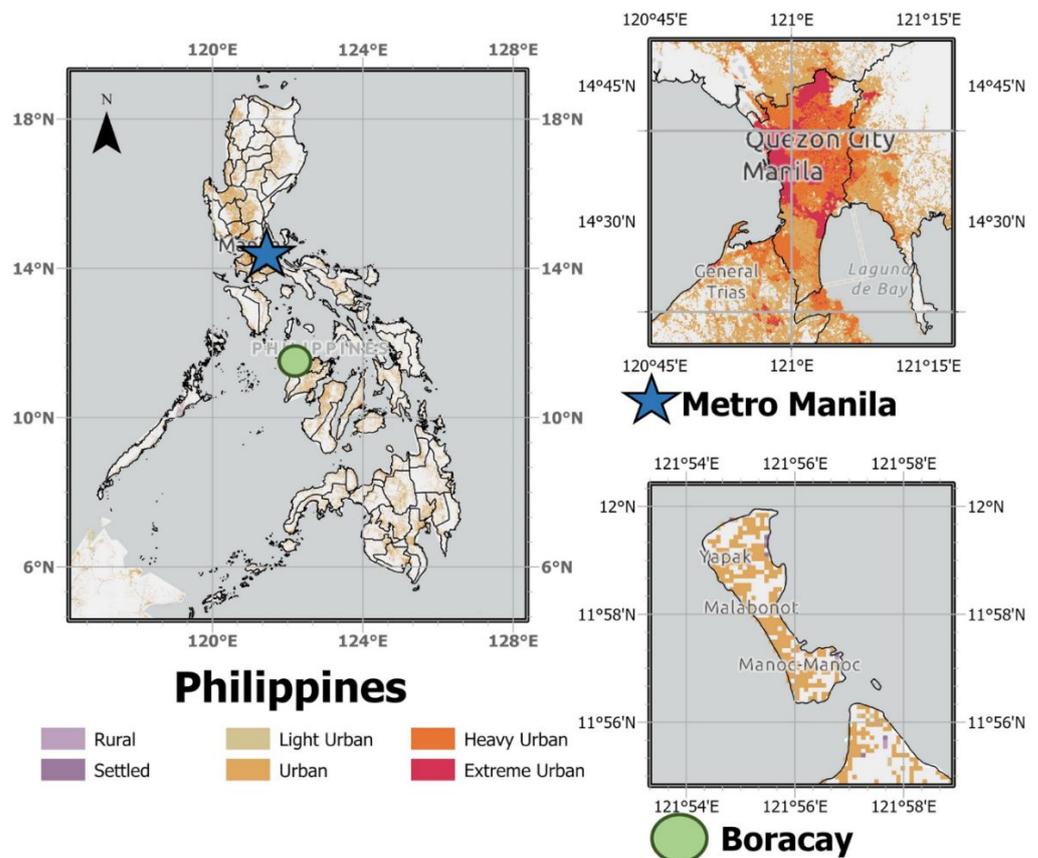
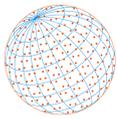


Fig. 1. Geographical locations of Metro Manila (star symbol) and Boracay (circular symbol) in the Philippines. Included in this figure is the layer of world population density estimate for 2016, which highlights the urbanized conditions in the two sites where at least 1,908 persons are residing per square kilometers (Urban). For extreme urban conditions like Metro Manila, 26,331 or more live per square kilometer which may influence the atmospheric conditions in this area.



2.2 Characterization of Regional PM_{2.5} and Aerosol Components Using MERRAero

MERRAero provides a simulated surface mass concentration of fine aerosols (PM with diameter $\leq 2.5 \mu\text{m}$) based on the GOCART concentrations of the major aerosol components. MERRAero total PM_{2.5} is given as follow:

$$\text{PM}_{2.5} = 1.375 \times (\text{SO}_4) + 1.4 \times [\text{OC}] + [\text{BC}] + [\text{DS}_{2.5}] + [\text{SS}_{2.5}] \quad (1)$$

where SO₄, OC, and BC are the simulated concentration of sulfate, organic carbon, and black carbon, respectively. The factor 1.375 was utilized to convert the sulfate concentration to neutralized ammonium sulfate, which is considered a major component of atmospheric aerosols. The 1.4 multiplier accounts for the conversion of organic carbon to organic matter (OM), a particular converter factor applicable for urban sites like Metro Manila and Boracay (Turpin and Lim, 2001). DS_{2.5} and SS_{2.5} are dust and sea salt with a size less than or equal to 2.5 μm . MERRAero provides global data for 72 atmospheric vertical layers over a uniform grid size of 0.5° latitude \times 0.625° longitude (approx. 55 km \times 70 km). More details on the simulation and reanalysis procedure used in MERRAero (e.g., assimilation of bias-corrected MODIS AOD) can be found in prior studies (Buchard *et al.*, 2016; Provençal *et al.*, 2017a; Provençal *et al.*, 2017b).

