- 1 An <u>Emerging</u> Aerosol Climatology via Remote Sensing over Metro Manila, Philippines 2
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23 Abstract

- 24 Aerosol particles in Southeast Asia have a complex life cycle and consequently are challenging
- 25 to characterize. The due to their complex life cycle within the diverse topography and weather in
- 26 the region-complicate the situation. An emerging aerosol climatology was established based on
- 27 AERONET data (December 2009 to October 2018) for clear sky days in Metro Manila,
- 28 Philippines. -Aerosol optical depth (AOD) values were highest infrom August, coinciding with
- 29 the summer southwest monsoon, due to October, partly to from fine particles from urban aerosol
- 30 particles, including soot. Also, August corresponds to, coinciding with the burning season in
- 31 Insular Southeast Asia when smoke is often transported to Metro Manila during the southwest
- 32 <u>monsoon</u>. Clustering of AERONET volume size distributions (VSD) resulted in five aerosol
- 33 particle sources based on the position and magnitude of their peaks in the VSD and the
- 34 contributions of specific particle species to AOD per cluster based on MERRA-2. The clustering
- 35 showed that the majority of aerosol particles above Metro Manila were from a clean marine
- 36 source (58%), which could be related to AOD values there being relatively smaller than in other
- 37 cities in the region. The following are the other particle sources over Metro Manila: fine polluted
- 38 (20%), mixed polluteddust (12%), urban/industrial (5%), and cloud processing (5%).
- 39 Furthermore, MERRA-2 AOD data over Southeast Asia were analyzed using empirical
- 40 orthogonal functions. Along with AOD fractional compositional contributions and wind regimes,
- 41 four dominant aerosol particle air masses emerged: two sulfate air masses from East Asia, an
- 42 organic carbon source from Indonesia, and a sulfate source from the Philippines. Knowing the
- 43 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
- 45 and their impact on regional weather.

46 1. Introduction

47 Although Southeast Asia is one of the most rapidly developing regions in the world, there have 48 been limited studies characterizing with a growing number of extensive research conducted 49 (Reid et al., 2023), there remain knowledge gaps related to aerosol particles in the area (Tsay et 50 al., 2013; Lee et al., 2018; Chen et al., 2020; Amnuaylojaroen, 2023). The region represents a 51 complex geographic, meteorological, and hydrological environment making it challenging to 52 understand aerosol particle characteristics, especially interactions between aerosol particles with 53 their environment (Reid et al., 2013). The island of Luzon in the Philippines in particular is very 54 populated and is characterized by high levels of anthropogenic emissions superimposed on 55 natural emissions from the surrounding waters (AzadiAghdam et al., 2019) and long-range 56 transport of emissions from areas such as Indonesia and East Asia (Braun et al., 2020; Hilario et 57 al., 2020a; Hilario et al., 2020b; Hilario et al., 2021a). Aerosol particle lifecycle in the region is 58 impacted by Philippine weather that is marked by two distinct monsoons, typhoons, the 59 intertropical convergent zone, 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 60 et al., 2021b). Studying this area is informative owing to the wide dynamic range in aerosol 61 particle and weather conditions, which are interconnected. The presence of The overlapping of 62 63 large fraction of cirrus clouds with lower clouds in the area (Hong and Di Girolamo, 2020) 64 makes space-borne remote sensing of aerosol particles very challenging (Reid et al., 2013; Lin et 65 al., 2014). These reasons motivated the NASA Cloud, Aerosol, and Monsoon Processes Philippines Experiment (CAMP²Ex) airborne measurement campaign in 2019 to understand the 66 interaction between tropical meteorology and aerosol particles (Di Girolamo et al., 2015; Reid et 67 68 al., 2023). Prior to the airborne measurements, intensive surface based measurements were conducted as part of the CAMP²Ex weatHEr and CompoSition Monitoring (CHECSM) study 69 70 between July 2018 and October 2019 However, those short terms measurements cannot provide an adequate assessment of aerosol behavior across all seasons and over many years. 71 72 Aerosol climatology studies in different regions have proved beneficial to understand temporal characteristics of aerosol particle concentrations and properties, in addition to identifying 73 74 potential source regions along with interactions with clouds and rainfall (Stevens and Feingold, 75 2009: Li et al., 2011: Tao et al., 2012: Crosbie et al., 2014: Kumar et al., 2015: Alizadeh-76 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 77 78 coverage of aerosol characteristics in specific locations with a column-based perspective from 79 the ground up. Aerosol climatology studies in different regions have proved beneficial to 80 understand temporal characteristics of aerosol particle concentrations and properties, in addition to identifying potential source regions along with interactions with clouds and rainfall (Stevens 81 and Feingold, 2009; Li et al., 2011; Tao et al., 2012; Crosbie et al., 2014; Kumar et al., 2015; 82 83 Alizadeh-Choobari and Gharaylou, 2017; Mora et al., 2017; Aldhaif et al., 2021). in specific 84 locations with a column based perspective from the ground up. To our knowledge, there has not 85 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 86 of 20,800 km⁻² (PSA, 2016; Alas et al., 2018). Studying this area is informative owing to the 87 88 wide dynamic range in acrosol particle and weather conditions, which are interconnected. 89 Aerosol particle lifecycle in the region is impacted by Philippine weather that is marked by two 90 distinct monsoons, typhoons, and impacts from El Niño Southern Oscillation and Madden Julian

91 Oscillation (Cruz et al., 2013; Xian et al., 2013; Reid et al., 2012; Reid et al., 2015; Hilario et al.,
92 2021b).

93 Regional analysis of aerosol particles in Southeast Asia and Asia in general show the prevalence 94 of biomass burning in the region, as well as the larger influence of anthropogenic emissions in 95 East Asia (Nakata et al., 2018). These large prevalent sources may overshadow other relevant but 96 weaker sources in the region, such as local sources. Due to the complex nature of acrosol particles, analysis techniques such as principal component analysis and clustering along with 97 98 recent improvements in gridded datasets help detect spatial and temporal patterns that would 99 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 100 101 Southeast Asia will help in distinguishing local and transported particles that influence the 102 aerosol climatology in Metro Manila. Most of the past studies involving long-term remotely 103 sensed aerosol particle data in Southeast Asia (Cohen, 2014; Nakata et al., 2018; Nguyen et al., 104 2019b) had no specific focus on the Philippines. The Philippines is considered as part of the 105 Maritime Continent (MC), the island nations sub-region of Southeast Asia. The other Southeast Asia sub-region, Peninsular Southeast Asia (PSEA), comprises those nations within the 106 107 continental Asia land mass. These two regions have separate aerosol sources and climate, where 108 MC is dependent on the intertropical convergent zone (ITCZ) and PSEA is dependent on both 109 the ITCZ and monsoon systems (Dong and Fu, 2015). Only the southern part of the Philippines 110 is climatologically part of MC (Ramage, 1971), however, and northwest Philippines, where Metro Manila is located, is affected by the monsoons and tropical cyclones aside from the ITCZ 111 (Chang et al., 2005; Yumul Jr et al., 2010; Bagtasa, 2017). These unique meteorological 112 113 influences and extensive local aerosol particle sources warrant a unique aerosol climatology over 114 Metro Manila, one of a polluted source in a tropical marine environment, and its effects on cloud 115 formation in the area. Aerosol effects on clouds in the marine environment are associated with 116 the largest uncertainties in climate change research (Hendrickson et al., 2021; Wall et al., 2022) 117 and the Philippines was ranked as the 5th country globally as most at risk to climate change and 118 extreme weather from 1997 to 2018 (Eckstein et al., 2018). There have been several surface measurements of aerosol particles made in Metro Manila for the past 20 years (Oanh et al., 2006; 119 120 Bautista VII et al., 2014; Cruz et al., 2019) but columnar ground-based measurements there are just beginning to be established (Dorado et al., 2001; Ong et al., 2016; Cruz et al., 2023). The 121 AERONET sun photometer is one of the first long-term column-based aerosol instruments in 122 123 Metro Manila and the Philippines (Ong et al., 2016). 124 The goal of this study is to use multi-year AERONET data in Manila Observatory along with other complementary datasets (MERRA-2, PERSIANN, MISR, HYSPLIT, and NAAPS) to 125

address the following questions: (1) what are the monthly characteristics of aerosol particles over

