https://doi.org/10.5194/egusphere-2023-197
Preprint. Discussion started: 27 March 2023

An Aerosol Climatology via Remote Sensing over Metro Manila, Philippines

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https://doi.org/10.5194/egusphere-2023-197
Preprint. Discussion started: 27 March 2023

Abstract

Aerosol particles in Southeast Asia have a complex life cycle and consequently are challenging to characterize. The diverse topography and weather in the region complicate the situation. An aerosol climatology was established based on AERONET data (December 2009 to October 2018) for clear sky days in Metro Manila, Philippines. Aerosol optical depth (AOD) values were highest in August, coinciding with the summer southwest monsoon, due partly to fine particles from urban aerosol particles, including soot. Also, August corresponds to the burning season in Insular Southeast Asia when smoke is often transported to Metro Manila. Clustering of AERONET volume size distributions (VSD) resulted in five aerosol particle sources based on the position and magnitude of their peaks in the VSD and the contributions of specific particle species to AOD per cluster based on MERRA-2. The clustering showed that the majority of aerosol particles above Metro Manila were from a clean marine source (58%), which could be related to AOD values there being relatively smaller than in other cities in the region. The following are the other particle sources over Metro Manila: fine polluted (20%), mixed polluted (12%), urban/industrial (5%), and cloud processing (5%). Furthermore, MERRA-2 AOD data over Southeast Asia were analyzed using empirical orthogonal functions. Along with AOD fractional compositional contributions and wind regimes, four dominant aerosol particle air masses emerged: two sulfate air masses from East Asia, an organic carbon source from Indonesia, and a sulfate source from the Philippines. Knowing the local and regional aerosol particle air masses that impact Metro Manila is useful in identifying the sources while gaining insight on how aerosol particles are affected by long-range transport and their impact on regional weather.

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1. Introduction

Although Southeast Asia is one of the most rapidly developing regions in the world, there have been limited studies characterizing aerosol particles in the area (Tsay et al., 2013; Lee et al., 2018; Chen et al., 2020). The region represents a complex geographic, meteorological, and hydrological environment making it challenging to understand aerosol particle characteristics, especially interactions between aerosol particles with their environment (Reid et al., 2013). The island of Luzon in the Philippines in particular is very populated and is characterized by high levels of anthropogenic emissions superimposed on natural emissions from the surrounding waters (AzadiAghdam et al., 2019) and long-range transport of emissions from areas such as Indonesia and East Asia (Braun et al., 2020; Hilario et al., 2020a; Hilario et al., 2020b; Hilario et al., 2021a). The presence of clouds in the area (Hong and Di Girolamo, 2020) makes space-borne remote sensing of aerosol particles very challenging (Reid et al., 2013; Lin et al., 2014). These reasons motivated the NASA Cloud, Aerosol, and Monsoon Processes Philippines Experiment (CAMP^{2}Ex) airborne measurement campaign in 2019 to understand the interaction between tropical meteorology and aerosol particles (Di Girolamo et al., 2015; Reid et al., 2023). Prior to the airborne measurements, intensive surface-based measurements were conducted as part of the CAMP^{2}Ex weatHEr and CompoSition Monitoring (CHECSM) study between July 2018 and October 2019.

Aerosol climatology studies in different regions have proved beneficial to understand temporal characteristics of aerosol particle concentrations and properties, in addition to identifying potential source regions along with interactions with clouds and rainfall (Stevens and Feingold, 2009; Li et al., 2011; Tao et al., 2012; Crosbie et al., 2014; Kumar et al., 2015; Alizadeh-Choobari and Gharaylou, 2017; Mora et al., 2017; Aldhaif et al., 2021). The NASA AErosol RObotic NETwork (AERONET) (Holben et al., 1998) is pivotal in providing broad temporal coverage in specific locations with a column-based perspective from the ground up. To our knowledge, there has not been a remote sensing-based aerosol climatology study for the Metro Manila region of Luzon, which has approximately 16 cities, a population of 12.88 million, and a high population density of 20,800 km^{-2} (PSA, 2016; Alas et al., 2018). Studying this area is informative owing to the wide dynamic range in aerosol particle and weather conditions, which are interconnected. Aerosol particle lifecycle in the region is impacted by Philippine weather that is marked by two distinct monsoons, typhoons, and impacts from El Niño-Southern Oscillation and Madden-Julian Oscillation (Cruz et al., 2013; Xian et al., 2013; Reid et al., 2012; Reid et al., 2015; Hilario et al., 2021b).

Regional analysis of aerosol particles in Southeast Asia and Asia in general show the prevalence of biomass burning in the region, as well as the larger influence of anthropogenic emissions in East Asia (Nakata et al., 2018). These large prevalent sources may overshadow other relevant but weaker sources in the region, such as local sources. Due to the complex nature of aerosol particles, analysis techniques such as principal component analysis and clustering along with recent improvements in gridded datasets help detect spatial and temporal patterns that would otherwise be difficult to make with noise interference and even weak signals (Li et al., 2013; Sullivan et al., 2017; Plymale et al., 2021). Understanding the dominant air masses around Southeast Asia will help in distinguishing local and transported particles that influence the aerosol climatology in Metro Manila.

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The goal of this study is to use multi-year AERONET data along with other complementary datasets to address the following questions: (1) what are the monthly characteristics of aerosol particles over Metro Manila, Philippines?; (2) what are the possible sources and factors influencing the observed characteristics?; (3) what relationships are evident between aerosol particles and cloud characteristics?; and (4) what are the regional and local aerosol particle air masses that influence Metro Manila?

2. Methods

This work relies on analysis of several datasets summarized in Table 1 and the following subsections. The common time range used for all datasets is between January 2009 and October 2018.

Table 1: Summary of datasets over Metro Manila used in this work covering the period from January 2009 to October 2018.

Parameter	Data Source	Spatial Coverage	Data Repository
Aerosol Optical Depth (500 nm)	AERONET	14.635°N, 121.078°E	(AERONET) https://aeronet.gsfc.nasa.gov
Asymmetry Factor (440 nm - 1020 nm)	AERONET	14.635°N, 121.078°E	
Extinction Angstrom Exponent (440 nm - 870 nm)	AERONET	14.635°N, 121.078°E	
Fine Mode Fraction	AERONET	14.635°N, 121.078°E	
Precipitable Water	AERONET	14.635°N, 121.078°E	
Single Scattering Albedo (440 nm - 1020 nm)	AERONET	14.635°N, 121.078°E	
Refractive Index (Real and Imaginary; 440 nm - 1020 nm)	AERONET	14.635°N, 121.078°E	
Volume Size Distribution	AERONET	14.635°N, 121.078°E	
Low Cloud Fraction (MODIS)	MERRA-2	14.25°N - 14.75°N, 120.9375°E - 121.5625°E	(MERRA-2) https://disc.gsfc.nasa.gov/
Planetary Boundary Layer Height	MERRA-2	14.25°N - 14.75°N, 120.9375°E - 121.5625°E	
Relative Humidity (975 mb)	MERRA-2	14.25°N - 14.75°N, 120.9375°E - 121.5625°E	
Sea Level Pressure	MERRA-2	14.25°N - 14.75°N, 120.9375°E - 121.5625°E	
Temperature (975 mb)	MERRA-2	14.25°N - 14.75°N, 120.9375°E - 121.5625°E	
Wind (975 mb)	MERRA-2	14.25°N - 14.75°N, 120.9375°E - 121.5625°E	
Precipitation	PERSIANN	14.5°N - 15.0°N, 120.75°E - 121.25°E	(PERSIANN) https://chrsdata.eng.uci.edu/

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2.1 Datasets

2.1.1 AERONET

The central dataset used is that of sun photometer measurements and derived (inversion) parameters from the AERONET (Holben et al., 1998) site at the Manila Observatory in Quezon City, Philippines (14.64° N, 121.08° E, ~70 m. a. s. l.). Direct sunlight extinction measurements were made at nominal wavelengths of 340, 380, 440, 500, 675, 870, 940, and 1020 nm, from which aerosol optical depth (AOD) was calculated (except for 940 nm, which is for water vapor) (Eck et al., 2013). AOD is a commonly used proxy for aerosol particle loading in the air column from the ground up (Holben et al., 2001); higher AOD translates to more aerosol particle extinction in the column above a location. The extinction angstrom exponent (EAE) and the fine mode fraction (FMF) are also AERONET direct sun products that are retrieved after the application of a spectral de-convolution algorithm (O'Neill et al., 2003). For the inversion products, it is through radiative retrievals that the volume size distribution (VSD) and complex refractive index (RI) are gathered (Schuster et al., 2005) and from which single scattering albedo (SSA) and asymmetry factor (AF) are calculated.

For the inversions, four wavelengths (440, 670, 870, and 1020 nm) of the radiometer spectral channels were chosen for diffuse radiance measurements and to avoid gas absorption (Dubovik et al., 1998). Version 3 Direct Sun and Inversion algorithms (AERONET, 2019; Giles et al., 2019) were used with the Almucantar Sky Scan Scenario to derive the following parameters with level 2.0 (automatically cloud-cleared and quality controlled datasets with pre- and post-field calibrations) data quality: column AOD (500 nm), fine mode fraction (500 nm), extinction angstrom exponent (440 – 870 nm), precipitable water (940 nm), SSA (440, 670, 870, and 1020 nm), asymmetry factor (440, 670, 870, and 1020 nm), refractive index (440, 670, 870, and 1020 nm), and VSD. The version 3 products are able to keep fine mode aerosol particle data (haze and smoke) as well as remove optically thin cirrus clouds in order to retain more aerosol particle measurements in the database (Giles et al., 2019). Cloud screening in the version 3 product improves remote sensing measurements in Southeast Asia in general, where cirrus clouds are pervasive (Reid et al., 2013). At most, a total of 29,037 direct sun and 1419 inversion AERONET daytime data points were available between January 2009 and October 2018.

2.1.2 MERRA-2

Modern Era-Retrospective Analysis for Research and Applications, Version 2 (MERRA-2: 0.5° × 0.625° approximate resolution) meteorological and aerosol particle composition data (Bosilovich, 2016; Gelaro et al., 2017; Randles et al., 2017) were acquired for the area around Manila Observatory (14.25°N – 14.75°N, 120.9375°E – 121.5625°E). The following products were used: M2I3NPASM Assimilated Meteorological Fields (3-hourly) for 975 mb level winds, temperature, relative humidity, and sea level pressure; M2T1NXFLX Surface Flux Diagnostics (1-hourly from 00:30 UTC time-averaged) 2D for planetary boundary layer height; and M2T1NXCSP COSP Satellite Simulator (1-hourly from 00:30 UTC time-averaged) for MODIS mean low cloud fraction (cloud top pressure > 680 hPa).

MERRA-2 meteorological and aerosol particle composition monthly data (Bosilovich, 2016; Gelaro et al., 2017; Randles et al., 2017) were also acquired for a larger region, the Southeast Asia region (0° - 30°N, 105°E – 135°E) for the period from 2009 to 2018. This is within the spatial domain of the CAMP^{2}Ex airborne measurement campaign which, as mentioned earlier, targets the interaction between tropical meteorology and aerosol particles. The following datasets

(0.5° latitude and 0.625° longitude resolution) were used: MERRA-2 tavgM_2d_aer_Nx: Aerosol Assimilation (M2TMNXAER) for Total AOD and speciated AOD (Sulfate, Black Carbon (BC), Organic Carbon (OC), Dust, and Sea Salt) and MERRA-2 instM_3d_ana_Np: Analyzed Meteorological Fields (M2IMNPANA) for 1000 hPa and 725 hPa level U and V winds. The monthly meteorological and aerosol particle composition data for the region will be used for empirical orthogonal functions, which will be described later.

2.1.3 MISR

Monthly AOD data (Level 3 Global Aerosol: 0.5° × 0.5° spatial resolution) from 2009 to 2018 are used from the Multi-angle Imaging SpectroRadiometer (MISR), (Diner et al., 2007; Garay et al., 2018). Level 3 products are global maps of parameters available in Level 2 (measurements derived from the instrument data) products. MISR has relatively more accurate AOD and agrees better with AERONET data compared to other satellite products due to its multi-angle measurements (Choi et al., 2019; Kuttippurath and Raj, 2021). Monthly median AOD (bin 0) were extracted for Southeast Asia (0.25° - 30.25°N, 104.75°E – 134.75°E) within the CAMP^{2}Ex region. They are used for comparison to the AERONET (over Metro Manila) and MERRA-2 (Southeast Asia) monthly AOD values.

2.1.4 PERSIANN

Hourly precipitation data were obtained from the Precipitation Estimation from the Remotely Sensed Information using Artificial Neural Networks (PERSIANN) (Nguyen et al., 2019) database of the Center for Hydrometeorology and Remote Sensing (CHRS) at the University of California, Irvine (UCI). Hourly data were accumulated for running three-day totals, which were compared to AERONET data. The data were averaged between the four grids that included the area of interest as well as ensuring a similar spatial domain (14.5°N - 15.0°N, 120.75°E - 121.25°E) to the MERRA-2 dataset.