The hourly and monthly surface simulated concentration of the atmospheric aerosol components were accessed using the Goddard Interactive Online Visualization ANd aNalysis Infrastructure (Giovanni) Air Quality Online tool (<http://giovanni.gsfc.nasa.gov>) (Acker and Leptoukh, 2007; Berrick *et al.*, 2009; Prados *et al.*, 2010). Giovanni is a free and readily available asynchronous web-service-based workflow management system that provides several NASA remote sensing data and other Earth Science data for different applications such as air quality analysis and public health improvement. Currently, it has more than 150 data sets with at least 1000 geophysical properties. Besides the data access, Giovanni was utilized in this study for spatial maps of several data variables, animations, and area-averaged time series.

2.3 Integrated Filter PM_{2.5} Collection

Ground-based filter collection was implemented in Boracay and Metro Manila using Gent Dichotomous sampler to capture both coarse and fine aerosols. The samplers were deployed twice a week (Wednesday and Sunday) to account for weekday and weekend emissions. Sampling time both starts and ends at midnight with a constant on and off procedure every hour to reduce the filter mass overloading, particularly during the period of heavy traffic emissions. Pre-weighed Nuclepore filters were used to collect particulate matter, which were stored at low-temperature conditions ($\sim 8^\circ\text{C}$) after the aerosol collection to reduce particle loss.

2.4 Gravimetric Analysis PM_{2.5} and Speciation of Black Carbon Sources

After the filter collection, nuclepore filters were conditioned in a clean room with controlled R.H. and temperature (i.e., 30–40% and 20–23°C) at least 24 hours before gravimetric mass analysis using a microbalance. An M34D Smokestain Reflectometer with $\epsilon = 7 \text{ m}^2 \text{ g}^{-1}$ (ϵ : the average fine particle mass absorption coefficient) and Multi-wavelength Absorption Black Carbon Instrument (MABI) were utilized to characterize the BC components of PM.

3 RESULTS AND DISCUSSION

3.1 Sources and Seasonal Profile of PM_{2.5}

Fig. 2 shows the eleven-year time series of the speciated components of PM_{2.5} generated from satellite data and MERRAero model simulation. The calculated PM_{2.5} mass concentrations of Metro Manila and Boracay were 12.3 ± 2.4 and $12.9 \pm 3.6 \mu\text{g m}^{-3}$, with a max of 20.4 and 25.7 $\mu\text{g m}^{-3}$ concentration, respectively. Included also in Fig. 2 are the ground-based analyses of PM_{2.5} using 24-hour integrated samplers (turquoise shaded region). Evidently, the MERRAero simulation underestimated the PM_{2.5} mass concentration, particularly for the metropolis site. The average PM_{2.5} masses of Metro Manila and Boracay were 26.7 ± 12.2 and $20.9 \pm 10.5 \mu\text{g m}^{-3}$, which the

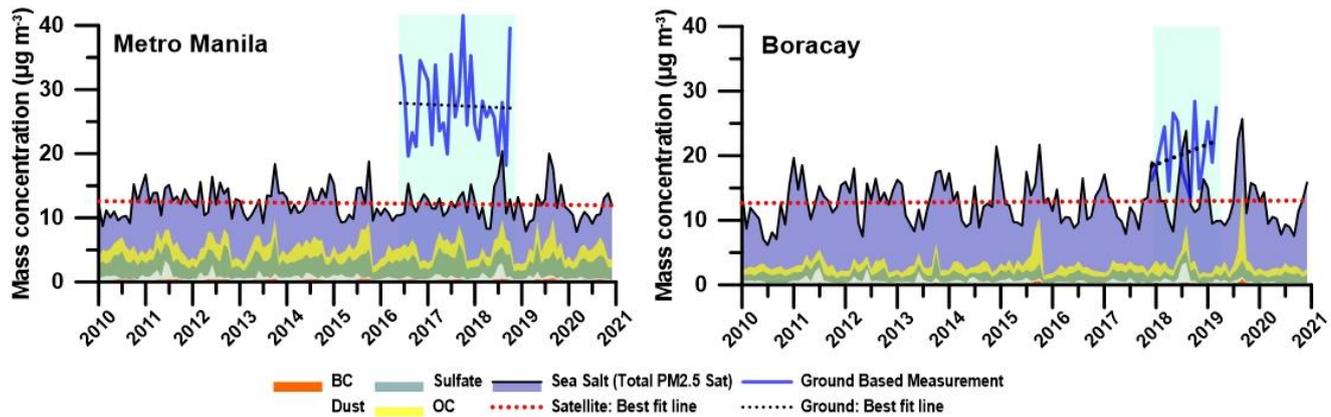
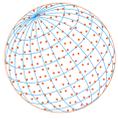