127 Metro Manila, Philippines?; (2) what are the possible sources and factors influencing the

128 observed characteristics?; (3) what relationships are evident between aerosol particles and cloud

129 characteristics?; and (4) what are the regional and local aerosol particle air masses that influence

130 Metro Manila?

131

132 **2. Methods**

- 133 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 October2018.
- 136 **Table 1:** Summary of datasets over Metro Manila used in this work covering the period from
- 137 January 2009 to October 2018.

Parameter	Data Source	Spatial Coverage	Data Repository <u>Time Coverage</u>
			(AERONET)
Aerosol Optical Depth (500 nm)	AERONET	14.635°N, 121.078°E	https://aeronet.gsfc.nasa.govJan 2009 - Oct 2018
Asymmetry Factor (440 nm - 1020 nm)	AERONET	14.635°N, 121.078°E	Jan 2009 - Oct 2018
Extinction Angstrom Exponent (440 nm -870 nm)	AERONET	14.635°N, 121.078°E	Jan 2009 - Oct 2018
Fine Mode Fraction	AERONET	14.635°N, 121.078°E	Jan 2009 - Oct 2018
Precipitable Water	AERONET	14.635°N, 121.078°E	Jan 2009 - Oct 2018
Single Scattering Albedo (440 nm - 1020 nm)	AERONET	14.635°N, 121.078°E	Jan 2009 - Oct 2018
Refractive Index (Real and Imaginary; 440 nm - 1020 nm)	AERONET	14.635°N, 121.078°E	Jan 2009 - Oct 2018
Volume Size Distribution	AERONET	14.635°N, 121.078°E	Jan 2009 - Oct 2018
Low Cloud Fraction (MODIS)	MERRA-2	14. 25 3°N - 14. 75 8°N, 120. 9375<u>75</u>°E - 121. 5625 25°E	(MERRA-2) https://disc.gsfc.nasa.gov/Jan 2009 - Dec 2018
Planetary Boundary Layer Height	MERRA-2	14. 25 3°N - 14. 75 8°N, 120. 9375 75°E - 121. <u>562525</u> °E 14. 25 3°N - 14. 75 8°N,	Jan 2009 - Dec 2018
Relative Humidity (975 mb)	MERRA-2	120. 9375<u>75</u>°E - 121. 5625<u>25</u>°E 14. 25 3°N - 14. 75 8°N.	Jan 2009 - Dec 2018
Sea Level Pressure	MERRA-2	120. 9375<u>75</u>°E - 121. 5625<u>25</u>°E 14. 25 3°N - 14. 75 8°N.	Jan 2009 - Dec 2018
Temperature (975 mb)	MERRA-2	120. 9375 75°E - 121. <u>562525</u> °E 14. 25 3°N - 14. 75 8°N,	<u>Jan 2009 - Dec 2018</u>
Wind (975 mb)	MERRA-2	120. 9375 <u>75</u> °E - 121. 5625 <u>25</u> °E	Jan 2009 - Dec 2018
Total Extinction Aerosol Optical Depth (550 nm)	MERRA-2	<u>14.3°N - 14.8°N, 120.75°E - 121.25°E</u>	Jan 2009 - Dec 2018
Sulfate, Black Carbon, Organic Carbon, Dust, and Sea Salt Extinction Aerosol Optical Depth (550 nm)	MERRA-2	<u>14.3°N - 14.8°N, 120.75°E - 121.25°E</u>	Jan 2009 - Dec 2018
Precipitation	PERSIANN	14. <u>53</u> °N - <u>15.014.8</u> °N, 120.75°E - 121.25°E	(PERSIANN) https://chrsdata.eng.uci.edu/Jan 2009 - Dec 2018

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139 **2.1 Datasets**

140 2.1.1 AERONET

141 The central dataset used is that of sun photometer measurements and derived (inversion)

- 142 parameters from the AERONET (Holben et al., 1998) site at the Manila Observatory in Quezon
- 143 City, Philippines (14.64°-N, 121.08°-E, ~70 m. a. s. l.). Direct sunlight extinction measurements
- 144 were made at nominal wavelengths of 340, 380, 440, 500, 675, 870, 940, and 1020 nm, from

145 which aerosol optical depth (AOD) was calculated (except for 940 nm, which is for water vapor)

- 146 (Eck et al., 2013). AOD is a commonly used proxy for aerosol particle loading in the air column
- 147 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
- 149 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.and from which single scattering albedo (SSA)
- and asymmetry factor (AF) are calculated. The AERONET observations were made during clear
- 154 and asymmetry factor (AF) are calculated. The AERONET observations were made during clear 155 sky conditions, which has been shown (Hong and Di Girolamo, 2022) to be able to represent all
- 156 sky conditions.
- 157 For the inversions, four wavelengths (440, 670, 870, and 1020 nm) of the radiometer spectral
- 158 channels were chosen for diffuse radiance measurements and to avoid gas absorption (Dubovik
- 159 et al., 1998). Version 3 Direct Sun and Inversion algorithms (AERONET, 2019; Giles et al.,
- 160 2019) were used with the Almucantar Sky Scan Scenario to derive the following parameters with
- 161 level 2.0 (automatically cloud-cleared and quality controlled datasets with pre- and post-field
- 162 calibrations) data quality: column AOD (500 nm), fine mode fraction (500 nm), extinction
- angstrom exponent (440 870 nm), precipitable water (940 nm), <u>SSAsingle scattering albedo</u>
- 164 (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 modeaerosol particle data (haze and smoke) as well as remove optically thin cirrus clouds in order to
- aerosol particle data (haze and smoke) as well as remove optically thin cirrus clouds in order toretain more aerosol particle measurements in the database (Giles et al., 2019). Cloud screening in
- 168 the version 3 product improves remote sensing measurements in Southeast Asia in general,
- 169 where cirrus clouds are pervasive (Reid et al., 2013). At most, a total of 29,037 direct sun and
- 170 1419 inversion AERONET daytime data points were available between January 2009 and
- 171 October 2018.
- 172 2.1.2 MERRA-2
- 173 Modern Era-Retrospective Analysis for Research and Applications, Version 2 (MERRA-2: 0.5°
- $174 \times 0.625^{\circ}$ approximate resolution) meteorological and aerosol particle composition <u>reanalysis</u> data
- 175 (Bosilovich, 2016; Gelaro et al., 2017; Randles et al., 2017) were acquired for the area around
- 176 Manila Observatory $(14.25^{\circ}N 14.75^{\circ}N, 120.9375^{\circ}E 121.5625^{\circ}E)$. The aerosol reanalysis
- 177 data includes data assimilation of AOD from the Moderate Resolution Imaging
- 178Spectroradiometer (MODIS: Terra, 2000 to present and Aqua, 2002 to present), Advanced Very
- High Resolution Radiometer (AVHRR, 1979-2002), and Multiangle Imaging SpectroRadiometer
 (MISR, 2000-2014) (Buchard et al., 2017; Rizza et al., 2019). The following products were used:
- 181 M2I3NPASM Assimilated Meteorological Fields (3-hourly) for 975 mb level winds,
- 181 M215NPASM Assimilated Meteorological Fields (5-hourly) for 975 lib level winds, 182 temperature, relative humidity, and sea level pressure; M2T1NXFLX Surface Flux Diagnostics
- 183 (1-hourly from 00:30 UTC time-averaged) 2D for planetary boundary layer height; and
- 184 M2T1NXCSP COSP Satellite Simulator (1-hourly from 00:30 UTC time-averaged) for MODIS
- mean low cloud fraction (cloud top pressure > 680 hPa); and M2T1NXAER Aerosol Diagnostics
- 186 (1-hourly from 00:30 UTC time-averaged) for Total AOD and speciated AOD (Sulfate, Black
- 187 <u>Carbon (BC), Organic Carbon (OC), Dust, and Sea Salt</u>).