2.1.5 NAAPS

Archived total and speciated optical depths along with surface concentrations of sulfate, dust, and smoke are used from the Navy Aerosol Analysis and Prediction System (NAAPS: 1/3° × 1/3° spatial resolution) (Lynch et al., 2016), which is publicly available at https://www.nrlmry.navy.mil/aerosol/. This reanalysis product relies on the Navy Global Environmental Model (NAVGEM) for meteorological fields (Hogan et al., 2014). Hourly maps were downloaded for Southeast Asia for aerosol particle events of interest based on AERONET data. These maps help in the identification of regional emission sources.

2.1.6 NASA Worldview

Archived maps of cloud fraction (Aqua MODIS and Terra MODIS) were downloaded from NASA Worldview (https://worldview.earthdata.nasa.gov) for events of interest based on AERONET data.

2.2 Clustering

Available AERONET VSD data (0.050 µm to 15.000 µm particle radius in 22 logarithmically equidistant bins, 1419 data points) were clustered via k-means clustering (Lloyd, 1982). The algorithm used was k-means++ (Arthur and Vassilvitskii, 2006). The ideal number of clusters was chosen based on relatively highest (>0.5) average silhouette value and the presence of a cluster with a second peak in the larger accumulation mode of the VSD. The clusters were

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analyzed based on their associated meteorological conditions and aerosol particle characteristics and were classified into air mass types (Table 2) based on previous studies (Pace et al., 2006; Kaskaoutis et al., 2007; Sorooshian et al., 2013; Kumar et al., 2014; Sharma et al., 2014; Che et al., 2015; Kumar et al., 2015).

Table 2: Summary of threshold values of aerosol optical depth (AOD), angstrom exponent (AE), and fine mode fraction (FMF) used to identify air mass types.

Air Mass Type	AOD	AE	FMF	Source
Clean Fine	< 0.1	> 1	> 0.7	Sorooshian et al., 2013
Polluted Fine	> 0.1	> 1	> 0.7	Sorooshian et al., 2013
Clean Coarse	< 0.1	< 1	< 0.3	Sorooshian et al., 2013
Polluted Coarse	> 0.1	< 1	< 0.3	Sorooshian et al., 2013
Desert Dust	> 0.3	-	< 0.6	Kaskaoutis et al., 2007
Clean Marine	< 0.2	-	< 0.7	Kaskaoutis et al., 2007
Urban/Industrial	> 0.2	-	> 0.8	Kaskaoutis et al., 2007

2.3 Extreme Event Analysis

Selected types of aerosol particle events were identified to characterize different types of sources and processes impacting aerosol particle columnar properties above Metro Manila. The three events are described below.

2.3.1 Smoke Long Range Transport

Events related to transported biomass burning/smoke were chosen based on the highest black carbon contribution to total AOD from the MERRA-2 dataset, high smoke contributions to AOD from NAAPS, and a dominant submicrometer peak in the AERONET VSD (Eck et al., 1999) over Metro Manila. Maps of surface smoke contributions from NAAPS as well as back-trajectories from the National Oceanic and Atmospheric Administration's (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015; Rolph et al., 2017) were used to identify the likely source and transport pathway for the smoke cases. Three-day back-trajectories with six-hour resolution were generated based on the NCEP/NCAR reanalysis meteorological dataset and with a resolution of 1° and a vertical wind setting of "model vertical velocity". Trajectories were computed for an end point with an altitude of 500 m above ground level at the Manila Observatory. This altitude represents the mixed layer based on related surface air quality studies (Crosbie et al., 2014; Mora et al., 2017; Schlosser et al., 2017; Aldhaif et al., 2020), including a previous study for the same area (Stahl et al., 2020).

2.3.2 Dust Long Range Transport

A dust transport case was identified based on the highest dust contribution to AOD from the MERRA-2 dataset, high dust contributions to AOD from NAAPS, and an enhanced coarse peak in the AERONET VSD (compared to the submicrometer fraction) (Eck et al., 1999) over Metro Manila. Surface dust concentrations from NAAPS along with HYSPLIT back-trajectories confirmed the plausibility of dust for this case.

2.3.3 Cloud Processing

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Cloud processing events were identified based on bimodal submicrometer VSDs (Eck et al., 2012) and a relatively large sulfate contribution to AOD over Metro Manila from the MERRA-2 dataset, since this species is predominantly produced via cloud processing (Barth et al., 2000; Faloona, 2009). The presence of clouds was verified with imagery from NASA Worldview in the path of air parcels reaching Metro Manila based on HYSPLIT back-trajectories.

2.4 Empirical Orthogonal Functions

Empirical orthogonal function (EOF) analysis was performed to be able to associate the air mass clusters identified earlier with regional scale aerosol particle sources. EOF analysis was done on the monthly AOD data (January 2009 to December 2018) from MERRA-2 for the Southeast Asia region for the months similar in scope to the AERONET data. EOF analysis needs a complete dataset with no data gaps, which is not available with pure satellite retrievals; MERRA-2 reanalysis data alleviate this issue.

The monthly MERRA-2 AOD maps (0° - 30°N, 105°E – 135°E with 0.5° latitude and 0.625° longitude resolution) (Lat: 61 rows x Lon: 49 columns) for the Southeast Asia region (presented subsequently) were first deseasonalized. Then, the AOD anomaly per grid per year (of the 120 months) was calculated by subtracting the monthly mean AOD from each value of a given month (Li et al., 2013). The anomalies per grid were weighted depending on their latitude by multiplying the anomalies by the square root of the cosine of their latitudes.

EOF, specifically singular value decomposition (SVD), analysis (Björnsson and Venegas, 1997) was then performed. To prepare the data for the analysis, they were transformed such that the final matrix was a 2D matrix (120 x 2989) with each row representing a year, and each column representing a grid in the map. The matrix was analyzed for eigenvalues using SVD in Matlab, which outputted the eigenvalue (S) and eigenvector (U: principal components and V: empirical orthogonal functions) matrices. The eigenvalues were, by default, arranged in descending order. Each PC time series was standardized by dividing each PC value by the standard deviation per PC time series (120 months).

An eigenvalue spectrum was also plotted based on the variance explained by each eigenvalue and error bars that were calculated using the North test (North et al., 1982). Then, the unweighted AOD anomalies were regressed onto the first three standardized PCs. Each grid therefore had a regression between 120 pairs (unweighted AOD anomalies vs standardized PCs). From the linear regression equation, the regression coefficient per grid was calculated and plotted.

2.5 Correlations

The first three standardized PCs of AOD anomalies were correlated to deseasonalized compositional AOD fractions (Sulfate, BC, OC, Dust, and Sea Salt). For each correlation, the t-test value was calculated, and the resulting t-score was compared to a t-critical value for ~n= 100 pairs (n is the number of pairs of data, in this case 120 months) for 0.90 confidence level, which is 1.660. Correlations that have t-values exceeding +1.660 or less than -1.660 (two-tailed test) are significant (90% confidence).

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3 Results and Discussion

3.1 Meteorology and Atmospheric Circulation

Knowledge of monthly behavior of weather in the study region helps interpretation of aerosol particle data. Philippine climate is influenced both by the winter northeast monsoon (~November to April, Amihan) and the summer southwest monsoon (~May to October, Habagat) (Coronas, 1920; Flores, 1969; Matsumoto et al., 2020). Median 3-hourly temperatures per month (MERRA-2, 975 mb) (Fig. 1a) ranged from 23.2 °C in January during the winter northeast monsoon, to 27.0 °C in May during the transition from the summer season, as defined in (Bañares et al., 2021), to the southwest monsoon. May was also the month with the lowest median 3-hourly relative humidity (76.6%) (MERRA-2, 975 mb) (Fig. 1b). The highest median level of relative humidity for a month was in August (86.5 %) during the summer southwest monsoon, which is also the time of the year (June to August) when rainfall peaks in the region where the sampling station (Manila Observatory) is located (Coronas, 1920; Cruz et al., 2013). The highest mean hourly precipitation (Fig. 1i) per month was from July (0.46 mm hr^{-1}) to September (0.42 mm hr^{-1}), while March exhibited the lowest mean hourly rainfall (0.02 mm hr^{-1}). Like relative humidity and precipitation, median precipitable water (from available AERONET data) (Fig. 1h) was highest in August (4.9 cm) and lowest in February and March (3.1 cm and 3.2 cm, respectively).

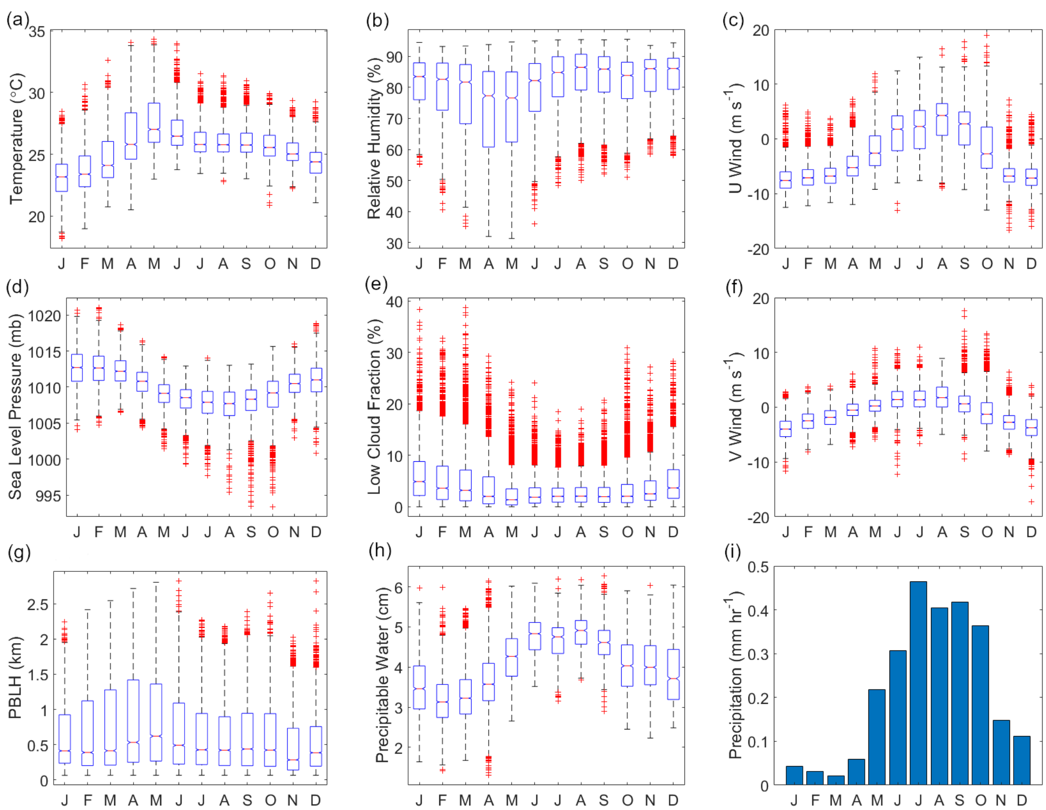

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Figure 1: Monthly characteristics of meteorological parameters for Metro Manila, Philippines based on data between January 2009 and October 2018. MERRA-2 parameters: (a) temperature, (b) relative humidity, (c/f) u and v wind at 975 mb, (d) sea level pressure, (g) planetary boundary layer height (PBLH), (e) low cloud fraction; AERONET: (h) precipitable water; PERSIANN: (i) mean hourly precipitation per month.

The lowest 3-hourly median pressures (MERRA-2) were observed (Fig. 1d) between July and September during the southwest monsoon season (~985.2 – 985.8 mb). This is also the time when the most number of tropical cyclones pass the island of Luzon (Wu and Choy, 2016). The highest 3-hourly median pressures (988.1 – 990.0 mb) were during the winter northeast monsoon.

Median winds (MERRA-2) were from the south/southwest direction from June to September (Fig. 1c and 1f), associated with the summer southwesterly monsoon. HYSPLIT back-trajectories show the same wind pattern (Fig. 2f to 2i). The highest median 3-hourly wind speeds (MERRA-2) (Fig. 1c and 1f) during the southwest monsoon were recorded for August (u: 4.2 m s^{-1} and v: 1.7 m s^{-1}). Median winds begin to transition in October and November (to the northeast monsoon: Amihan) (Fig. 2j and 2k) coming from the east/northeast and maintained until February (Fig. 2b), which is towards the end of the winter northeast monsoon. There were generally higher wind speeds and the highest median 3-hourly wind speeds of the year (MERRA-2) (Fig. 1c and 1d) in January (u: -7.6 m s^{-1} and v: -4.0 m s^{-1}). Median winds shifted toward a more easterly source from March to May (transition time before the Habagat monsoon) (Fig. 2c to 2e) accompanied by decreasing median 3-hourly wind speeds (u = -6.8 m s^{-1}, v = -1.9 m s^{-1} to u: -2.6 m s^{-1}, v = 0.2 m s^{-1}).