Fig. 2. The stacked concentration of the PM composition of Metro Manila (left) and Boracay (right). The black line represents the simulated mass concentration of PM_{2.5}, calculated from the sum of the masses of individual species. The red dotted line shows the regression line of the PM mass during the 11-year measurement. The turquoise shaded region is the period when ground measurements PM collections were implemented in Metro Manila (Valenzuela City) and Boracay.

simulated PM_{2.5} only estimated by 50 and 74%, respectively. The lower mass concentration from MERRAero was well expected due to unaccounted particulate nitrate (NO₃⁻) in the PM mass construction (see Eq. (1)) (Provençal *et al.*, 2017a). In a highly urbanized area like Metro Manila, NO₃ is a major component of particulate matter, generated from the oxidation of nitrogen dioxides (NO_x).

The weather pattern of the Philippines, which impacts the concentration of atmospheric aerosols, is influenced by two major seasons that are classified based on temperature and rainfall amount. Between June to November, the Philippines is under a rainy season while the rest of the year is characterized by a hot dry season (Matsumoto *et al.*, 2020). T-test analysis indicated a significant difference ($p < 0.01$) in PM mass of Metro Manila observed between the dry ($13.0 \pm 1.2 \mu\text{g m}^{-3}$) and wet ($11.5 \pm 1.0 \mu\text{g m}^{-3}$) seasons. In comparison, the average regional fine aerosols masses of Boracay during the two seasons were comparable ($\sim 12 \mu\text{g m}^{-3}$). The distinction of the mass of PM of National Capital Region due to seasons was attributed to the enhanced organic matter during the wet season ($2.29 \pm 0.29 \mu\text{g m}^{-3}$; Dry: $1.64 \pm 0.14 \mu\text{g m}^{-3}$), particularly between August–October when transboundary plumes with a high concentration of biomass burning compounds. This will further be explained in the succeeding section. Moreover, the pronounced occurrence of typhoons during the wet season, particularly between July and September, also elevated the concentration of PM of Manila during the 2nd half of the year. Approximately 20 tropical cyclones influence the meteorological conditions of the northern side of the Philippines (e.g., Luzon island) and the air quality. Before the typhoon landfalls, stagnant meteorological conditions induced the accumulation of air pollutants, as observed in prior studies (Chow *et al.*, 2018; Lin *et al.*, 2021; Wu, 2021). The detailed effect of typhoons on Manila's air quality will be further explored in succeeding studies.

The two sites recorded comparable mass concentrations (i.e., $\sim 12 \mu\text{g m}^{-3}$, see Fig. 3), yet with different distribution of major PM components. The Boracay site was highly influenced by sea salt, 75% of the calculated PM mass. The proximity of these regional background stations to coastal and marine sites enhanced the concentration of seas salt components of aerosols. On the other hand, the highly urbanized region of Metro Manila reported only 55% mass accounted to sea salt due to the dominant contribution of sulfates from vehicular, industrial, and fossil fuel combustion emissions. Here, we chiefly attributed the elevated concentration of sulfate to transportation activities, which is highly rampant in the heavily urbanized metropolis. Petroleum-based fuels used in automobiles consist of sulfur compounds that are oxidized to SO₂ during the combustion process (Xie *et al.*, 2020). Indeed, particulate inorganic nitrate is a better tracer for transportation activities. However, simulation of particulate bound nitrate is still unavailable in MERRAero. During the eleven-year measurement, 22% of the mass of atmospheric aerosol was from sulfate, while the Boracay has less than 10% of PM_{2.5} accounted to sulfate. On average, the sulfate mass concentration in the megacity was $2.62 \mu\text{g m}^{-3}$, while Boracay reported $1.24 \mu\text{g m}^{-3}$