- 188 MERRA-2 meteorological and aerosol particle composition monthly <u>mean reanalysis</u> data
- 189 (Bosilovich, 2016; Gelaro et al., 2017; Randles et al., 2017) were also acquired for a larger
- 190 region, $(30^{\circ} \times 30^{\circ})$, the Southeast Asia region $(0^{\circ}-N-30^{\circ}N, 105^{\circ}E-135^{\circ}E)$ for the period from
- 191 2009 to 2018. This is within the spatial domain of the CAMP²Ex airborne measurement
- 192 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 500 nm
- AOD and speciated 500 nm AOD (Sulfate, Black Carbon (BC), Organic Carbon (, OC), Dust,
- and Sea Salt) and MERRA-2 instM_3d_ana_Np: Analyzed Meteorological Fields
- 197 (M2IMNPANA) for 1000 hPa and 725 hPa level U and V winds. <u>The total MERRA-2 AOD for</u>
- 198 <u>the region (mean over $30^{\circ} \times 30^{\circ}$ region) was used along with MISR AOD data (mean over $30^{\circ} \times 30^{\circ}$ region) was used along with MISR AOD data (mean over $30^{\circ} \times 30^{\circ}$ region)</u>
- 199 <u>30° region) to assess the influence of long-range sources to the aerosol column over Manila</u>
- 200 <u>Observatory</u>. The monthly meteorological and aerosol particle composition data for the region
- 201 will be used for empirical orthogonal functions, which will be described later.
- 202 2.1.3-MISR
- 203 Monthly AOD data (Level 3 Global Aerosol: $0.5^{\circ} \times 0.5^{\circ}$ spatial resolution) from 2009 to 2018
- are used from the Multi-angle Imaging SpectroRadiometer (MISR), (Diner et al., 2007; Garay et
- 205 al., 2018). Level 3 products are global maps of parameters available in Level 2 (measurements
- 206 derived from the instrument data) products. MISR has relatively more accurate AOD and agrees
- 207 better with AERONET data compared to other satellite products due to its multi-angle
- 208 measurements (Choi et al., 2019; Kuttippurath and Raj, 2021). Monthly median AOD (bin 0)
- 209 were extracted for Southeast Asia (0.25° 30.25°N, 104.75°E 134.75°E) within the CAMP²Ex
- 210 region. They are used for comparison to the AERONET (over Metro Manila) and MERRA 2
- 211 (Southeast Asia) monthly AOD values.
- 212 2.1.4 PERSIANN
- 213 Hourly precipitation data were obtained from the Precipitation Estimation from the Remotely
- 214 Sensed Information using Artificial Neural Networks (PERSIANN) (Nguyen et al., 2019a)
- 215 database of the Center for Hydrometeorology and Remote Sensing (CHRS) at the University of
- 216 California, Irvine (UCI). Hourly data were accumulated for running three-day totals, which were
- 217 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 $(14.5^{\circ}N 15.0^{\circ}N, 120.75^{\circ}E 121.05^{\circ}N)$
- 219 $121.25^{\circ}E$) to the MERRA-2 dataset.

220 <u>2.1.4 MISR</u>

- 221 <u>Monthly 500 nm AOD data (Level 3 Global Aerosol: $0.5^{\circ} \times 0.5^{\circ}$ spatial resolution in the region</u> 222 0.25° N - 30.25^{\circ}N and 104.75^{\circ}E - 134.75^{\circ}E) from 2009 to 2018 are used from the Multi-angle
- Imaging SpectroRadiometer (MISR), (Diner et al., 2007; Garay et al., 2018) as regional
- (Southeast Asia) baseline remote sensing data to support the Manila Observatory AERONET
- data. The regional $(30^{\circ} \times 30^{\circ})$ MISR data was used to confirm regional sources of aerosols that
- 226 may be influencing the AOD over Metro Manila. Level 3 MISR products are global maps of
- parameters available in Level 2 (measurements derived from the instrument data) products.
- 228 MISR is ideal for remote sensing in the CAMP²Ex region because it has an overpass at 10:30
- AM ECT (descending mode) (when cirrus is minimal) and its retrievals have been shown to be
- 230 <u>unimpacted by small cumulus (Zhao et al., 2009)</u>, which are typical in the region. <u>MISR has</u>
- 231 relatively more accurate AOD and agrees better with AERONET data compared to other satellite

- 232 products due to its multi-angle measurements (Choi et al., 2019; Kuttippurath and Raj, 2021).
- 233 The MISR sampling noise is relatively small due to the large domain and seasonal averages that
- are considered in this study. MISR is also the only passive sensor that speciates aerosol particle
- 235 size and shape. All these factors led to the choice of using regional MISR data to associate long-
- 236 range sources influencing AERONET data in Manila Observatory. Monthly mean AOD (bin 0)
- 237 were extracted for Southeast Asia $(0.25^{\circ}N 30.25^{\circ}N, 104.75^{\circ}E 134.75^{\circ}E)$ within the
- 238 <u>CAMP²Ex region. Monthly mean AOD values were then calculated for each 0.5° grid point and</u>
- $\frac{1}{240} \frac{1}{1000} \frac{1}{1000}$
- AOD and angstrom exponent in the CAMP²Ex domain ($6.5^{\circ}N 22.5^{\circ}N$, $116.5^{\circ}E 128.5^{\circ}E$;
- ACD and angstrom exponent in the CAWF Ex domain (0.5 N = 22.5 N, 110.5 E = 128.5 E,
 March 2000 to December 2020) are also used to support the findings from the AERONET data.