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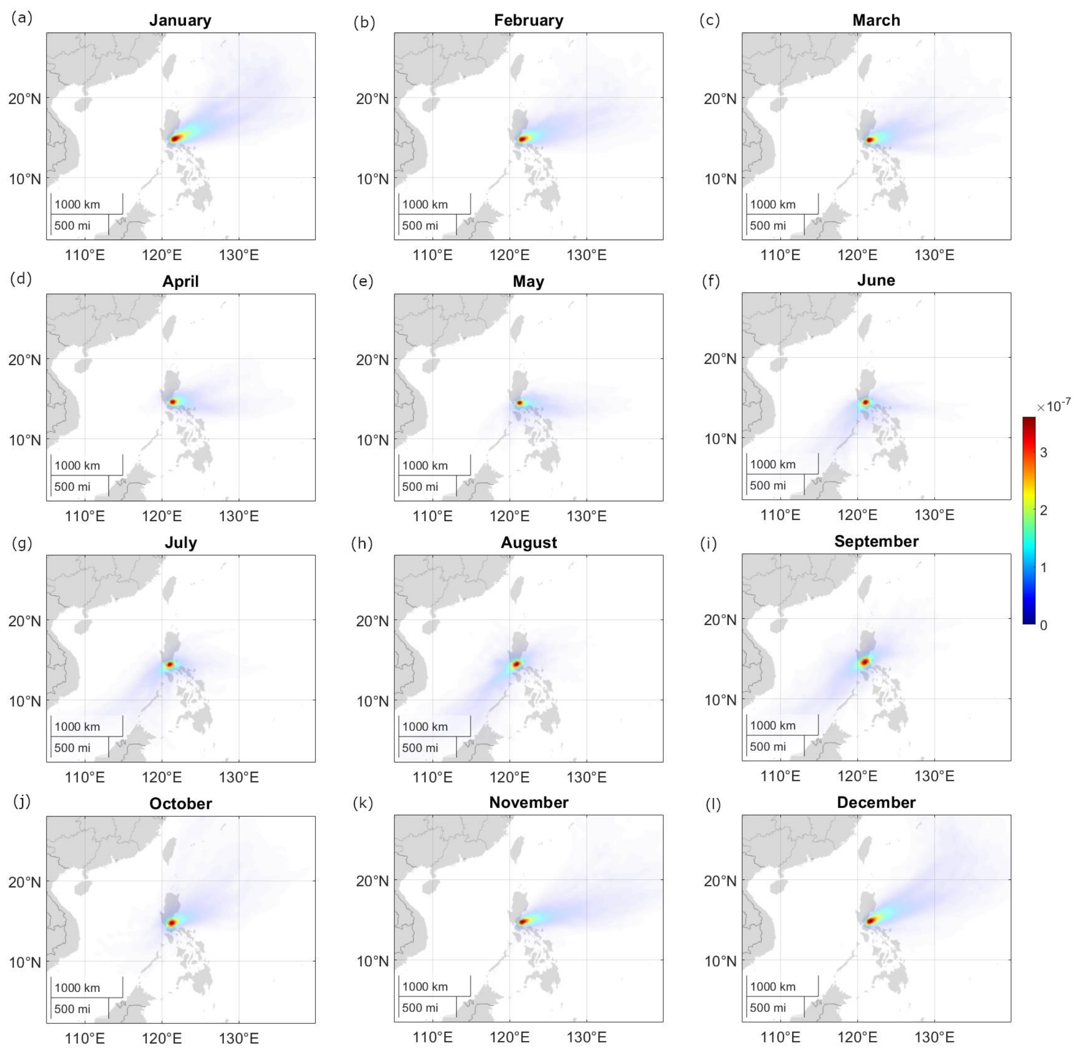

Figure 2: Density plots of trajectories reaching Manila Observatory per month from 2009 to 2018.

The transition times between the monsoons (when the wind directions shift and wind speeds change) are also the times of the highest (May, Fig. 1g, 621.2 m) and lowest (November, Fig. 1g, 279.6 m) median planetary boundary layer heights (MERRA-2). The median planetary boundary layer height was highest during the period (May) of highest temperatures, lowest relative humidity, reduced air pressure, and lowest monthly median low cloud fraction (MERRA-2) (Fig. 1e) (1.4 %). The lowest monthly median planetary boundary layer height was observed during the period (November) when temperatures were beginning to cool and air pressure was rising. The monthly maximum low cloud fraction was lowest in July (18.5 %) during the summer southwest monsoon while the monthly median and monthly maximum low cloud fractions

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(MERRA-2) (Fig. 1e) were highest (38.3 % max, 4.9 % median) in January during the winter northeast monsoon.

3.2 Aerosol Particle Characteristics

3.2.1 Aerosol Optical Depth

Monthly median AOD (AERONET) (Fig. 3a) over the Manila Observatory was highest from August (0.21) to October (0.23) around the time of the summer monsoon when winds were coming from the southwest (Figs. 2h to 2i) (Holben et al., 2001). This is the same time of year when biomass burning activities occur in the region southwest of Metro Manila. AOD over the larger Southeast Asia region from MISR and MERRA-2 (Fig. 4) had a similarly large peak from September to October which, however, was second only in magnitude to a March peak, which is influenced by biomass burning in Peninsular Southeast Asia (PSEA) (Gautam et al., 2013; Hyer et al., 2013). This larger peak in March, attributed to PSEA west of the Philippines, was not as prevalent in the AERONET AOD data due to the dominant easterly winds in the Philippines in March (Fig. 2c) and more localized sources.

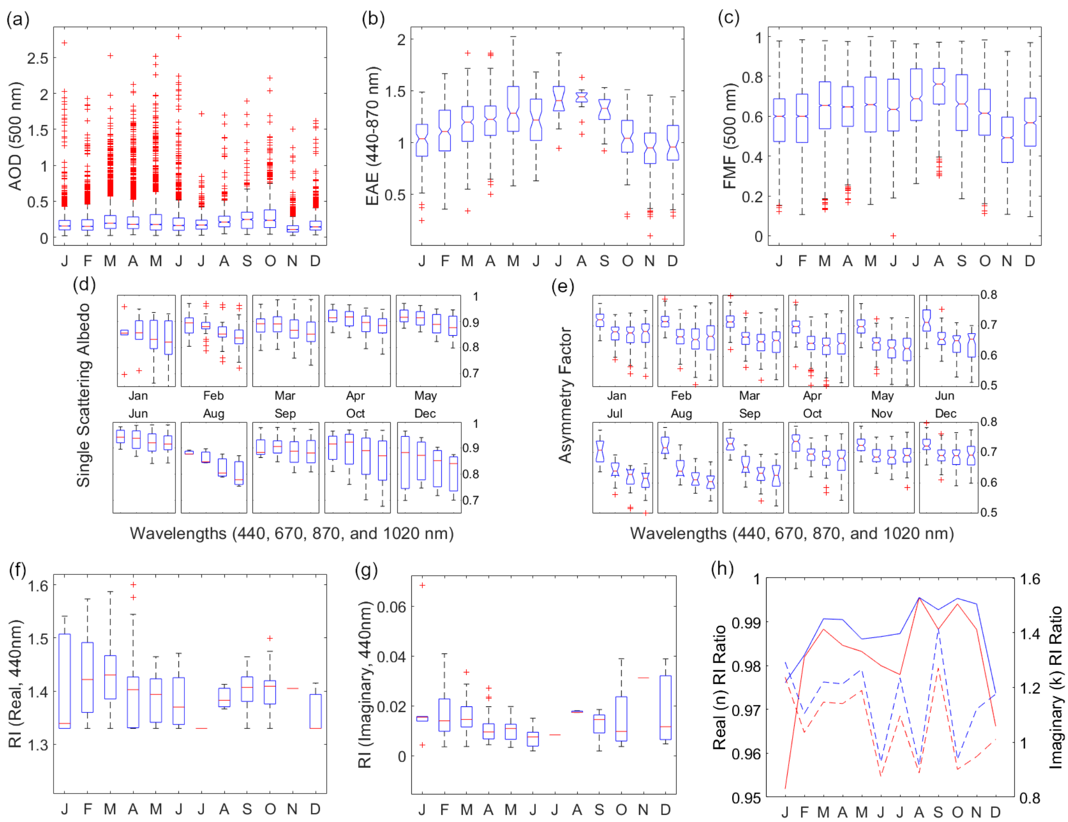

Figure 3: Monthly characteristics of AERONET aerosol particle parameters: (a) aerosol optical depth (AOD), (b) extinction angstrom exponent (EAE), (c) fine mode fraction (FMF), (d) single scattering albedo (SSA), (e) asymmetry factor (AF), (f) real and (g) imaginary refractive index (RI) values (440 nm), and (h) refractive index ratios (where the blue line is the ratio of RI at 440

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nm and 675 nm, the red line is the ratio of RI at 440 nm and the average RI for the 670–1020 nm wavelengths, and the broken lines are the imaginary refractive index ratios) for Metro Manila, Philippines based on data between January 2009 and October 2018.

There is a notable dip in the monthly median AERONET AOD from the peak in October to the lowest monthly median AOD (0.11) in November, just slightly above defined background levels (<0.1) (Holben et al., 2001), when the windspeeds were picking up and were coming from the east to northeast directions (Fig. 2k) in the direction of the Philippine Sea and the West Pacific Ocean. This dip was also observed in the regional AOD data (MISR and MERRA-2, Fig. 4).

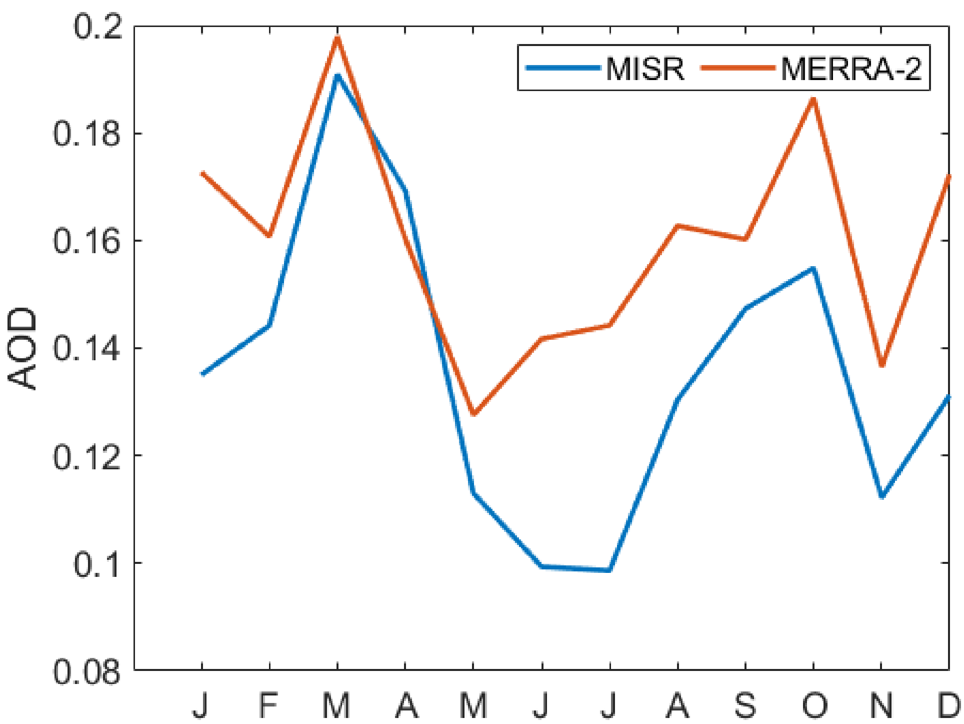

Figure 4: Monthly median AOD in Southeast Asia from 2009 to 2018 from MISR (blue line) and MERRA-2 (red line).

There were 338 instances (~1.2 % of the time based on the total number of 28,538 valid AERONET AOD data points) of AOD values exceeding 1, indicative of heavy aerosol particle loading (Huang et al., 2021). Because AOD is extrinsic (it depends on mass), AOD describes total aerosol particle loading and we examine other aerosol particle parameters from AERONET to make more informed inferences about size and composition.

3.2.2 Extinction Angstrom Exponent and Fine Mode Fraction

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The extinction angstrom exponent (EAE) relates the extinction of light at specific wavelengths and is indicative of aerosol particle size (Ångström, 1929). The EAE is usually greater for smaller particles (~4 for very small particles that undergo Rayleigh scattering and 0 for particles as large as cloud drops) (Bergstrom et al., 2007), except for when the coarse mode has a large impact on the angstrom exponent (Schuster et al., 2006). The highest monthly median EAE (Fig. 3b) from 2009 to 2018 over the Manila Observatory was observed from July (~1.4) to September (~1.3), during the southwest monsoon. This period is associated with the biomass burning southwest of the Philippines (Oanh et al., 2018; Stahl et al., 2021; Crosbie et al., 2022). The median (per month) EAE ranged from ~0.9 in November to ~1.4 in August, a range which is within the values from previous studies collected from mixed sites and urban/industrial areas with both fine and coarse particles (Eck et al., 2005; Giles et al., 2012).