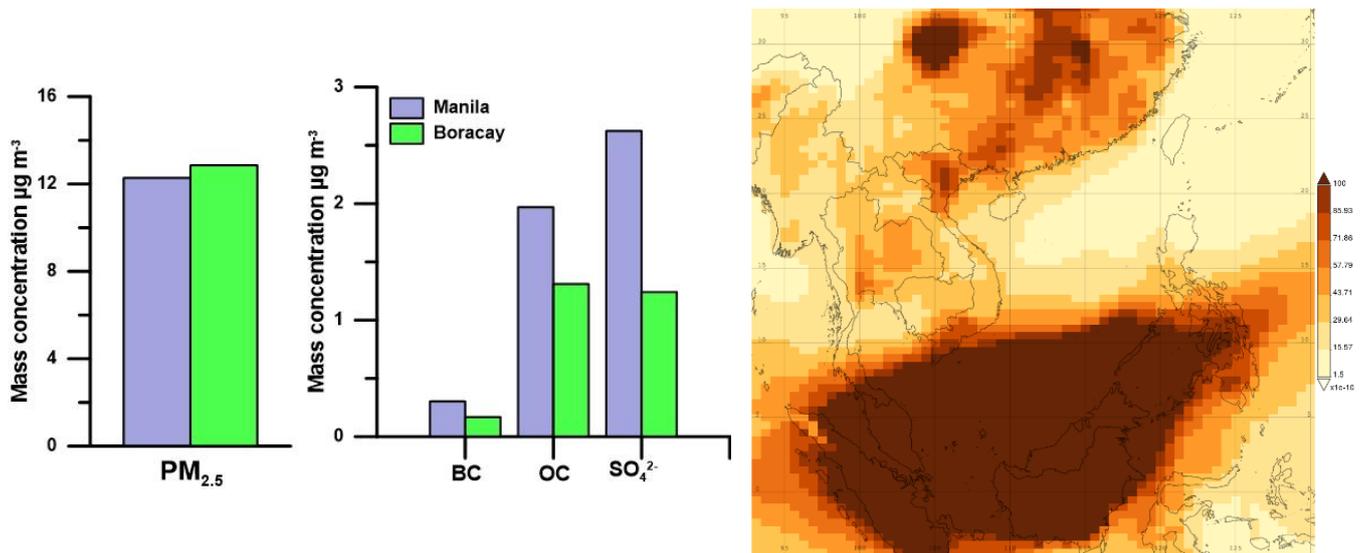
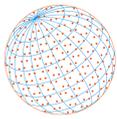


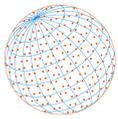
Fig. 3. (left) Comparison of $\text{PM}_{2.5}$, BC, OC, and SO_4 average mass concentration of Manila and Boracay. (Right) Time-averaged map of organic carbon in South East Asia during the strong transboundary transport of BB plumes of Sept 2019.

(see Fig. 3). This clearly shows the evident influence of anthropogenic activities, particularly from the transportation sector, in the atmospheric aerosol mass in highly urbanized areas like Metro Manila.

3.2 Variability of Regional Black Carbon, Organic Matter, and Sulfate

The regional BC of Metro Manila and Boracay also showcased the differences in the contribution of anthropogenic activities in the two areas. During the eleven-year measurement period, Metro Manila recorded $0.30 \pm 0.16 \mu\text{g m}^{-3}$ average concentration of BC, twice the mean values observed in Boracay ($0.17 \pm 0.16 \mu\text{g m}^{-3}$, see Fig. 3). The black carbon in the megacity was enhanced by the extensive vehicular activities, which emit high levels of BC. Moreover, Metro Manila is also dominated by vehicles with pre-EURO engines (e.g., public utility vehicles) that exacerbated the black carbon in the megacity. A previous study about carbonaceous aerosol in Metro Manila indicated that inefficient combustion processes dominated the megacity (Bautista *et al.*, 2014). Surprisingly, the suburban site of Boracay reported 28 days with more than $1.0 \mu\text{g m}^{-3}$ BC from satellite measurements, compared to only 16 days in the Metropolis area. High BC loading days in Boracay occurred mostly during September 2015 and 2019, which was suspected of coming from transboundary transport of biomass burning. Backward trajectory analysis (see Fig. S1 in supplement) indicated that the majority of the plumes arriving in September 2019 were predominantly coming from the southeast (i.e., Malaysia, Cambodia, and Vietnam) while other years showed mixed sources (i.e., west the Pacific Ocean). Comparison with ground-based measurement indicated that the regional BC concentration can account for 42% of the locally detected BC in Boracay while only 5% in Manila. MERRAero underestimates the mass concentrations of aerosol components in highly urbanized regions such as Metro Manila due to several point sources (e.g., vehicular and industrial), which the satellite data cannot completely simulate.