243 2.1.5 NAAPS

- Archived <u>maps of total</u> and speciated optical depths <u>along withand</u> surface concentrations of
- sulfate, dust, and smoke for Southeast Asia are used from the Navy Aerosol Analysis and
- Prediction System (NAAPS: $1/3^{\circ} \times 1/3^{\circ}$ spatial resolution) (Lynch et al., 2016), and which is are
- 247 publicly available at https://www.nrlmry.navy.mil/aerosol/. This reanalysis product relies on the
- 248 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 associate possible regional emission
- sources. to extreme aerosol loading events in Manila Observatory. Previous studies have used
- NAAPS data for an overview of aerosol sources in specific regions of interest (Ross et al., 2018;
 Foth et al., 2019; Markowicz et al., 2021; Harenda et al., 2022; Mims III, 2022). More recent
- studies show the need to improve aerosol representation in NAAPS (Edwards et al., 2022), so we
- will use NAAPS qualitatively, together with MERRA-2 compositional AOD data and back-
- trajectories, for an overview of aerosol sources that may contribute to extreme events with high
- 257 AOD from AERONET.
- 258 2.1.6 NASA Worldview<u>HYSPLIT</u>
- 259 Archived maps of cloud fraction (Aqua MODIS and Terra MODIS)-were downloaded from
- 260 NASA Worldview (https://worldview.earthdata.nasa.gov) for events of interest based on 261 AERONET data.
- 261 AER 262

263 2.22.1 Clustering

- 264 Available AERONET VSD data (0.050 μm to 15.000 μm particle radius in 22 logarithmically
- 265 equidistant bins, 1419 data points) were clustered via k-means clustering (Lloyd, 1982). The
- 266 algorithm used was k-means++ (Arthur and Vassilvitskii, 2006). The ideal number of clusters
- 267 was chosen based on relatively highest (>0.5) average silhouette value and the presence of a
- 268 cluster with a second peak in the larger accumulation mode of the VSD. The clusters were
- 269 analyzed based on their associated meteorological conditions and acrosol particle characteristics
- 270 and were classified into air mass types (Table 2) based on previous studies (Pace et al., 2006;
- 271 Kaskaoutis et al., 2007; Sorooshian et al., 2013; Kumar et al., 2014; Sharma et al., 2014; Che et
- 272 al., 2015; Kumar et al., 2015).
- 273 **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	<u>>0.7</u>	Sorooshian et al., 2013
Polluted Fine	>0.1	>1	<u>>0.7</u>	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	-	<u>>0.8</u>	Kaskaoutis et al., 2007

275

276 2.32.1 Extreme Event Analysis

277 Selected types of aerosol particle events-were identified to characterize different types of sources

- 278 and processes impacting aerosol particle columnar properties above Metro Manila. The three
- 279 events are described below.

280 2.3.12.1.1 Smoke Long Range Transport

281 Events related to transported biomass burning/smoke were chosen based on the highest black

282 carbon contribution to total AOD from the MERRA-2 dataset, high smoke contributions to AOD

283 from NAAPS, and a dominant submicrometer peak in the AERONET VSD (Eck et al., 1999)

284 over Metro Manila. Maps of surface smoke contributions from NAAPS as well as backBack-

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.

288 Three provide support for the AERONET monthly aerosol characteristics and the chosen case

studies. Three and seven-day back-trajectories with six-hour resolution were generated based on

290 the NCEP/NCAR reanalysis meteorological dataset and with a resolution of 1° and a vertical

291 wind setting of "model vertical velocity". <u>The three-day data were used to map the density of</u>

trajectories reaching Manila Observatory in each month from 2008 to 2019. The seven-day data

293 <u>were used in the analysis of the case studies.</u> Trajectories were computed for an end point with

- 294 an altitude of 500 m above ground level at the Manila Observatory. This altitude represents the 295 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)7 al., 2020).

298 <u>2.1.7 NASA Worldview</u>

299 Archived maps of cloud fraction (Aqua MODIS and Terra MODIS) over Metro Manila and

- Southeast Asia were downloaded from NASA Worldview (https://worldview.earthdata.nasa.gov)
 for events of interest based on AERONET data.
- 302

303 <u>2.2 Clustering</u>

Available AERONET VSD data (0.050 μm to 15.000 μm particle radius in 22 logarithmically

305 <u>equidistant discrete points, 1419 data points</u>) were clustered via k-means clustering (Lloyd,

- 306 <u>1982</u>). The algorithm used was k-means++ (Arthur and Vassilvitskii, 2006). The ideal number
- 307 <u>of clusters was chosen based on relatively highest (>0.5) average silhouette value and the</u>
- 308 presence of a cluster with a second peak in the larger accumulation mode of the VSD. The
- 309 <u>clusters were analyzed based on their associated meteorological conditions and aerosol particle</u>

310 <u>characteristics and were classified into air mass types (Table 2) based on estimates from previous</u>

- 311 <u>studies (Dubovik et al., 2002; Pace et al., 2006; Kaskaoutis et al., 2007; Kaskaoutis et al., 2009;</u>
- 312 <u>Sorooshian et al., 2013; Kumar et al., 2014; Sharma et al., 2014; Che et al., 2015; Kumar et al.,</u>
- 2015; Deep et al., 2021). The first four mentioned air mass types in Table 2 are the most general,
 and four more classifications based on aerosol particle sources are included. The urban/industrial
- air mass type here refers to local combustion along with long-range transported biomass burning
- 316 (Kaskaoutis et al., 2009). While these classifications are not rigid definitions of air masses, they
- help in understanding the sources that contribute to aerosols in Metro Manila and in identifying
- 318 cases where certain sources are more influential than others.
- 319 **Table 2:** Summary of threshold values of aerosol optical depth (AOD), angstrom exponent (AE),
- 320 <u>fine mode fraction (FMF), and single scattering albedo (SSA) used to identify air mass types.</u>

Air Mass Type	AOD	AE	FMF	<u>SSA</u>	Source
Clean Fine	<u>< 0.1^a</u>	<u>> 1^a</u>	$> 0.7^{a}$	±.	Sorooshian et al., 2013
Polluted Fine	<u>>0.1^a</u>	<u>> 1^a</u>	$\geq 0.7^{a}$	±.	Sorooshian et al., 2013
Clean Coarse	<u>< 0.1^a</u>	<u>< 1^a</u>	<u>< 0.3^a</u>	±.	Sorooshian et al., 2013
Polluted Coarse	<u>> 0.1^a</u>	<u>< 1^a</u>	<u>< 0.3^a</u>	±.	Sorooshian et al., 2013
Clean Marine	$\leq 0.2^{b}$	<u>< 0.9^d</u>	=	<u>0.98^e</u>	Kaskaoutis et al., 2009 Dubovik et al., 2002
Urban/Industrial	$\geq 0.2^{b}$	$\geq 1^d$	±.	$\frac{0.9}{0.98^{e}}$	Kaskaoutis et al., 2009 Dubovik et al., 2002
Biomass Burning	=	<u>> 1.4^a</u>	Ξ	$\frac{0.89}{0.95^{e}}$	<u>Deep et al., 2021</u> <u>Dubovik et al., 2002</u>
Desert Dust	<u>> 0.3^c</u>	<u>< 1^d</u>	±.	<u>0.92-</u> <u>0.93^e</u>	Kaskaoutis et al., 2009 Deep et al., 2021 Dubovik et al., 2002
^a from MODIS	_	^c AOD at	<u>400 nm</u>	<u>e SSA at 440 nm</u>	
^b AOD at 500 nm	-	^d AE at 38	<u>80 nm to 87</u>	-	