EAE increases with AOD (Fig. S1), which means that the greater particle loading is contributed by smaller particles (Smirnov et al., 2002). Of the high loading cases (AOD >1), the EAE values ranged from 0.6 to 1.6, indicating fine mode particles (Che et al., 2015). The EAE values in August were the highest compared to other months including having the highest minimum value of any month (0.71) (Fig. S1), due to smaller particles (~EAE >1 for fine particles, Table 2). The lowest EAE values (0.08) and thus the largest particles were observed in December.

The fine mode fraction (FMF) describes the prevalence of fine mode particles in the column of air above the surface. The fine mode fraction (Fig. 3c) from 2009 to 2018 was highest in August (monthly median of 0.75) and lowest in November (monthly median of 0.45). This is consistent with the EAE values discussed earlier with the prevalence of smaller particles in August and larger particles in November. In August (Fig. 2h) the southwest monsoon is known to coincide with transported fine smoke particles to Luzon. In November (Fig. 2k), the prevalent winds may have already shifted to easterly (Matsumoto et al., 2020) implying more marine-related sources associated with coarser particles.

3.2.3 Single Scattering Albedo

The single scattering albedo (SSA) is the most important aerosol particle parameter determining whether aerosol particles will have a warming or cooling effect (Reid et al., 1998). SSA is the ratio of the scattering coefficient to the total extinction (scattering and absorption) coefficient (Bohren and Clothiaux, 2006) of aerosol particles. Higher SSAs are related to more reflective aerosol particles while more absorbing aerosol particles will have lower SSA values; values range from 1 (reflective) to 0 (absorbing). Monthly median SSA values were largest in June (0.94, 440 nm), suggesting the presence of more reflective aerosol particles, and smallest in August (0.78, 1020 nm). August is when biomass burning is prevalent to the southwest of the Philippines and associated with soot particles that are absorptive.

The sensitivity of SSA to different wavelengths depends on the type of aerosol particles present. More specifically, aerosol particle size and refractive index (which is related to aerosol particle composition) both affect the SSA (Dubovik and King, 2000; Bergstrom et al., 2007; Moosmüller and Sorensen, 2018). For dust-type particles, SSA increases with wavelength because of lower dust absorption in the higher visible to infrared wavelengths (Dubovik et al., 2002), while for urban particles (including black carbon), which absorb light at longer wavelengths, SSA decreases with wavelength (Reid et al., 1998; Bergstrom et al., 2002). The presence of organic carbon may affect this spectral dependence; however, because organic particles absorb in the

UV, this lowers SSA at wavelengths shorter than 440 nm (Kirchstetter et al., 2004). Monthly median SSA generally decreased with increasing wavelength for all months with available data (Fig. 3d) presumably due to the influence of more urban particles in contrast to dust. Noteworthy though are the monsoon transition months of April, September, and October (Fig. 3d), which had increased SSA from 440 nm to 670 nm, possibly from organics along with black carbon due to transported smoke. The back-trajectories for these months (Figs. 2d, 2i, and 2j) suggest sources from the northeast that are closer to Luzon during these months compared to other months. This indicates the possibility of more local sources. Increasing the certainty of sources associated with aerosol particles necessitates looking at other available aerosol particle parameters, discussed subsequently.

3.2.4 Asymmetry Factor

The asymmetry factor quantifies the direction of scattering of light due to aerosol particles, with values ranging from -1 (back scatter) to 0 (uniform scattering) to 1 (forward scatter). It is important in modeling climate forcing because it affects the vertical distribution of the radiation in the atmosphere (Kudo et al., 2016; Zhao et al., 2018). The asymmetry factor is dependent on particle size, shape, and composition and the value of 0.7 is used in radiative models (Pandolfi et al., 2018).

Lower asymmetry factors are related to smaller particles (at constant AOD) (Bi et al., 2014). Measured values due to biomass burning, for example, are 0.54 (550 nm) in Brazil (Ross et al., 1998) and 0.45 – 0.53 (550 nm and including dust) over central India (Jose et al., 2016). There have been relatively higher values observed in western, central, and eastern Europe (0.57 – 0.61 at 520 – 550 nm) (Pandolfi et al., 2018) and the U.S. East Coast (0.7 at 550 nm) (Hartley and Hobbs, 2001). In Norway, the asymmetry factor for background summer conditions was 0.62 and was higher in the springtime at 0.81 (862 nm) during Arctic haze events (Herber et al., 2002). Highest values are associated with dust such as those measured in the Sahara being 0.72 – 0.73 (500 nm) (Formenti et al., 2000). Over Metro Manila, the asymmetry factors from the AERONET data at the 675, 870, and 1020 nm were similar across months (Fig. 3e). The monthly median asymmetry factors at 440 nm ranged from 0.70 (April and May) to 0.74 (October), while for 670, 870, and 1020 nm the monthly median asymmetry factors were smaller and ranged from 0.62 – 0.69. These values were closely related to those observed over the U.S. East Coast as mentioned earlier, perhaps due to the proximity of the location to the coast (10 km east of Manila Bay and 100 km west of the Philippine Sea) as well as its location in Manila, which is a large local source due mostly to vehicles (Cruz et al., 2019).

The monthly median asymmetry factor in Metro Manila was greatest towards the end of the year (October to December) for all the wavelengths, suggesting larger particles when winds (Figs. 2j to 2l) come from the Philippine Sea in the northeast. It was in March and April that the monthly median asymmetry factor was minimal for 440 nm and in August for 670, 870, and 1020 nm. These were the times when the aerosol particles were smallest. March to April represents the driest time of the year in Manila (Fig. 1b and 1h) perhaps preventing particle growth and where the local sources may be dominating, even as back-trajectories (Fig. 2c and 2d) extend all the way from the Philippine Sea to the east. This is corroborated by results from other studies showing that the asymmetry factor seems to be enhanced by relative humidity (Zhao et al., 2018). The unexpected low asymmetry factor values in August, however, are probably because of the source of the particles. August had the highest relative humidity and precipitable water

(Fig. 1b and 1h) but is also when the back-trajectories (Fig. 2h) were from the southwest, possibly affected by the Indonesia fires, which could have transported more non-hygroscopic fine particles.

Fine particles have been observed to exhibit decreasing asymmetry factors with increasing wavelength (Bergstrom et al., 2003). This trend is observed in all the months for the monthly median asymmetry factors (Fig. 3e) suggesting the predominance of smaller aerosol particles. The greatest decrease in the asymmetry factor (all wavelengths) was in August, consistent with the lowest observed values of the year (670, 870, and 1020 nm). Transported biomass burning particles are the probable dominant particles during this time. They are usually composed of hygroscopic inorganics, non-hygroscopic soot, and relative non-hygroscopic organic fractions (Petters et al., 2009). Knowing the composition of biomass burning particles over the study region will help in the understanding of hygroscopicity and its impacts on radiation.

3.2.5 Refractive Index

Refractive index is an intrinsic parameter as it does not depend on the mass or the size of particles, and thus can be used to infer aerosol particle composition (Schuster et al., 2016). Refractive index measurements are complex since they include real and imaginary parts related to light scattering and absorption, respectively. All aerosol particles scatter light but only certain types absorb light significantly. The most prominent particle absorbers in the atmosphere are soot carbon, brown carbon (organic carbon that absorbs light), and free iron from dust (hematite and goethite in the ultraviolet to mid-visible) (Schuster et al., 2016). For this study, we examine refractive index values at 440 nm wavelength because this is the wavelength used to calculate SSA (Andrews et al., 2017). Pure sources of soot carbon have the highest real refractive index values (~1.85) as well as the highest imaginary refractive index (~0.71), both independent of wavelength (Koven and Fung, 2006; Van Beelen et al., 2014). Brown carbon and dust have relatively lower real refractive index values at 440 nm (~1.57 and ~1.54) and imaginary refractive index values (~0.063 and ~0.008) that decrease with increasing wavelength (Xie et al., 2017).

In this study the range of the monthly median real refractive index values (440 nm) was from 1.33 (December and January) to 1.43 (March) (Fig. 3f). Water uptake by aerosol particles decreases the real refractive index values (Xie et al., 2017) and thus the lowered real refractive indices over the Manila Observatory can be due to the presence of more water in the atmosphere in general and/or the increased presence of more hygroscopic particles. December and January are not necessarily the months that have the highest moisture content, but they are months when back-trajectories reaching the column over the Manila Observatory are from the Philippine Sea to the northeast presumably transporting hygroscopic particles. As reported in previous sections, relatively larger particles are observed around this time of the year and thus sea salt can be an important contributor. The greatest change in the monthly median real refractive index with increasing wavelength also was observed in December (Fig. 3h), possibly due the increased fractional contribution of constituents other than soot carbon (because the real refractive index of soot carbon is invariant with wavelength). Noteworthy as well is the month of August (Fig. 3f), which has the smallest range of real refractive index values, possibly indicating a more homogenous aerosol particle source compared to other months. August is the month with the highest relative humidity (Fig. 1b) as well as highest precipitable water (Fig. 1h), while this is

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also the month when long-range biomass burning emissions are observed to be highest, and when the real refractive index values would otherwise be expected to be highest.

Water content seems to play a significant role in the real refractive index values in Manila. March, when the monthly median real refractive index values are highest (Fig. 3f), is when precipitable water vapor (Fig. 1h) is among the lowest in the year. The months around March are also when maximum real refractive indices (1.57 in February, 1.59 in March, and 1.60 in April) were observed (Fig. 3f). March was when there was a relatively small change in real refractive index value with wavelength perhaps related to greater soot carbon fractions during this time, due possibly to the contribution of biomass burning from Peninsular Southeast Asia (Shen et al., 2014). Looking more closely at the imaginary refractive index values will help elucidate this issue.

Monthly median imaginary refractive index values (440 nm) ranged from 0.007 in June to 0.015 in September and December (Fig. 3g). These are low compared to those of the pure soot carbon mentioned earlier because of the mixed nature of the sampling site with contributions from brown carbon and dust. The highest imaginary refractive index values in September and December suggest the greatest fractional contribution of soot because the highest imaginary refractive index values are associated with soot. These are also similar in magnitude to biomass burning particles in the Amazon (0.013) (Guyon et al., 2003). The key distinction between soot carbon and other major absorbers (brown carbon and dust) is that its imaginary refractive index is invariant with wavelength. Both brown carbon and dust exhibit a decrease in the imaginary refractive index with increasing wavelength (Xie et al., 2017). The ratios of imaginary refractive index values (440 nm to average of 670–1020 nm) (Fig. 3h) show a relative invariance with wavelength (ranging from 0.88 to 1.4), which indicates the dominance of soot as the major absorber in the region (Eck et al., 2003). While observed wavelength invariance points to high soot contributions, the size of the particles can help distinguish between brown carbon, which reside mainly in the fine mode, and dust sources, which yield more coarse particles (Schuster et al., 2016). September is during the southwest monsoon, which is when, as noted in the earlier sections, fine particles were most prevalent. This is also the time when the imaginary refractive index varied most with wavelength (1.4 ratio of the imaginary refractive index at 440 nm and the imaginary refractive index average for 670 nm to 1020 nm in Fig. 3h) possibly with greater absolute contributions from brown carbon, even with the highest soot carbon fractional contributions. Brown carbon has been observed both from primary and aged aerosol particle emissions from biomass burning (Saleh et al., 2013). As noted earlier, December also had the highest imaginary refractive index values as well as relatively coarser particles, possibly due to larger dust absolute contributions even with the highest soot carbon fraction contributions. The lowest monthly median imaginary refractive index values in June, on the other hand, when fine mode particles prevail suggest highest fractional contributions of brown carbon relative to other months (Fig. 3h).

3.2.6 Volume Size Distributions

The volume size distribution (VSD) is another way to be able to more deeply characterize aerosol particles, specifically related to their effect on climate, weather, and clouds (Haywood and Boucher, 2000; Feingold, 2003). In the Manila Observatory dataset, there was a bi-modal VSD for the entire dataset (Fig. 5a). The fine mode median values peaked in the accumulation mode at 0.148 µm particle radius while the coarse mode median values peaked at 3.857 µm. The

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median coarse mode amplitudes were higher than the fine mode amplitudes for most of the year (DJF, MAM, and SON, Fig. 5b), except during the southwest monsoon (JJA) when the fine mode amplitude was higher. This is consistent with observations earlier of fine mode prevalence during the southwest monsoon. Median VSD amplitudes (Fig. 5c) were greater in the afternoon, with higher peaks for both the fine and coarse modes, compared to the morning. There was a slightly larger coarse median amplitude, compared to the accumulation mode median amplitude, for both the morning and afternoon size distributions. While the VSDs confirm several observations based on the analysis of the aerosol particle parameters presented earlier, not much further information is gained especially regarding chemical composition. Size distributions are a result of contributions from multiple sources, and thus being able to discriminate the sources based on their characteristic size distributions will help identify relevant sources.