Organic matter was the major fraction of the fine aerosols for both sites. During the eleven-year measurements, the average OM masses in $\text{PM}_{2.5}$ were 1.97 ± 0.72 and $1.31 \pm 1.17 \mu\text{g m}^{-3}$ (see Fig. 3), which accounts for 16 and 10% of $\text{PM}_{2.5}$ in Metro Manila and Boracay, respectively. Similar to BC, no evident monthly and yearly trends were observed except for the two periods (September 2015 and 2019) with prominent spikes of OM, particularly in Boracay with less amount of OM in the atmosphere. The OM in Boracay reached as high as 7.29 and $10.86 \mu\text{g m}^{-3}$ during the two events, which were 456 and 729% increase from the eleven-year average. Based on the remote analysis of organic carbon in Southeast Asia (see Fig. 3 and Fig. S2 in supplement), the enhancement of surface-level organic carbon during the two periods started in the southwest of the Philippines, which drastically increased the OM in the mid and southern regions of the Philippine as these events were also observed in Southern Philippines (see Fig. S3 in supplement).



The suspected source of high OM, as well as BC, was biomass burning, which is rampant in equatorial Southeast Asia between August to October. Most of the fire spots in SEA were shrubland and forest fire, which accounted for more than 75% of the biomass burning events (Yin, 2020). This was further attenuated by the general wind circulation occurring during this period. During the wet season, the southwest monsoon (“Habagat”) brings winds that typically originate from equatorial SEA countries with frequent occurrence of biomass burning events (Matsumoto *et al.*, 2020).

As indicated earlier, sulfate had a substantial mass burden (22%) to the PM mass calculated in the megacity. In highly polluted environments such as China, the rapid transformation of SO₂ to sulfate has an important contribution to severe haze conditions, even in cloud-free scenarios (Wang *et al.*, 2016). In Metro Manila, a linear relationship between the proxy of the sulfate formation, the molar ratio of SO₄²⁻ and SO₂, and PM_{2.5} mass was observed as shown in Fig. 4. Even at relatively clean atmospheric conditions (PM_{2.5} < 50 μg m⁻³) compared to China, the sulfate formation in the particle phase still influenced the formation and variability of fine aerosols of Metro Manila. In the megacity, photochemistry was one of the drivers of the formation of sulfate aerosols. Fig. 4 shows the diurnal profile of the proxy of the sulfate formation, which showcases the efficient transformation of SO₂ during noontime. Between 2010 and 2020, the average proxy value was 0.19 ± 0.05, similar to a temperate city in China (Wang *et al.*, 2016). The SO₄-SO₂ ratio reached a value of more than one between 11:00 to 15:00, highlighting the importance of gas-phase oxidation of SO₂ by hydroxyl radical. Moreover, the monthly trends of the SO₄-SO₂ ratio in Fig. 4 illustrate a bimodal feature, with evident enhancements between March to April and September to October. The first peak was attributed to the elevated photochemistry during the summer season, while atmospheric water content induced the formation of sulfate during the latter part of the year. Metro Manila experiences a rainy/wet season during the 2nd half of the