321

322 <u>2.3 Extreme Event Analysis</u>

Aerosol particle events based on the three clusters with the highest VSD concentrations were
 identified to characterize different types of sources and processes impacting aerosol particle

325 <u>columnar properties above Metro Manila. The three events are described below.</u>

326 <u>2.3.1 Smoke Long Range Transport</u>

327 Events related to transported biomass burning/smoke were chosen from the AERONET VSD

data that were clustered as urban/industrial (with a dominant submicrometer peak) (Eck et al.,

329 1999) over Metro Manila. Cases with the highest black carbon contribution to total AOD from

330 the MERRA-2 dataset were considered. Maps from NAAPS of high smoke contributions to

AOD and surface smoke contributions in the direction of back-trajectories HYSPLIT were used

332 <u>to provide support for the likely source and transport pathway for the smoke cases.</u>

333 2.3.2 Dust Long Range Transport

- A dust transport case <u>over Metro Manila</u> was identified based on the highest dust contribution to
- AOD from the MERRA-2 dataset, high dust contributions to AOD from NAAPS, and AERONET
- 336 <u>VSD dust cluster (with</u> an enhanced coarse peak in the AERONET VSD (compared to the
- submicrometer fraction) (Eck et al., 1999)-over Metro Manila., the highest dust contribution to
- AOD from the MERRA-2 dataset, and high dust contributions to AOD from NAAPS. Surface
- dust concentrations from NAAPS along with<u>the</u> HYSPLIT back-trajectories confirmedimproved
- 340 the plausibility of dust for this case.
- 341 2.3.3 Cloud Processing
- 342 Cloud processing events were identified based on bimodal submicrometer VSDs (Eck et al.,
- 343 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 <u>qualitatively</u> with <u>MODIS (Aqua and Terra)</u>
- 346 imagery from NASA Worldview in the path of air parcels reaching Metro Manila based on
- 347 HYSPLIT back-trajectories.
- 348

349 **2.4 Empirical Orthogonal Functions**

- Regional analysis of aerosol particles in Southeast Asia and Asia in general show the prevalence
- 351 of biomass burning in the region, as well as the larger influence of anthropogenic emissions in
- 352 East Asia (Nakata et al., 2018). These large prevalent sources may overshadow other relevant but
- 353 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. Empirical orthogonal function (EOF) analysis was
- 360 performed to be able to associate the air mass clusters identified earlier with regional scale
- 361 aerosol particle sources.
- 362 <u>To contextualize the analysis of aerosol particle masses in Metro Manila, major regional sources</u>
- 363 of aerosol particles in Southeast Asia were identified based on the dominant principal
- 364 components from empirical orthogonal (EOF) analysis of AOD. EOF analysis was done on the
- 365 monthly AOD data (January 2009 to December 2018) from MERRA-2 for the Southeast Asia
- 366 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; like MISR; the
- 368 MERRA-2 reanalysis data alleviatedataset alleviates this issue.
- 369 The monthly MERRA-2 AOD maps ($0^{\circ} 30^{\circ}$ N, 105° E 135° E with 0.5° latitude and 0.625°
- 370 longitude resolution) (Lat: 61 rows x Lon: 49 columns) for the Southeast Asia region (presented
- 371 subsequently) were first deseasonalized. Then, the AOD anomaly per grid per year (of the 120
- 372 months) was calculated by subtracting the monthly mean AOD from each value of a given month
- 373 (Li et al., 2013). The anomalies per grid were weighted depending on their latitude by
- 374 multiplying the anomalies by the square root of the cosine of their latitudes.

- EOF, specifically singular value decomposition (SVD), analysis (Björnsson and Venegas, 1997)
- 376 was then performed. To prepare the data for the analysis, they were transformed such that the
- 377 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 <u>outputted</u> the eigenvalue (S) and eigenvector (U: principal components and V:
- 380 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
- 382 standard deviation per PC time series (120 months).
- 383 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
- 385 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
- 388 plotted. Each grid on the Southeast Asia map was colored based on the calculated regression
 389 coefficient value.
- 390

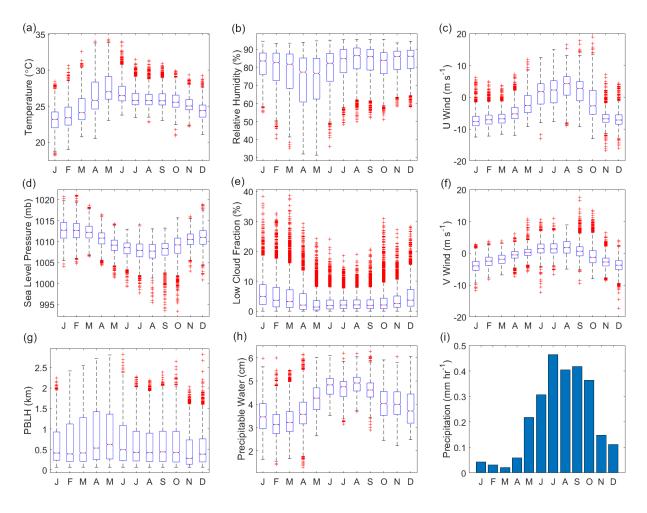
391 **2.5 Correlations**

- 392 The first three standardized PCs of AOD anomalies were correlated to deseasonalized
- 393 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 $\sim n = 100$
- 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
- 397 significant (90% confidence).
- 398

399 **3 Results and Discussion**

400 **3.1 Meteorology and Atmospheric Circulation**

- 401 Knowledge of monthly behavior of weather in the study region helps interpretation of aerosol 402 particle data. Philippine climate is influenced both by the winter northeast monsoon 403 (~November to April, Amihan) and the summer southwest monsoon (~May to October, Habagat) 404 (Coronas, 1920; Flores, 1969; Matsumoto et al., 2020). Median 3-hourly temperatures at 975 mb 405 per month (MERRA-2, 975 mb) (Fig. 1a) ranged from 23.2 °C in January during the winter 406 northeast monsoon, to 27.0 °C in May during the transition from the summer season, as defined 407 in (Bañares et al., 2021), to the southwest monsoon. May was also the month with the lowest 408 median 3-hourly relative humidity (76.6%) (MERRA-2, 975 mb) (Fig. 1b). The highest median 409 level of relative humidity at 975 mb for a month was in August (86.5 %) during the summer 410 southwest monsoon, which is also the time of the year (June to August) when rainfall peaks in 411 the region where the sampling station (Manila Observatory) is located (Coronas, 1920; Cruz et 412 al., 2013). The highest mean hourly precipitation (Fig. 1i) per month was from July (0.46 mm hr⁻
- 413 ¹) to September (0.42 mm hr⁻¹), while March exhibited the lowest mean hourly rainfall (0.02 mm
- 414 hr⁻¹). Like relative humidity and precipitation, median precipitable water (from available
- AERONET data) (Fig. of 513 points in August, 4015 points in February, and 5049 points in
- 416 <u>March) (Fig.</u> 1h) was highest in August (4.9 cm) and lowest in February and March (3.1 cm and
- 417 3.2 cm, respectively).
- 418