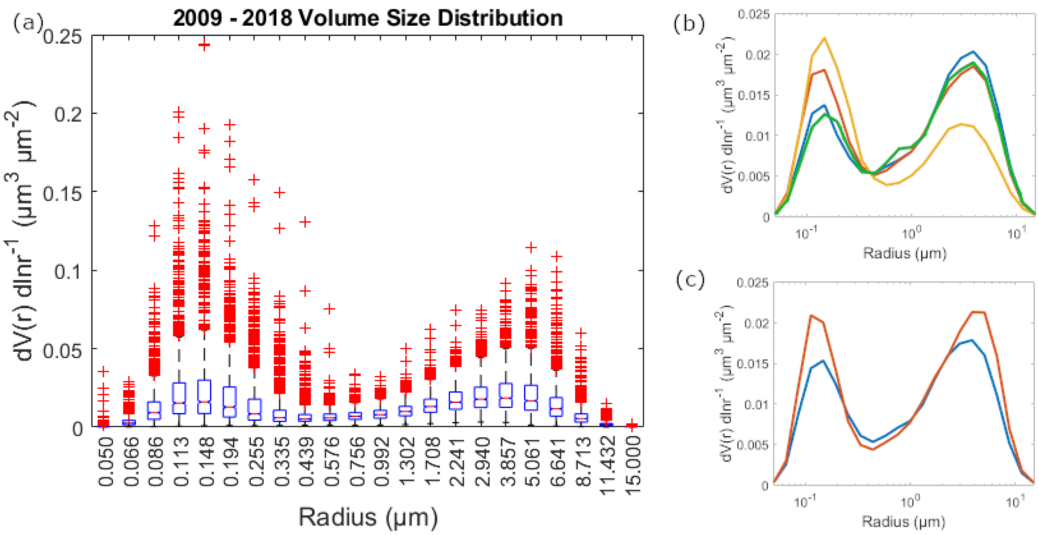

Figure 5: (a) VSD results derived from AERONET measurements at Metro Manila between January 2009 and October 2018. Median VSDs over the study period based on (b) season (blue: DJF, red: MAM, orange: JJA, green: SON) and (c) time of day (blue: AM, red: PM).

3.3 Clusters

3.3.1 VSD Cluster Profiles

Five clusters were identified to best represent the VSD (Fig. 6a). The average of the VSDs in each cluster varied depending on the height of the peaks in the accumulation mode and the coarse mode. In Metro Manila, the accumulation mode is associated with aged aerosol particles and combustion (Cruz et al., 2019). The majority of the data (830 count out of 1419 total VSD profiles) were clustered together in a profile (cluster 1) that had relatively low average magnitudes for both the accumulation (0.01 μm^3 μm^{-2}) and coarse (0.02 μm^3 μm^{-2}) modes, with the magnitude of the coarse mode peak slightly higher than the magnitude of the accumulation mode. The next prevalent cluster profile (284 counts, cluster 2) had an average fine mode peak (0.04 μm^3 μm^{-2}) which was more than twice as much than the previous profile but with a similar coarse mode peak (0.02 μm^3 μm^{-2}). The average coarse mode peak (0.04 μm^3 μm^{-2}) was the

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highest (compared to the four other cluster profiles) for the third prevalent cluster profile (166 counts, cluster 3); cluster 3 was also slightly shifted in the coarse mode to a higher radius (5.06 µm) compared to other clusters. The coarse mode dominated this VSD compared to other profiles (lower magnitude for the accumulation mode peak, 0.02 $\mu m^3\ \mu m^{-2}$). The two remaining cluster profiles exhibited high average magnitudes in both the accumulation and coarse modes. The fourth prevalent cluster profile (74 counts, cluster 4) had the highest average absolute magnitude in the accumulation mode (0.11 $\mu m^3\ \mu m^{-2}$), while the fifth prevalent cluster profile (65 counts, cluster 5) had a slightly smaller accumulation mode peak (0.07 $\mu m^3\ \mu m^{-2}$) that was shifted to a slightly higher radius (0.19 µm compared to 0.15 µm). Both clusters 4 and 5 had similar average coarse mode peak magnitudes (0.04 $\mu m^3\ \mu m^{-2}$).

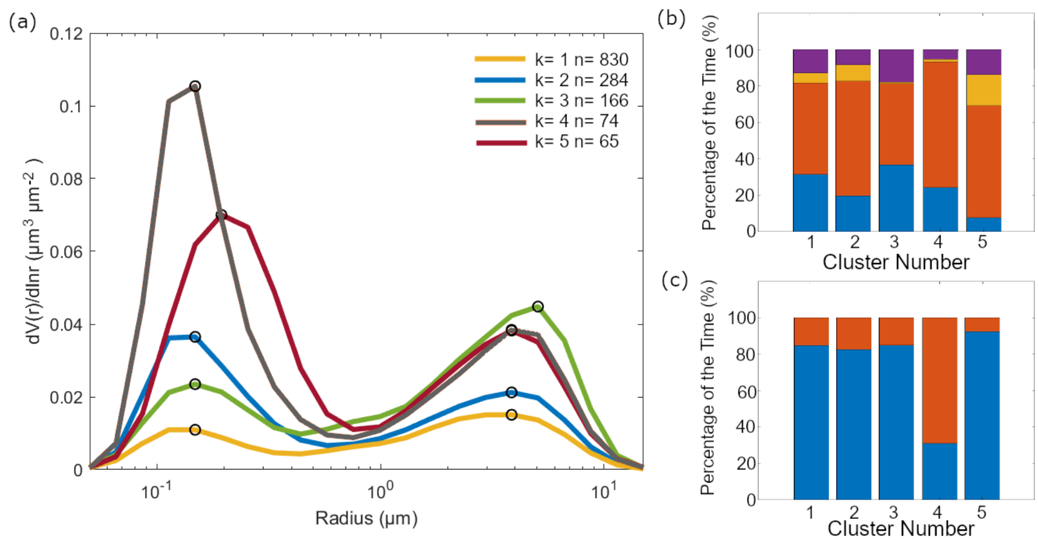

Figure 6: (a) Cluster analysis of VSD data yielding five characteristic and averaged VSDs with the number of points per cluster shown in the legend. The black circles on the curves show the peak locations in the submicrometer (<1 µm) and coarse (≥1 µm) modes. The relative abundance of each cluster is shown for different (b) seasons (blue: DJF, red: MAM, orange: JJA, violet: SON) and (c) times of day (blue: AM, red: PM).

The clusters were distributed across seasons (Fig. 6b), with clusters 1 and 2 being the most evenly distributed among the clusters. Cluster 3, which had the highest coarse mode peak, had the greatest contribution from September to November compared to other clusters. Cluster 4, which had the highest accumulated mode peak compared to other clusters, had the greatest contribution from March to May as well as to afternoon VSDs compared to other clusters (Fig. 6b and 6c). Relative contributions of VSDs from June to August were highest for cluster 5, which had the shifted accumulated mode peak.

Median total (AERONET) AOD values (Fig. 7b) were lowest (0.12) for cluster 1, though it had the second highest sea salt fractional contributions (Fig. 7a) to total AOD (MERRA-2) (31%)

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among all the clusters. Cluster 2 had relatively mid-range median total AOD values (0.27) that, along with clusters 4 and 5, were dominated by sulfate and organic carbon (46% and 20%). Cluster 3 had similar, but slightly lower median total AOD (0.25) compared to cluster 2. Cluster 3 was distinct because it had the largest total (0.04) and fractional contribution (37%) from sea salt among all clusters. Clusters 4 and 5 had the highest median total AOD values (0.47 and 0.56), with cluster 5 having the highest absolute and fractional sulfate contributions (0.14 and 64%) among the clusters. Integrating the above results with their corresponding aerosol particle properties can help associate the clusters to air masses.

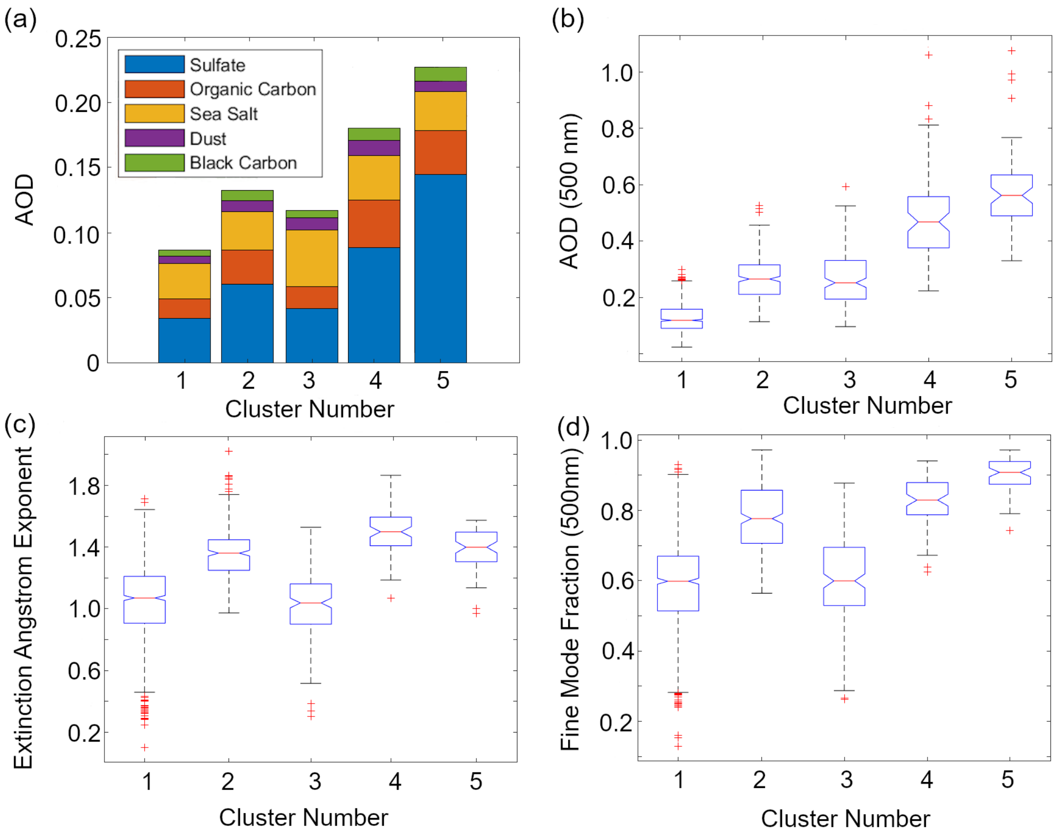

Figure 7: (a) Average compositional contributions to aerosol optical depth (AOD) from MERRA-2 per identified cluster. Boxplots of AERONET (b) total AOD (500 nm), (c) extinction angstrom exponent (EAE, 440 nm – 870 nm total), and (d) fine mode fraction (FMF, 500 nm) per cluster.

3.3.2 Air Mass Types

Air masses have been classified in previous studies based on their AOD, EAE, and FMF values (e.g., Aldhaif et al, 2021). The criteria from different studies (Table 2) were applied per cluster. Median total AOD of cluster 1 (0.12) was less than 0.2 (Fig. 7b), which is the threshold for sea

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salt sources (Kaskaoutis et al., 2007). Half of the data points in cluster 1 also fall below the threshold for clean environments (AOD < 0.1) (Sorooshian et al., 2013). Based on its median FMF (0.60) and EAE (1.07, where EAE < 1 is coarse and EAE >1 is fine) values (Fig. 7c and 7d), cluster 1 is a mixture of fine and coarse particles. Cluster 1 is the only cluster with a median that meets that threshold value for clean marine sources (AOD < 0.2), and we know from Sect. 3.3.1 that its average VSD magnitude was greater for the coarse fraction and that its sea salt contribution to total AOD was second greatest among the clusters. Thus, we can say that most probably cluster 1 is a background clean marine source, since it also is predominant throughout the seasons (Fig. 6b). This makes sense given the proximity of the ocean to Metro Manila from both the east and the west.

Most of the data from the other clusters all fall in the polluted category (Table 2), based on their median total AODs (>0.1) (Fig. 7b). Cluster 2 has a median FMF value of 0.78 (Fig. 7c), which suggests that most of the particles in this air mass are in the fine fraction. They are, however, not sufficiently dominant in the aerosol for them to be typical of urban/industrial sources. The average VSDs (Fig. 6a) of cluster 2 similarly suggest that their relative accumulation mode magnitude is higher than the coarse magnitude, but not much higher. Like cluster 1, cluster 2 is also more evenly distributed across the seasons (Fig. 6b) and could be a fine polluted background source. Metro Manila is a megacity with continuous and large amounts of sources that could be, due to its proximity to the ocean, interacting with the background.