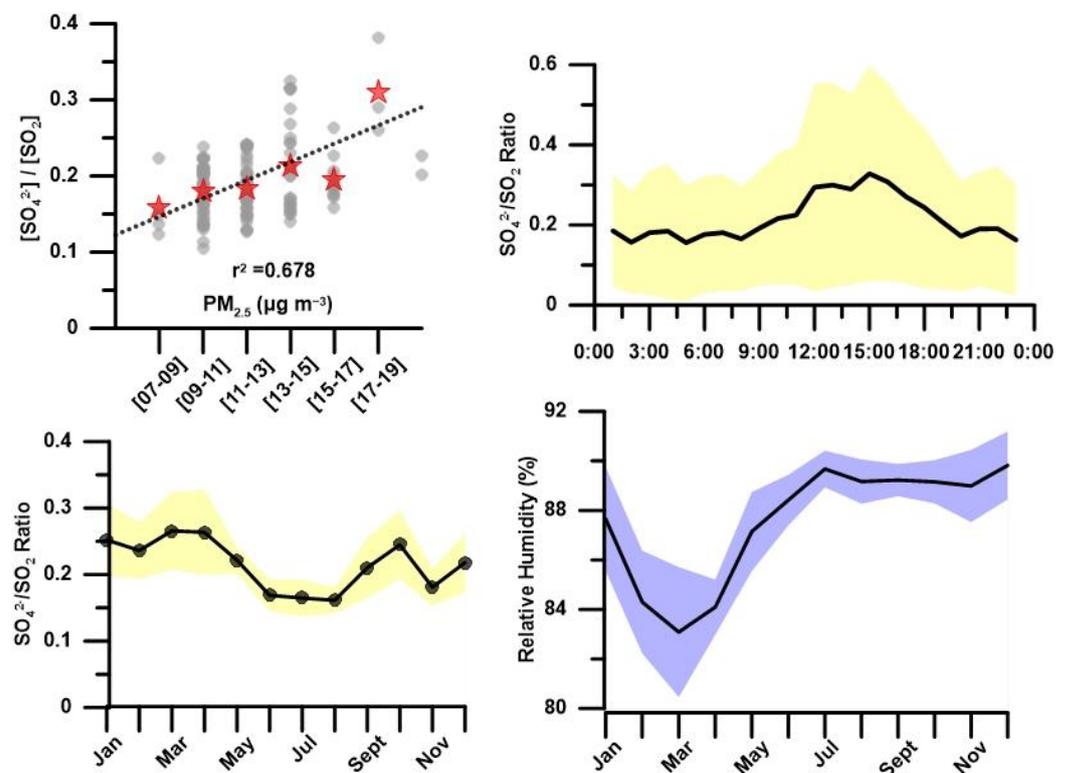
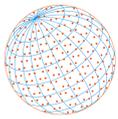


Fig. 4. Formation of sulfate aerosols in Metro Manila. (a) The ratio of sulfate to SO₂ binned to the aerosol mass concentration of PM_{2.5}. The red star represents the average value per clustered mass concentration while the dotted line is the best fit line of the average PM mass and SO₄-SO₂ ratio. (b) Diurnal profile of the proxy of transformation of SO₂. (c) The monthly trend of the SO₄-SO₂ ratio during the eleven-year measurement. (d) Relative humidity in Manila. Note that the y-axis starts at 80%. The shaded regions in plots (b)–(d) are standard deviations.



year, supported by the trend of R.H. given in Fig. 4. High aerosol liquid water content induces multiphase oxidation of SO₂ in aerosol particles (Cheng *et al.*, 2016; Liu *et al.*, 2021), which explains the enhancement of both the SO₂-SO₄ ratio and sulfate concentration. For Boracay, a consistent value of more than unity (Ave. = 1.95 ± 1.32) was recorded throughout the year, which was attributed to the interference of marine sulfate generated from sea spray events.

3.3 Characterization of Key Sources: Biomass Burning vs. Anthropogenic Emissions

The surface concentrations of OM and SO₄ from MERRAero were previously utilized for PM_{2.5} source attribution (Saunders and Waugh, 2015). The ratio of the two aerosol components was used as a metric to identify the sources of aerosols, particularly in distinguishing emissions from anthropogenic and biomass burning sources. Metro Manila and Boracay, even both are urbanized areas, had dissimilar source profiles based on the ratio of OM and SO₄. The megacity recorded an average ratio of 0.77 ± 0.20 , while Boracay had a mean value close to unity (1.06 ± 0.55). The urbanized island had 48 months with values greater than one throughout the measurement period, which accounted for 37% of the study period. On the other hand, Manila had fewer months (14) with ratios exceeding 1.0 due to elevated sulfate aerosol mass concentration. The ratios indicate that the anthropogenic emissions, particularly vehicular emission, dominated the sources of aerosol in Metro Manila, in contrast, equal contributions from human-related activities and biomass burning (e.g., forest fire) sources influenced the variability of PM in Boracay. The monthly trend of OM-SO₄ of Boracay and Metro Manila in Fig. 5 demonstrates the evident influence of biomass burning between August and October, particularly for Boracay with seven months with ratios above 2.0 that typically occurred during these months. Furthermore, the max values of the OM-SO₄ ratio of Boracay occurred during September 2015 (3.67) and 2019 (4.20), which coincided with months that were suspected to be highly impacted by biomass burning events generated from equatorial SEA countries.