- 419
- 420

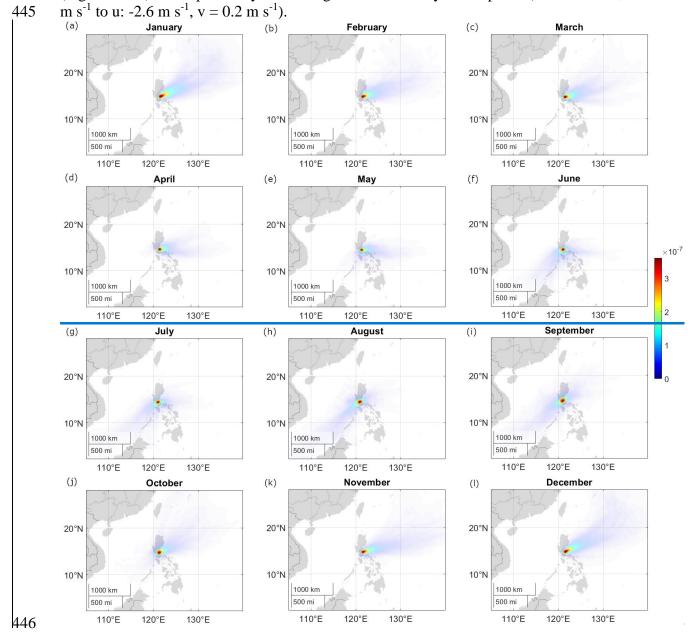
Figure 1: Monthly characteristics of meteorological parameters for Metro Manila, Philippines
based on data between January 2009 and October 2018. MERRA-2 parameters: (a) temperature
at 975 mb, (b) relative humidity at 975 mb, (c/f) u and v wind at 975 mb, (d) sea level pressure,
(g) planetary boundary layer height (PBLH), (e) low cloud fraction; (cloud top pressure > 680
hPa); AERONET: (h) precipitable water; (data counts per month Jan: 2131, Feb: 4015, Mar:
5049, Apr: 5844, May: 3448, Jun: 1696, Jul: 652, Aug: 513, Sep: 753, Oct: 1700, Nov: 2084,
Dec: 1449); PERSIANN: (i) mean hourly precipitation per month.

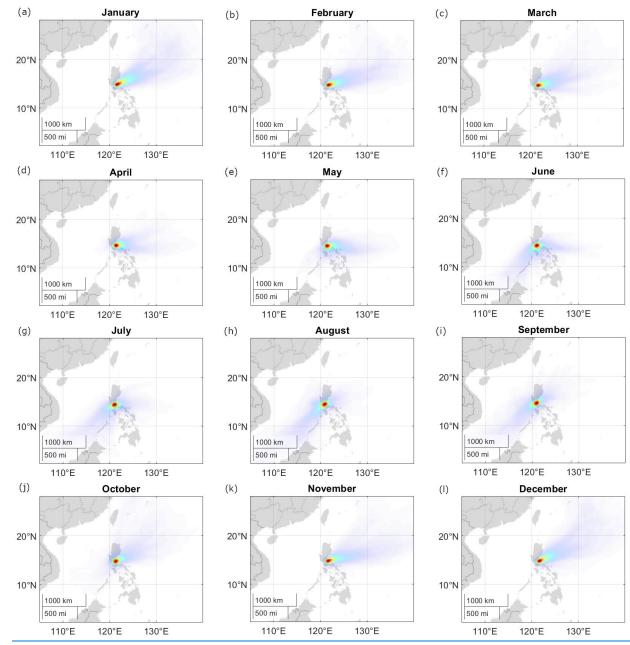
428

429 The lowest 3-hourly median pressures (MERRA-2) were observed (Fig. 1d) between July and

- 430 September during the southwest monsoon season (~985.2 985.8 mb). This is also the time
- 431 when the most number of tropical cyclones pass the island of Luzon (Wu and Choy, 2016). The
- 432 highest 3-hourly median pressures (988.1 990.0 mb) were during the winter northeast
- 433 monsoon.
- 434 Median winds (MERRA-2) were from the south/southwest direction from June to September
- 435 (Fig. 1c and 1f), associated with the summer southwesterly monsoon. HYSPLIT back-
- 436 trajectories show the same wind pattern (Fig. 2f to 2i). The highest median 3-hourly wind speeds
- 437 (MERRA-2) (Fig. 1c and 1f) during the southwest monsoon were recorded for August (u: 4.2 m
- 438 s⁻¹ and v: 1.7 m s⁻¹). Median winds begin to transition in October and November (to the northeast

- 439 monsoon: Amihan) (Fig. 2j and 2k) coming from the east/northeast and maintained until
- 440 February (Fig. 2b), which is towards the end of the winter northeast monsoon. There were
- 441 generally higher wind speeds and the highest median 3-hourly wind speeds of the year
- 442 (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)
- 444 (Fig. 2c to 2e) accompanied by decreasing median 3-hourly wind speeds ($u = -6.8 \text{ m s}^{-1}$, v = -1.9





447

Figure 2: Density plots of <u>HYSPLIT</u> trajectories reaching Manila Observatory per month from 2009 to 2018. Red denotes areas with the greatest number of back trajectories within a 100 km

450 radius. The colors represent density value contributions to Matlab-calculated cumulative

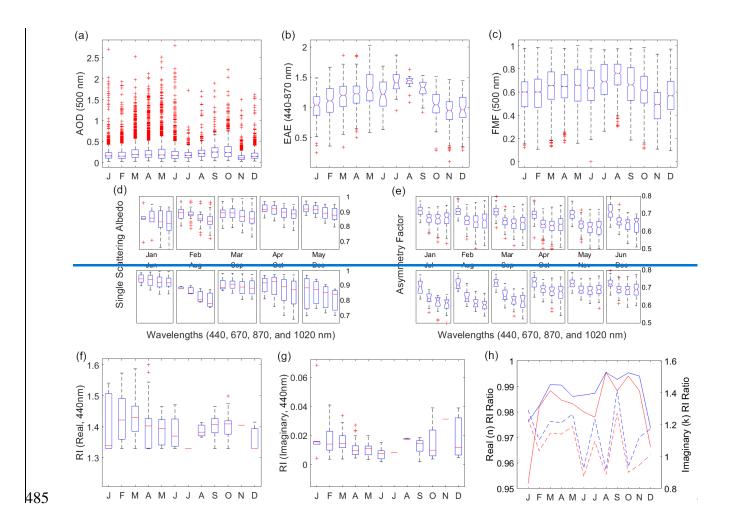
- 451 probability distribution surfaces (100 km radius) from coordinates of three-day back trajectories
- 452 of the specific months.
- 453 The transition times between the monsoons (when the wind directions shift and wind speeds
- 454 change) are also the times of the highest (May, Fig. 1g, 621.2 m) and lowest (November, Fig. 1g,
- 455 279.6 m) median planetary boundary layer heights (MERRA-2). The median planetary boundary
- 456 layer height was highest during the period (May) of highest temperatures, lowest relative
- 457 humidity, reduced air pressure, and lowest monthly median low cloud fraction (MERRA-2) (Fig.
- 458 1e) (1.4 %). The lowest monthly median planetary boundary layer height was observed during