Based on its median EAE value (1.04) (Fig. 7d), cluster 3 is mixed but mostly in the coarse fraction, consistent with its VSD profile (Fig. 6a) which has the highest coarse magnitude (FMF = 0.60) compared to the other clusters. The contribution of data from September to February is greatest in cluster 3, consistent with expected coarser particles during this period when the winds are initially shifting from the southwest before becoming more northeasterly, as previously noted. This air mass can be a mixed polluted air mass, which is possibly transported due to the large sea salt contribution to total AOD (Sect. 3.3.1).

Both clusters 4 and 5 have median total FMF (0.83 and 0.91) (Fig. 7c) values exceeding the mark (> 0.8, Table 2) for urban/industrial air masses. Combining this and results from the previous sections confirms that cluster 4 can be an urban/industrial source given that it had the highest median accumulated mode peak and organic carbon contribution to total AOD among the clusters.

Cluster 5 had the highest median total AOD and FMF values (Fig. 7c). It also had the highest (Fig. 7a) sulfate contribution to total AOD as well as a shifted accumulation mode peak (Fig. 6a). These characteristics suggest that cluster 5 is a possible cloud processing air mass (Eck et al., 2012). The larger peak in the accumulation mode is possibly the cloud signature. Previous studies have attributed this larger mode to cloud processing due to the conversion of SO_2 to sulfate (Hoppel et al., 1994). Cloud processing is a major source of sulfate (Barth et al., 2000).

The distribution of the air masses based on the abundance of the VSD profiles per cluster suggest prevalent clean marine (58% of the total VSD counts) and background fine polluted (20%) air masses over Metro Manila. The mixed polluted (12%), urban/industrial (5%), and cloud processing (5%) air masses contribute 22% all together. We can investigate more deeply and look at specific case studies that can better describe the air masses identified here.

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3.4 Case Studies

Selected case studies are used to highlight periods with the highest AOD values and strongest clear sky (no rain and heavy clouds) daytime aerosol particle sources within the sampling period. As such, the clusters that are associated with the selected case studies are the clusters (3-5) with higher VSD magnitudes.

3.4.1 Long Range Transport of Smoke

Both cases of long-range transport of smoke discussed below have similar VSDs (Fig. 8a and 9a) to the urban/industrial cluster VSD (cluster 4, Fig. 6a). Organic carbon was the dominant contributor to AOD (Fig. 8b and 9b) for both long-range transport cases. The first of two events occurred around 1 April 2020 with smoke presumed to come from East Asia. The VSD of this specific case (Fig. 8a) is most like the urban/industrial cluster (cluster 4 in 3.3.2, Fig. 6a) because of the high magnitude of its accumulated mode peak, its timing (April), and the enhanced organic carbon contribution to AOD in the area (Fig. 8b). Though the absolute black carbon contribution to AOD was highest here compared to the other case studies, and in general for the AERONET data, it was organic carbon that was more prevalent in terms of contribution to total AOD. Smoke is comprised of both soot carbon and organic carbon, amongst other constituents (Reid et al., 2005).

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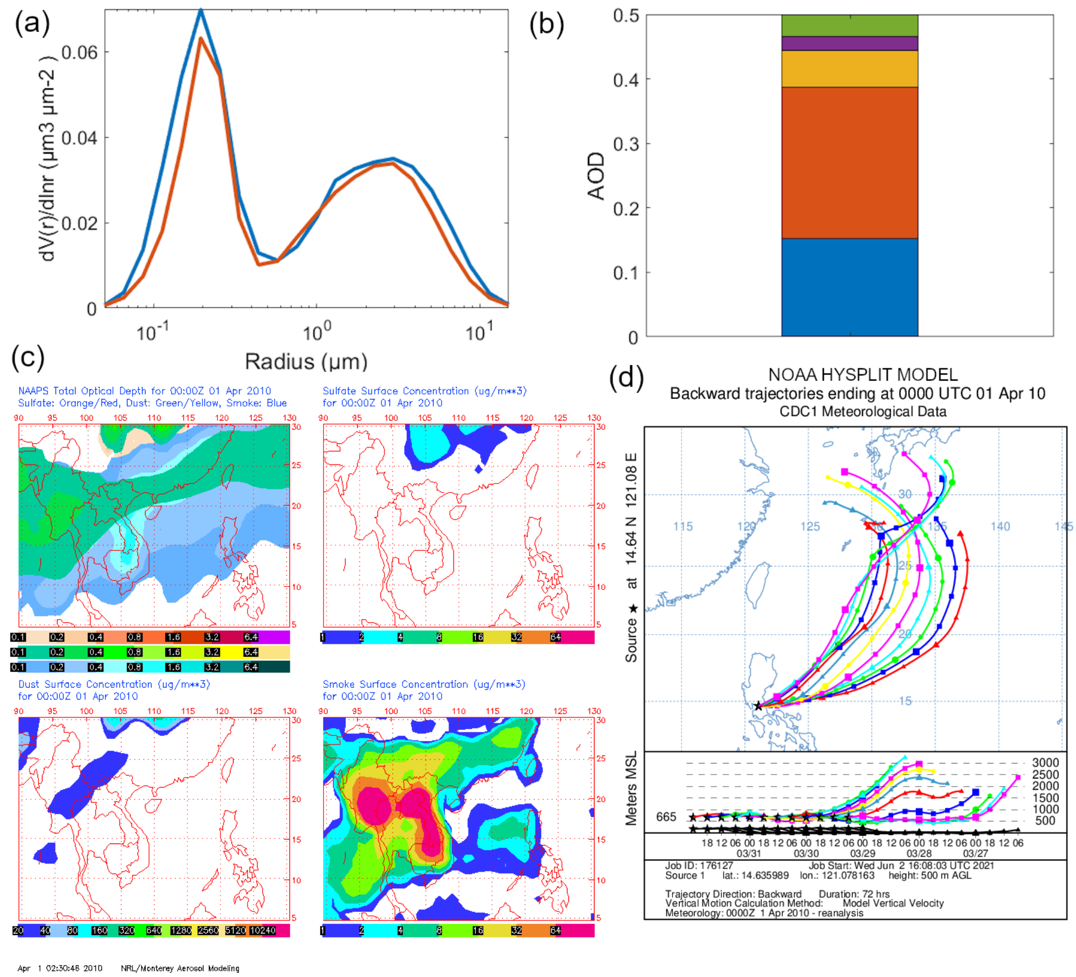

Figure 8: Case study of long-range transport (smoke – East Asia) around 1 April 2010. (a) AERONET VSDs at (blue) 00:01 and (red) 00:26 UTC, (b) MERRA-2 hourly (green: black carbon, violet: dust, yellow: sea salt, orange: organic carbon, blue: sulfate) compositional contributions to AOD closest in time to 00:01 UTC, (c) NAAPS maps of total and compositional hourly AOD (orange/red: sulfate, green/yellow: dust, blue: smoke) and sulfate, dust, and smoke surface concentrations at 00:00 UTC, and (d) HYSPLIT three-day back-trajectories arriving at Manila Observatory at 00:00 UTC.

The smoke contribution to AOD from NAAPS (Fig. 8c) for the first smoke case was visible in the Philippines (0.2) and seemed to come from East Asia were the smoke contribution to AOD was greater (reaching 0.8) especially in Peninsular Southeast Asia. Smoke surface concentrations were also widespread (Fig. 8c) with greatest concentrations in East Asia that reached the Western Philippines, though seemingly disconnected over the sea. There were observed biomass burning emissions in the Peninsular Southeast Asia (southern China, Burma, and Thailand) at this time (Shen et al., 2014). The direction of the air mass coming into Metro Manila was from

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the northeast, which curved from the west in the direction of East Asia based on HYSPLIT back-trajectories (Fig. 8d).

The second smoke case was on 15 September 2009 with the source being Southeast Asia. The back-trajectories of this case study (Fig. 9d) are from the southwest of the Philippines, and in the direction of the Malaysia and Indonesia. NAAPS maps likewise show elevated AOD, specifically smoke contribution to AOD (Fig. 9c), as well as enhanced smoke surface contributions in the area around Metro Manila for this second smoke case study. The observed AOD and smoke surface concentration increased specifically from the southwest of the Philippines in the same direction of the back-trajectories. There were fires in the lowland (peat) forests of Borneo around this time (NASA, 2009). MERRA-2 AOD contributions for this case were greatest due to organic carbon as well as sulfate (Fig. 9b), and the absolute black carbon contributions were greatest compared to other cases. The VSD of this smoke case from Southeast Asia (Fig. 9a) resembled that from long-range transported smoke from East Asia (Fig. 8a) and the urban/industrial air mass (cluster 4, Fig. 6a). This case occurred in the afternoon, which was the prevalent time that the urban/industrial air mass was observed (Fig. 6c).

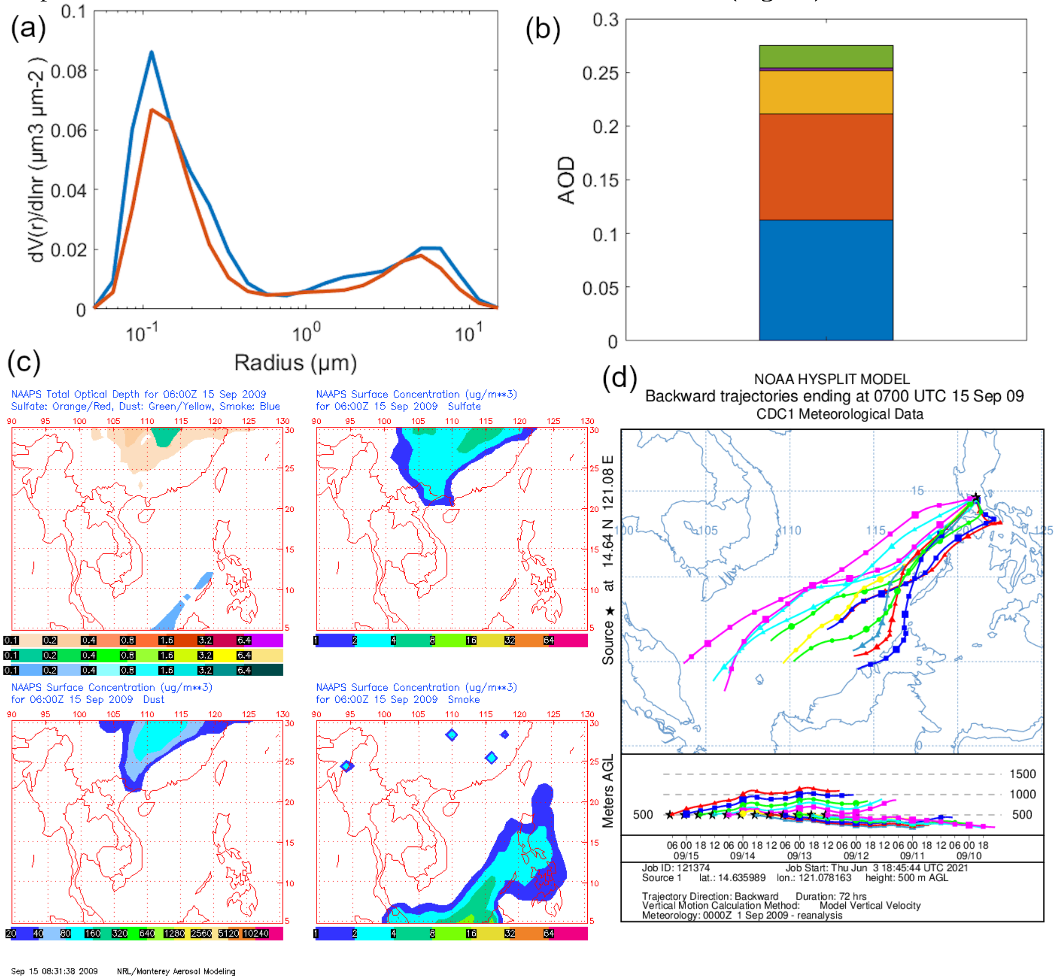

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Figure 9: Case study of long-range transport (smoke – Southeast Asia) around 15 September 2009. (a) AERONET VSDs at (blue) 07:27 and (red) 07:52 UTC, (b) MERRA-2 hourly (green: black carbon, violet: dust, yellow: sea salt, orange: organic carbon, blue: sulfate) compositional contributions to AOD closest in time to 07:27 UTC, (c) NAAPS maps of total and compositional hourly AOD (orange/red: sulfate, green/yellow: dust, blue: smoke) and sulfate, dust, and smoke surface concentrations at 06:00 UTC, and (d) HYSPLIT three-day back-trajectories arriving at Manila Observatory at 07:00 UTC.

3.4.2 Long Range Transport of Dust

The VSD of this specific case on 24 March 2018 (Fig. 10a) was most similar to the mixed polluted cluster (cluster 3), which had a mixed size distribution but a more dominant coarse contribution. This is consistent with the most dominant contribution to AOD in the area, which was sea salt and dust (Fig. 10b). The back-trajectories were from East Asia around the same latitude as Taiwan (Fig. 10d). That area, at that time, had increased AOD in general from sulfate and dust (Fig. 10c). The dust and sulfate seemed to have been transported to Metro Manila from East Asia based on the NAAPS sulfate and dust surface concentrations (Fig. 10c).