Integrated filter-based analysis using multi-wavelength absorption black carbon instrument can distinguish vehicle and biomass burning sources, which can be compared with OM-SO₄ ratio from MERRAero. More information regarding the MABI's instrument design, function, and interpretation of BC data from the measurement of filters is discussed elsewhere (Manohar *et al.*, 2021). During the field measurement, the MABI estimated an average concentration of 9.2 ± 3.2 (Metro Manila) and $2.0 \pm 2.2 \mu\text{g m}^{-3}$ (Boracay) of BC attributed to vehicle emissions (BC_{vehicular}). On the other hand, 0.134 ± 0.30 and $0.53 \pm 0.42 \mu\text{g m}^{-3}$ of BC of the metropolis and tourist spot

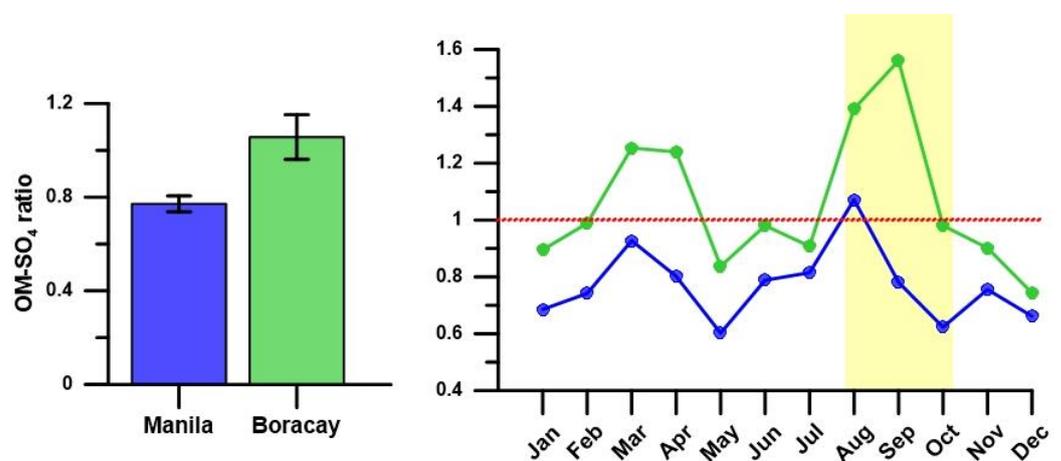
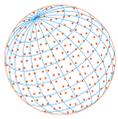


Fig. 5. (left) Comparison of mean OM-SO₄ ratio of Manila (blue) and Boracay (green) in the last 11 years. The error bar is given by the confidence interval at 95%. (Right) Average monthly profile of OM-SO₄ ratio of the two sites. The red line indicates the unity (one) value which indicates an equal contribution of biomass burning and anthropogenic sources. The yellow shaded region highlights the months with extensive transboundary biomass burning pollution from other SEA countries.



originated from biomass burning ($BC_{\text{biomass burning}}$) based on the differentiation of MABI. The median ratio between the vehicle and BB emissions (ratio = $BC_{\text{vehicular}}/BC_{\text{biomass burning}}$) were 9181 and 3.8 for Metro Manila and Boracay (see supplement for data basis of the median ratios), which highlights the comparable contribution of BB and the transportation sector in the Boracay while vehicle emissions dominated Manila. This is consistent with the results from the ratio of OM and SO_4 in Boracay with a mean ratio value of unity. Clearly, the MERRAero can efficiently simulate the regional mass concentration of surface fine particulate matter and its speciated components based on the comparison with conventional integrated filter analyses.

4 ATMOSPHERIC IMPLICATION AND CONCLUSION

Long-term trends of the surface-level fine atmospheric aerosol of two important tropical urban areas in the Philippines with varying atmospheric conditions and pollutant sources were characterized using reconstructed $PM_{2.5}$ mass from the individual concentrations of PM components generated using NASA's MERRAero. The variabilities of the regional black carbon, sulfate, and organic matter during the eleven-year measurement were also probed to understand the differences of the sources and formation pathway of PM in Manila and Boracay.

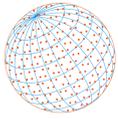
The average simulated mass concentrations of $PM_{2.5}$ in the two sites were comparable ($\sim 12 \mu\text{g m}^{-3}$), which underestimated the measurements using the traditional integrated PM filter technique by at least 50%. The absence of the particulate nitrate in the MERRAero modeling procedure was one of the primary reasons, which should be integrated to fully describe the regional PM mass particularly in urban regions with high emission and production of NO_x . Furthermore, fine aerosol was higher during the wet season in Manila, which was in contrast with the precipitation effect on the removal of air pollutants. Transboundary plumes containing traces from combustion processes and frequent occurrence of the tropical cyclones during the 2nd half of the year exceeded the aerosol sink due to wet deposition.