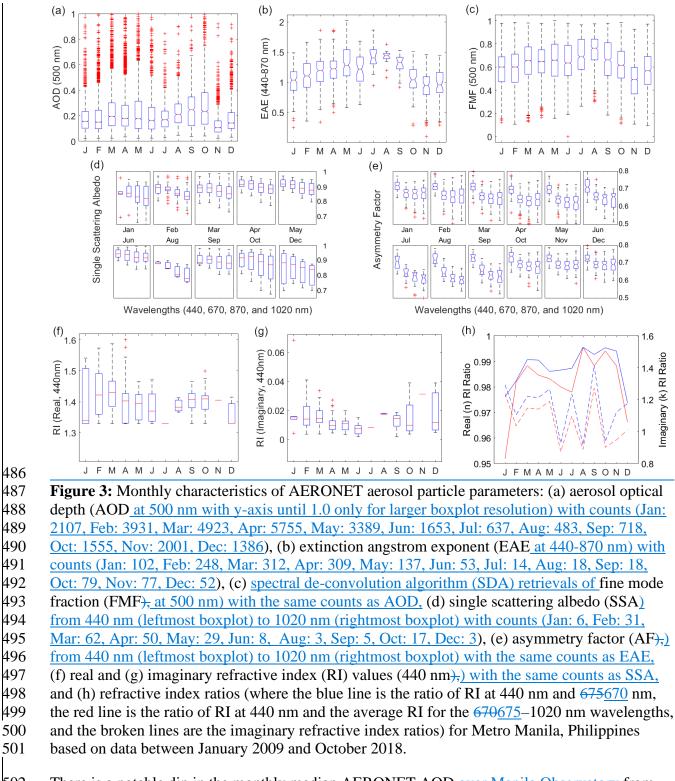
- the period (November) when temperatures were beginning to cool and air pressure was rising.
- 460 The monthly maximum low cloud fraction was lowest in July (18.5 %) during the summer
- 461 southwest monsoon while the monthly median and monthly maximum low cloud fractions
- 462 (MERRA-2) (Fig. 1e) were highest (38.3 % max, 4.9 % median) in January during the winter463 northeast monsoon.
- 464

465 **3.2 Aerosol Particle Characteristics**

- 466 3.2.1 Aerosol Optical Depth
- 467 Monthly median AOD (AERONET, 500 nm) (Fig. 3a) over the Manila Observatory was highest
- 468 from August (0.21) to October (0.23) around the time of the summer monsoon when winds were
- 469 coming from the southwest (Figs. 2h to 2i) (Holben et al., 2001). This is the same time of year
- 470 when biomass burning activities occur in the <u>Indonesian</u> region southwest of Metro Manila.
- 471 (Glover and Jessup, 1998; Kiely et al., 2019; Cahyono et al., 2022). Studies have shown that
- AOD in the Philippines increases during the biomass burning season in Indonesia (Nguyen et al.,
- 473 <u>2019b; Caido et al., 2022). Regional AOD (550 nm)</u> over the larger Southeast Asia
- 474 regiondomain from MISR and MERRA-2 (Fig. 4) had a similarly large peak from around the
- 475 <u>same time beginning in</u> September to<u>until</u> October which, however, was second only in
- 476 magnitude to a March peak, which is influenced by biomass burning in Peninsular Southeast
- 477 Asia (PSEA) (Gautam et al., 2013; Hyer et al., 2013<u>; Dong and Fu, 2015; Wang et al., 2015;</u>
- 478 <u>Yang et al., 2022</u>). This is consistent with the peak in speciated AOD due to fine (radii <0.7
- 479 μm), spherical, and absorbing aerosols that were observed by MISR from March to April (Fig.
- 480 <u>S1).</u> This larger peak in March, attributed to PSEA (which is ~2000 km west of the
- 481 Philippines,), was not as prevalent in the AERONET AOD data over Manila Observatory in
- 482 <u>Metro Manila</u> due to the dominant easterly winds in the Philippines in March (Fig. 2c) and more
- 483 localized sources.

484





502 There is a notable dip in the monthly median AERONET AOD <u>over Manila Observatory</u> from

the peak in October to the lowest monthly median AOD (0.11) in November, (Fig. 3a), just

slightly above defined background levels (<0.1) (Holben et al., 2001), when the windspeeds

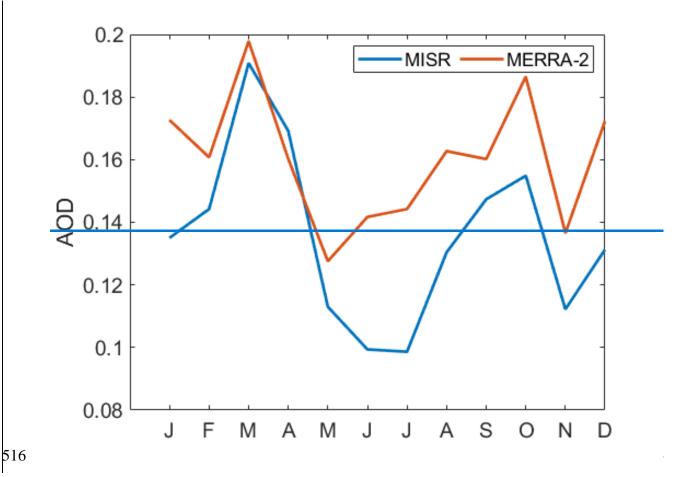
505 were picking up and were coming from the east to northeast directions (Fig. 2k) in the direction

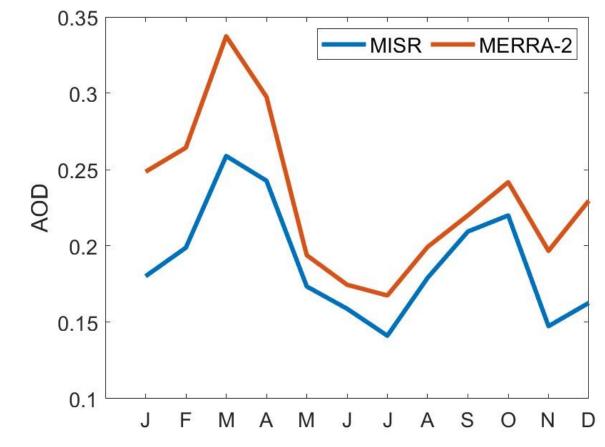
506 of the Philippine Sea and the West Pacific Ocean. This dip was also observed in the regional

507 AOD data (MISR and MERRA-2, Fig. 4). This dip was also observed in the regional $(30^{\circ} \times 30^{\circ})$ 508 AOD data (MISR and MERRA-2, Fig. 4). This is most probably due to the decrease in the AOD 509 contribution from fine (radii <0.7 µm) and spherical particles based on size speciated MISR 510 AOD (Fig. S1). Larger and non-spherical particle contributions to AOD increase in November in 511 the Southeast Asia region. The MERRA-2 AOD is relatively higher than the MISR AOD 512 probably due to assimilation of MODIS data into MERRA-2. Studies in Asia (Xiao et al., 2009; 513 Oi et al., 2013: Choi et al., 2019) have observed relatively higher MODIS AOD compared to

- 513 Qi et al., 2013; Choi et al., 2019) have observed relatively higher MODIS AOD compared to
- 514 MISR AOD.