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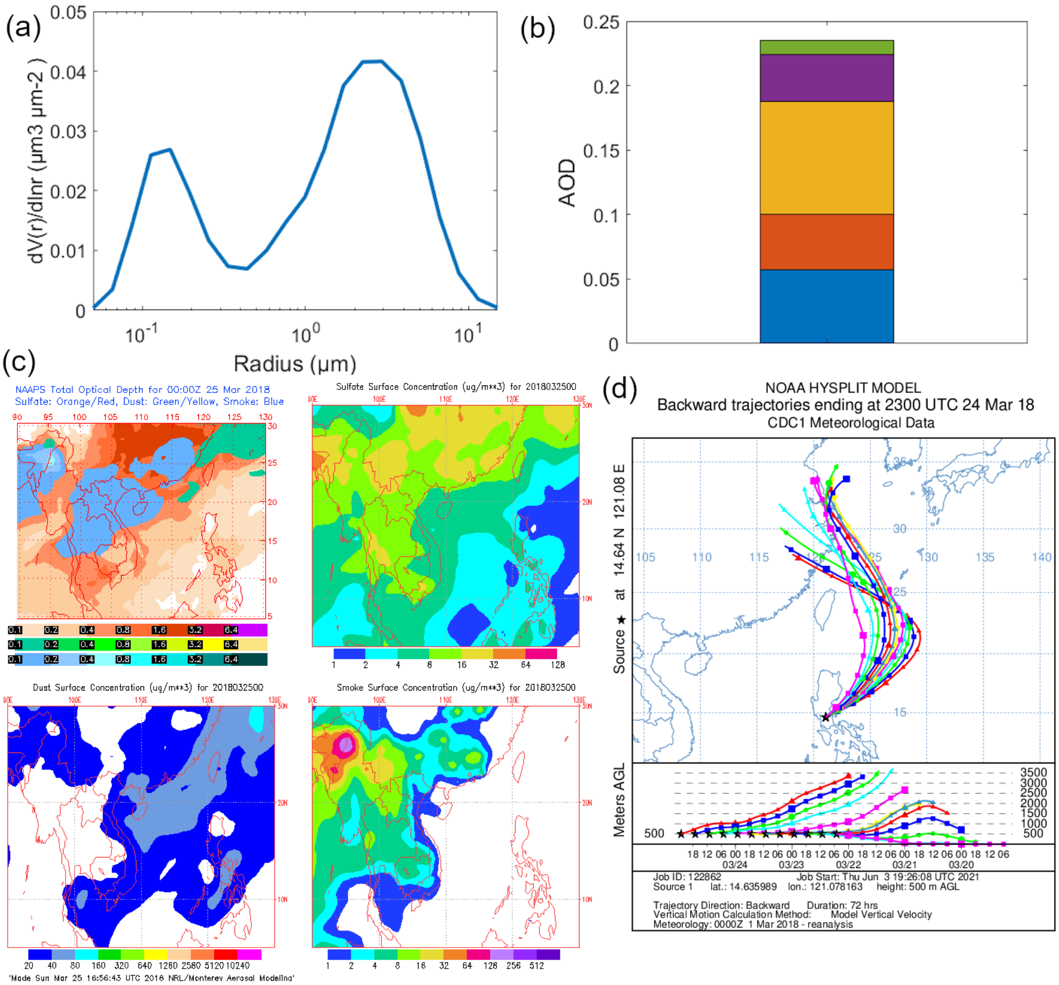

Figure 10: Case study of long-range transport (dust) around 24-25 March 2009. (a) AERONET VSD at (blue) 23:23 UTC, (b) MERRA-2 hourly (green: black carbon, violet: dust, yellow: sea salt, orange: organic carbon, blue: sulfate) compositional contributions to AOD closest in time to 23:23 UTC, (c) NAAPS maps of total and compositional hourly AOD (orange/red: sulfate, green/yellow: dust, blue: smoke) and sulfate, dust, and smoke surface concentrations at 00:00 UTC on March 25, and (d) HYSPLIT three-day back-trajectories arriving at Manila Observatory at 23:00 UTC.

3.5 Cloud Processing

Sulfate dominated the AOD (Fig. 11b) for this case on 26 August 2009 in the area around Metro Manila. This along with its VSD exhibiting a second peak (Fig. 11a) in the accumulation mode make it very similar to the cloud processing cluster (cluster 5). Sulfate has been known to be enhanced through chemical productions in clouds and is used as a signature for cloud processing (Barth et al., 2000; Ervens et al., 2018). Aqueous production of sulfate is significant in areas with

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sources and clouds (Barth et al., 2000), and this case study has both. Aside from the high sulfate contribution to AOD, the cloud fraction is very high (~100%) in the area of the back-trajectories (Aqua/MODIS, Terra/MODIS, Fig. S2). Interestingly, there is no regional AOD elevation observed in the NAAPS maps (Fig. 11c) for this time. There are increased surface smoke and sulfate levels in East Asia as well as southwest of the Philippines, and though the back-trajectories (Fig. 11d) do show a northeastward direction, they do not reach far enough into mainland East Asia. It is possible that even while there are known regional sources of sulfate in Southeast Asia (Smith et al., 2011; Li et al., 2017), this case could be local to the Philippines. There is in fact a large power plant northwest of Metro Manila (Jamora et al., 2020).

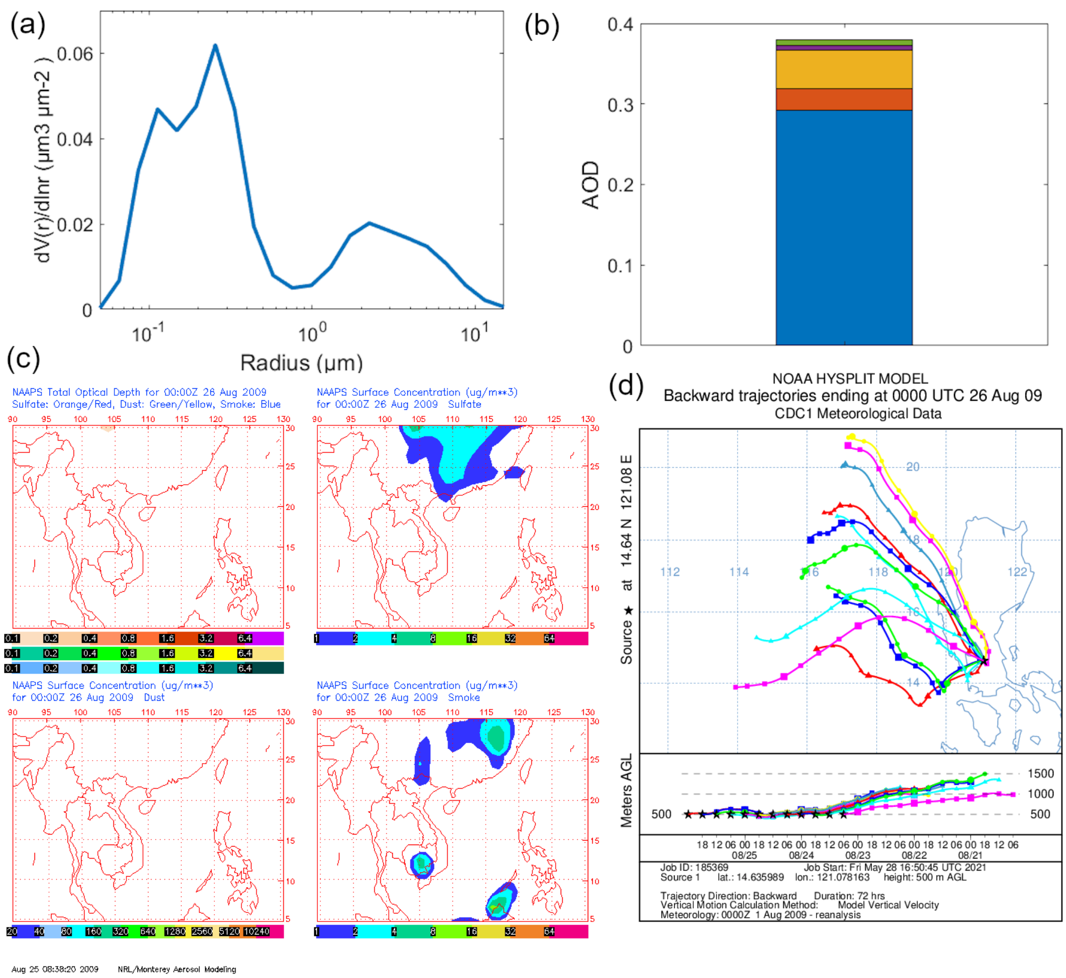

Figure 11: Case study of cloud processing on 26 August 2009. (a) AERONET VSDs at 00:18 UTC, (b) MERRA-2 hourly (green: black carbon, violet: dust, yellow: sea salt, orange: organic carbon, blue: sulfate) compositional contributions to AOD closest in time to 00:18 UTC, (c) NAAPS maps of total and compositional hourly AOD and contributions and smoke surface

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concentrations at 00:00 UTC, and (d) HYSPLIT three-day back-trajectories arriving at Manila Observatory at 00:00 UTC.

3.6 EOF Analysis of AOD in Southeast Asia

To contextualize the analysis of aerosol particle masses in Metro Manila, major regional sources of aerosol particles in Southeast Asia were identified based on the dominant principal components from EOF analysis of AOD. Three principal components (PC, Fig. 12) explained most of the data variance (73.77%) (Fig. 12a) and were all well-separated from each other and are therefore most probably the major distinct aerosol particle sources in the region. They will be the focus of the subsequent discussion.

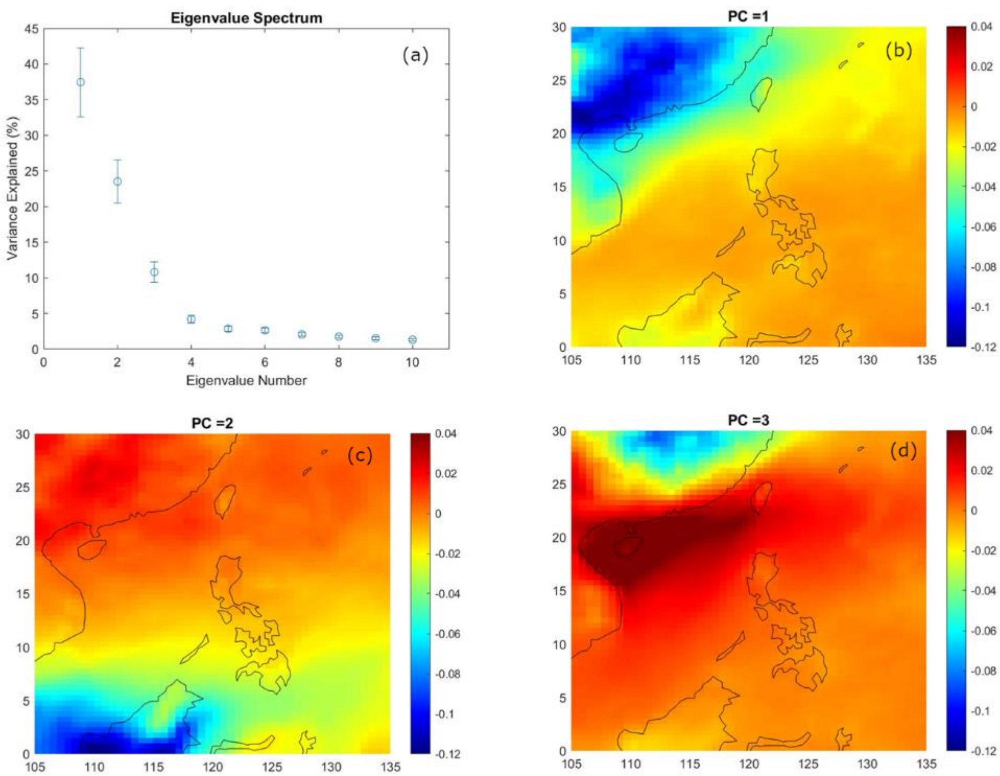

Figure 12: Results of the singular value decomposition. (a) Eigenvalue spectrum of the first ten eigenvalues, (b-d) maps of the coefficients of regression AOD anomalies onto the first three principal components.

The first PC explains 37.46% of the data variance (Fig. 12a) and, based on the map of the regression coefficients (Fig. 12b), separates mainland East Asia from the Philippines and Indonesia. East Asia is a globally recognized source for high AOD (Li et al., 2013), and its contribution to particles in Southeast Asia possibly corresponds to the first PC. The second PC explains 25.51% of the data variance (Fig. 12a) and separates the southern Southeast Asia from northern Southeast Asia at around 15°N (Fig. 12c). Southern Southeast Asia is a known regional

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source of aerosol particles due to biomass burning (Cohen et al., 2017) and could be associated with the second PC. The third PC explains 10.80% of the data variance (Fig. 12a) and separates northern East Asia from southern East Asia mainland and the rest of Southeast Asia (Fig. 12d).