Even with similar $PM_{2.5}$ concentrations, Manila and Boracay each had unique aerosol component distributions, which reflect the difference in the sources of the pollutants in these regions. The atmospheric aerosols in the nation's capital had a strong contribution from sulfate aerosols (22%) due to rampant industrial and vehicle emissions, while only 10% of the mass of $PM_{2.5}$ in the well-recognized tourist spot was attributed to SO_4 . This was complemented by the black carbon level in Metro Manila, which was twice the concentration of the observed BC mass in Boracay. The abundance of vehicles (e.g., public utility jeepneys) equipped with pre-EURO engines emit a substantial amount of BCs and other pollutants that pose detrimental health risks, especially to people frequently exposed to vehicle emissions (e.g. traffic enforcers).

Organic matter concentrations in both sites were also a prominent component of fine aerosols (> 10%), particularly in Boracay during transboundary transport of biomass burning pollutants from equatorial SEA countries. Two exceptional cases of extreme enhancement of OM ($> 8 \mu\text{g m}^{-3}$) were recorded in Boracay, which were both attributed to the biomass burning events from equatorial SEA countries and some parts of the southern Philippines (i.e., Mindanao). The dominant wind pattern (southwest monsoon) during the wet season also facilitated the efficient transport of OM.

The source of the sulfate aerosols was also probed based on the proxy factor for the transformation of SO_2 to sulfate. The average ratio of sulfate to SO_2 in Boracay was above unity, which highlights other relevant sources of sulfate (i.e., sea spray) while close to 20% of SO_2 in the Manila was converted sulfate aerosols. The conversion of SO_2 to sulfate in Metro Manila showed a bimodal distribution. The enhancement during the first half of the year was accounted to the elevated photochemistry during the high temperature of the summer season. The humid conditions during the 2nd half of the year (R.H. > 90%) induced the multiphase oxidation of SO_2 , which was an integral component of haze events observed in polluted cities like Beijing. More work should be focused on the formation of sulfate aerosols to curb the level of fine aerosols in highly urbanized regions like Manila

The ratio of organic matter and sulfate was utilized to assess the dominant source of the pollutants, either biomass burning or anthropogenic emissions. Boracay reported an average value of one, which indicates the mixed contribution of two sources in the tourist spot. On the other



hand, Metro Manila had a mean ratio of 0.7, signifying the evident influence of human-related activities (e.g., vehicular emissions) in the Metropolis region. The OM–SO₄ ratio also stressed the occurrence of biomass burning events. For instance, the transboundary biomass burning observed in Boracay in September 2015 and 2019 recorded a value of more than 3.5, which was thrice the average ratio, evidently showing the effectivity of the simple factor in capturing the changes in atmospheric pollutant conditions. This was complemented with the multiwavelength analysis of BC that revealed the prevailing role of vehicle emissions in Manila while smoke from biomass burning had a comparable influence in Boracay. Overall, the long-term trend analysis of PM in Manila and Boracay showcased the different sources of pollutants and atmospheric conditions in two regions, which require unique approaches to reduce the exposure to harmful PM components. Furthermore, the results presented here underscore the effectivity of MERRAero in simulating the ground level conditions of regional atmospheric aerosols although new developments should be implemented to effectively capture the absolute concentration of PM (e.g., simulation of NO₃ aerosols).

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ADDITIONAL INFORMATION AND DECLARATIONS

Conflict of Interest

The authors declare no conflict of interest.

Data Availability

The data that support the findings of this study are available upon reasonable request from the corresponding author.

Author Contributions

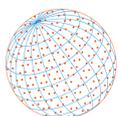
Conceptualization: Christian Mark Salvador, Angel T. Bautista VII, Raymond J. Sugcang, and Preciosa Corazon B. Pabroa; **Data Curation, Formal Analysis and Investigation:** Christian Mark G. Salvador, Iara Chantrelle Coronel, Angel T. Bautista VII, Rey Y. Capangpangan, Preciosa Corazon B. Pabroa; **Funding Acquisition:** Christian Mark Salvador, Mark Anthony M. Lavapie, Rey Y. Capangpangan, Preciosa Corazon B. Pabroa; **Investigation, Writing- Original draft preparation:** Christian Mark Salvador, Angel T. Bautista VII and Preciosa Corazon B. Pabroa; **Writing- Reviewing and Editing:** All authors reviewed the manuscript and provided comments.

Supplementary Material

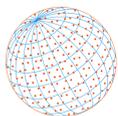
Supplementary material for this article can be found in the online version at <https://doi.org/10.4209/aaqr.220026>

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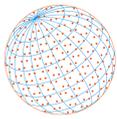
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