To gain confidence in the association of the PCs with their sources, we present correlation maps between the first three PCs to the fractional contributions of sulfate and organic carbon to AOD for the entire dataset.

The correlation maps of the first PC and the sulfate contribution to AOD (Fig. 13a and 13d) show high and statistically significant correlations (gray areas) in mainland East Asia and Taiwan, parts of western Philippines and Borneo, which are the probable sulfate sources. Clues from the mean monthly wind vector maps in April (Fig. 14a and 14d) and mean monthly AOD in either March or April (Fig. S3c or S3e) most resembling the features of regression map of the first PC (Fig. 12b) and the PC time series peaking in March (Fig. S4) together suggest that the first PC may be associated with air masses that are present around March or April. Emissions sources and meteorology that are dominant during the peak dates in the PC time series offer clues to the attribution of each PC. The Southeast Asia region and the Philippines is influenced by the monsoon systems (Coronas, 1920; Matsumoto et al., 2020) and February to March is the time when the winds are transitioning from the northeasterly to easterly. The first PC could be affected by the easterly winds, which are dominant around March when its PC values peaked. The higher-level winds (free troposphere) (Fig. 14a) in April are from the west in mainland East Asia and are from the east in the Philippines and it is possible that the different wind regimes are distinguishing the sulfate sources in East Asia and the Philippines and beyond. Sulfate is a known product of industry in East Asia (Smith et al., 2011; Li et al., 2017) while the West Luzon and West Visayas islands have large power plants (Jamora et al., 2020).

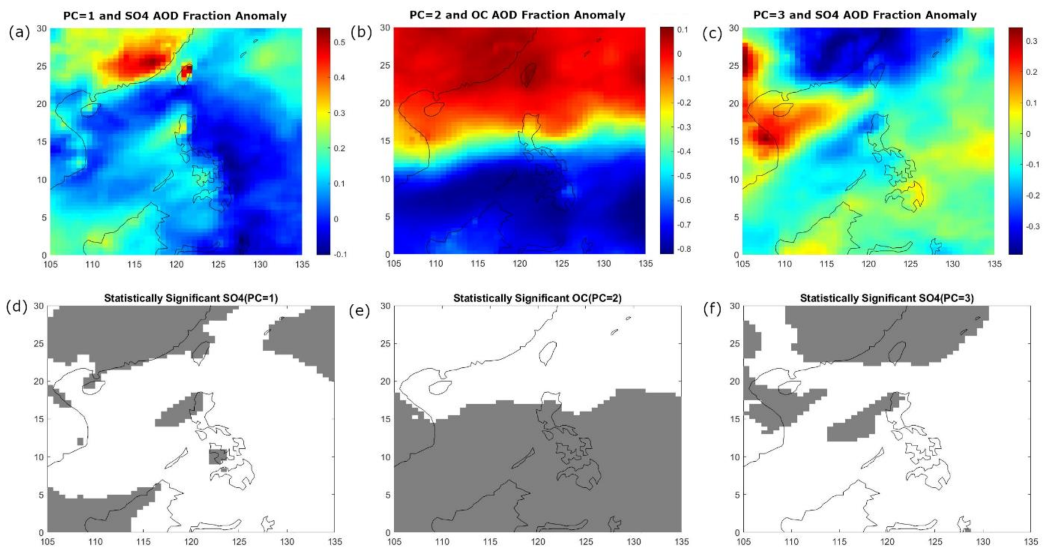

Figure 13: Correlation coefficients of principal components with (a/c) sulfate AOD fraction and (b) organic carbon AOD fraction. Statistically significant (90%, d-f) areas are shaded gray.

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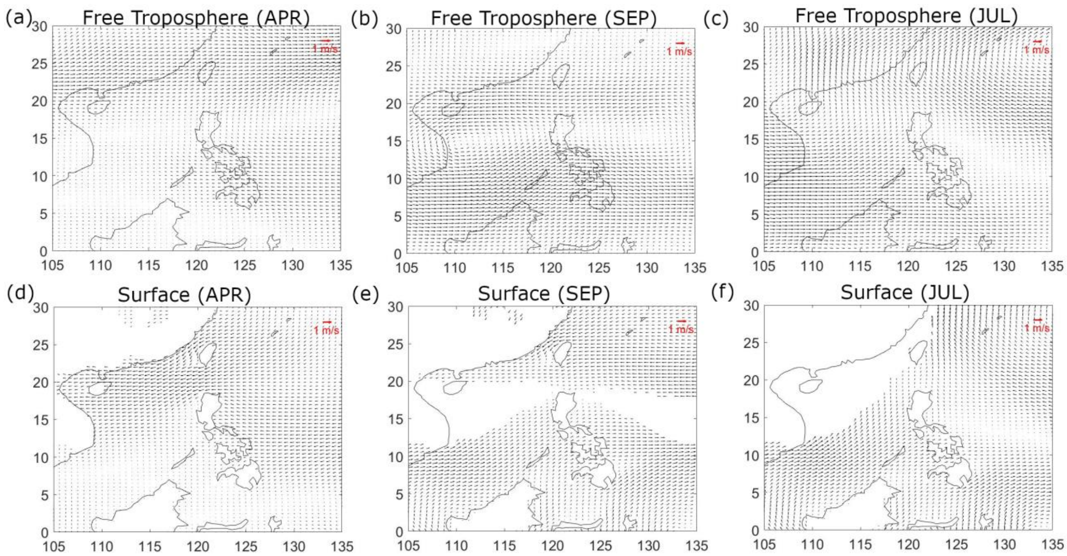

Figure 14: Monthly averaged winds for (a & d) April, (b & e) September, and (c & f) July from MERRA-2 at (725 hPa, a-c) the free troposphere approximate and at (1000 hPa, d-f) the surface.

The correlation maps of the second PC and the OC contribution to AOD (Fig. 13b and 13e) show high and statistically significant correlations from 0°N to 15°N. The large magnitude of the correlation coefficient (gray areas in Fig. 13b) stands out in the southern Southeast Asia and is the potential OC source. In this case, it is known that Indonesia is a major source of biomass burning during its fire season (Glover and Jessup, 1998), and thus the local significance established in the southern Southeast Asia is most likely due to the Indonesia biomass burning source. The burning season in Indonesia is from August to October, and that is the same time when the AOD values peak in the area (Fig. S3h, S3i, and S3j), as well as the peak of the second PC in the time series (Fig. S4). Winds are usually from the southwest and west due to the southwest monsoon from September to October, when the second PC peaked, and thus the second PC may be related to the southwest monsoon. During the same time the surface and free troposphere mean monthly winds (Fig. 14b and 14e) are from the southwest (in the general direction of Indonesia) towards the south portion of Southeast Asia and thus corroborate the observation that the second PC may be highlighting the regional effect of the Indonesia forest fires. Of interest is the line of separation of the northern and southern Southeast Asia in the principal component that is within the area of the monsoon trough (Wang et al., 2007). This line is also evident in the surface and the free troposphere maps where the southwest winds from the area of Indonesia meet the easterlies in north Southeast Asia (Fig. 14b and 14e) and which thus appears to be limiting the dispersion of the biomass burning emissions to southern Southeast Asia.

The third PC was also well correlated to the sulfate AOD fraction though, compared to the first PC correlation maps, there were distinctions between the northern and southern East Asia regions (Fig. 13c and 13f). The local Philippine source still came out in the correlation maps as a significant source. It was not clear from the PC time series (Fig. S4), which showed peaks in the third PC in February, how the dates were related to the PC profile. The free troposphere winds in

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July (Fig. 14c), as well as the AOD monthly mean map in July (Fig. 14c), however, showed more similarities to the third PC regression map. Both showed a delineation between the northern East Asia and southern East Asia (including Hong Kong) features. Mean winds (Fig. 14c) in the free troposphere are from the west, due to the southwest monsoon, in the area around the Philippines, and they were from the northeast in north Southeast Asia. The interface of the winds is within the approximate location of the monsoon trough in July (Wang et al., 2007), and it is thus possible that the monsoon trough is causing the separation of the sulfate sources. This could be investigated further. The monsoon trough has been noted to scavenge aerosol particles from southern Southeast Asia (Reid et al., 2013). It is evident from the analysis that meteorology affects the transport and processing of aerosol particles in region which along with local sources contribute to the aerosol composition in Southeast Asia (Cruz et al., 2019; AzadiAghdam et al., 2019; Braun et al., 2020; Hilario et al., 2020b; Hilario et al., 2022).

4. Conclusion

Metro Manila has both urban and industrial local sources known to contribute to the dominance of fine mode particles in its air (Cruz et al., 2019). Ten years of AERONET data in Manila Observatory suggest that aerosol particles in Metro Manila were mixed in size but with a prevalent fine mode fraction (>50% FMF) throughout the year. Background clean marine aerosol particles (58% of the time) and fine polluted aerosol particles (20% of the time) were the most dominant clear sky day sources impacting the atmospheric column over Metro Manila based on cluster analysis of volume size distributions. The proximity of Metro Manila to the sea both in the east and west along with local sources, transportation being the most prominent, contribute to the prevalence of the marine and fine particles. The prevalence of marine particles could explain the relatively small AOD values in Metro Manila compared to other Southeast Asian megacities (Reid et al., 2013).

Regional sources and meteorology also impact monthly aerosol optical depth trends in Metro Manila from EOF analysis. Biomass burning from Borneo and Sumatra emerged in the study as the second most prevalent regional anthropogenic aerosol particle source in Southeast Asia. Though the monsoon trough limits the dispersion of aerosol particles throughout the entire Southeast Asia, biomass burning emissions impact southern Southeast Asia including Metro Manila during the southwest monsoon (July to September). The monsoon winds facilitate the transport of fine particles during the peak burning season in Borneo and Sumatra (August-September). This is experienced in Metro Manila as higher than usual aerosol particle loadings during the same period. Climatologically, August is also when aerosol optical depth peaked over Metro Manila, concurrent with greatest fine mode fractions that were relatively absorbing and non-hygroscopic possibly due to increased organic and elemental carbon fractional contributions. Though not as strong a source as the Borneo and Sumatra case, the peninsular Southeast Asia burning season (March-April) also contributed to extreme aerosol particle concentrations over Metro Manila.

High aerosol particle loadings due to transported dust, probably from East Asia, were observed in Metro Manila during the transition period between the southwest and northeast monsoons and during the northeast monsoon (December to February). These extreme events are transient because the lowest median aerosol particle loadings of the year were observed during the northeast monsoon when annual wind speeds were highest. Particles then were observed to be largest in diameter, with the greatest coarse fraction contribution and most hygrosocopicity, compared to other months of the year. This is probably due to constituents other than soot, especially aged dust (Kim and Park, 2012; Geng et al., 2014) and sea salt which the northeast winds appear to be bringing in from the general direction of the Luzon Island and the Philippine Sea (West Pacific Ocean).

Cloud processing is one of the cases that were linked to very high aerosol particle loading in Metro Manila. This is associated with sulfate sources, which appear more localized in nature because of a power plant nearby. This sulfate source seems to be distinct from the industrial sulfate air mass from East Asia, which is the most dominant regional aerosol particle source in Southeast Asia (Li et al., 2013). Winds appear to limit the mixing of this notable East Asia air mass with local industrial sources in the region including the Philippines and Indonesia.

The formation of cloud systems in Southeast Asia is complex due to intersecting large- and small-scale mechanisms. Additionally, the interaction of particles and clouds in Southeast Asia is not yet well understood. In Metro Manila, both topography and meteorology affect aerosol particle distribution (Cruz et al., 2023). This baseline study on the aerosol particle characteristics in Metro Manila and in regional Southeast Asia shows how meteorology impacts varied aerosol particle sources (e.g., sulfate, elemental carbon, and organic carbon) and their distribution in the region. This can help in mitigating aerosol particle sources in the region and in the deepening of the understanding of the relationship of aerosol particles, meteorology, and clouds.

Data availability

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Author contributions

GRL and AS designed the experiment. NL, SNU, GRL, GFG, HJO, JBS, and MTC, carried out various aspects of the data collection. GRL, AS, JBS, MOC, MRH, and CC conducted analysis and interpretation of the data. GRL prepared the manuscript draft with contributions from the coauthors. AFA, LDG, MRH, GRL, and AS reviewed and edited the manuscript. AS led the management and funding acquisition. All authors approved the final version of the manuscript.

Competing interests

We declare that Armin Sorooshian is a member of the editorial board of Atmospheric Chemistry and Physics. The peer-review process was guided by an independent editor, and the authors have also no other competing interests to declare.

Acknowledgements

The authors acknowledge support from NASA grant 80NSSC18K0148 in support of the NASA CAMP^{2}Ex project, in addition to ONR grant N00014-21-1-2115. We acknowledge the US Naval Research Laboratory for providing the AERONET instrument. We acknowledge the use of imagery from the NASA Worldview application (https://worldview.earthdata.nasa.gov), part of the NASA Earth Observing System Data and Information System (EOSDIS).

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