515





517

518 **Figure 4:** Monthly <u>medianmean</u> AOD (550 nm) in Southeast Asia ($30^{\circ} \times 30^{\circ}$) from 2009 to 2018 from MISR (blue line) and MERRA-2 (red line).

520 There were 338 instances (~1.2 % of the time based on the total number of 28,538 valid

521 AERONET AOD data points) of AOD values exceeding 1, indicative of heavy aerosol particle

522 loading (Huang et al., 2021). Because AOD is extrinsic (it depends on mass), AOD describes

523 total aerosol particle loading and we examine other aerosol particle parameters from AERONET

to make more informed inferences about size and composition.

525 3.2.2 Extinction Angstrom Exponent and Fine Mode Fraction

526 The extinction angstrom exponent (EAE) relates the extinction of light at specific wavelengths

527 and is indicative of aerosol particle size (Ångström, 1929). The EAE is usually greater for

528 smaller particles (~4 for very small particles that undergo Rayleigh scattering and 0 for particles

529 as large as cloud drops) (Bergstrom et al., 2007), except for when the coarse mode has a large

530 impact on the angstrom exponent (Schuster et al., 2006). The EAE is usually greater for smaller

particles (~4 for very small particles that undergo Rayleigh scattering, > 2 for small particles, <

532 <u>1 for large particles like sea salt and dust, and 0 for particles as large as cloud drops) (Schuster et</u>

533 <u>al., 2006; Bergstrom et al., 2007).</u> The highest monthly median EAE (Fig. 3b) from 2009 to 2018

534 over the Manila Observatory was observed from July (~1.4) to September (~1.3), during the 535 southwest monsoon. This period is associated with the biomass burning southwest of the 536Philippines (Oanh et al., 2018; Stahl et al., 2021; Crosbie et al., 2022). The median (per month)537FAF

- 537 EAE ranged from ~0.9 in November to ~1.4 in August, a range which is within the values from 538 previous studies collected from mixed sites and urban/industrial areas with both fine and coarse
- particles (Eck et al., 2005; Giles et al., 2012). The high EAE over Manila Observatory from July
- 539 particles (Eck et al., 2005, Glies et al., 2012). <u>The high EAE over Walling Observatory from Jur</u> 540 to September is probably regional in nature based on the MISR data showing increased EAE
- 541 with increased AOD from fine, spherical, and absorptive particles (Fig. S1) in Southeast Asia
- 542 during the same months. The high EAE over Manila Observatory from July to September is
- 543 consistent with the regional (30° latitude x 30° longitude) MISR data that shows increased AOD
- 544 from fine, spherical, and absorptive particles (Fig. S1) in Southeast Asia during the same months.
- 545 This suggests that the high EAE observed at the Manila Observatory during these months is not
- 546 <u>necessarily from local sources.</u>
- EAE increases with AOD (Fig. $\frac{\$4\$2}{\$}$), which means that the greater particle loading is
- 548 contributed by smaller particles (Smirnov et al., 2002). Of the high loading cases (AOD >1),
- 549 <u>over Manila Observatory</u>, the EAE values ranged fromwere mostly greater than 0.6 to 1.6,8
- indicating fine mode particles (Che et al., 2015). The EAE values in August were the highest
- 551 compared to other months including having the highest minimum value of any month (0.71)
- (Fig. $\frac{S1S2}{C}$), 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, which again may be regional in nature with MISP EAE also lawset during this time with increased AOD from larger
- 554 regional in nature with MISR EAE also lowest during this time with increased AOD from larger
 555 and non-spherical particles (Fig. S1).
- and non-spherical particles (112. 51).
- 556 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
- 559 with the EAE values discussed earlier with the prevalence of smaller particles in August and
- 560 larger particles in November. In August (Fig. 2h) the southwest monsoon is known to coincide
- 561 with <u>transported the transporting of</u> fine smoke particles to Luzon. In November (Fig. 2k), the
- 562 prevalent winds may have already shifted to easterly (Matsumoto et al., 2020) implying more
- 563 marine-related sources associated with coarser particles.
- 564 3.2.3 Single Scattering Albedo
- The single scattering albedo (SSA) is the most important aerosol particle parameter determining
- 566 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
- 569 aerosol particles while more absorbing aerosol particles will have lower SSA values; values
- 570 range from 1 (reflective) to 0 (absorbing). Monthly median SSA values were largest in June
- 571 (0.94, at 440 nm), suggesting the presence of more reflective aerosol particles, and smallest in
- August (0.78,88 at 440 nm and 0.78 at 1020 nm) suggesting more absorptive particles that are
- 573 <u>similar in range to the SSA of biomass burning particles (Table 2)</u>. August is when biomass
- 574 burning is prevalent to the southwest of the Philippines and associated with soot particles that are 575 absorptive
- 575 absorptive.
- 576 The sensitivity of SSA to different wavelengths depends on the type of aerosol particles present.
- 577 More specifically, aerosol particle size and refractive index (which is related to aerosol particle
- 578 composition) both affect the SSA (Dubovik and King, 2000; Bergstrom et al., 2007; Moosmüller

579 and Sorensen, 2018). For dust-type particles, SSA increases with wavelength because of lower 580 dust absorption in the higher visible to infrared wavelengths (Dubovik et al., 2002), while for

- 581 urban particles (including black carbon), which absorb light at longer wavelengths, SSA
- 582 decreases with wavelength (Reid et al., 1998; Bergstrom et al., 2002). The presence of organic
- 583 carbon may affect this spectral dependence; however, because organic particles absorb in the
- 584 UV, this lowers SSA at wavelengths shorter than 440 nm (Kirchstetter et al., 2004). Monthly
- 585 median SSA generally decreased with increasing wavelength for all months with available data
- 586 (Fig. 3d) presumably due to the influence of more urban particles in contrast to dust.
- 587 Noteworthy though are the monsoon transition months of April, September, and October (Fig.
- 588 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
- 590 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
- 592 sources associated with delosor particles necessitates looking at other available delosor particles
- 593 parameters, discussed subsequently.

594 3.2.4 Asymmetry Factor

- 595 The asymmetry factor quantifies the direction of scattering of light due to aerosol particles, with
- 596 values ranging from -1 (back scatter) to 0 (uniform scattering) to 1 (forward scatter). It is
- 597 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
- 600 al., 2018).

601 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.,
- 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
- 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
- 610 AERONET data at the 675, 870, and 1020 nm were similar across months (Fig. 3e). The monthly
- 611 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
- 614 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
- 616 local source due mostly to vehicles (Cruz et al., 2019).
- 617 The monthly median asymmetry factor in Metro Manila was greatest towards the end of the year
- 618 (October to December) for all the wavelengths, suggesting larger particles when winds (Figs. 2j
- to 21) come from the Philippine Sea in the northeast. It was in March and April that the monthly
- 620 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
- 624 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.,
- 626 2018). The unexpected low asymmetry factor values in August, however, are probably because
- 627 of the source of the particles. August had the highest relative humidity and precipitable water
- 628 (Fig. 1b and 1h) but is also when the back-trajectories (Fig. 2h) were from the southwest,
- 629 possibly affected by the Indonesia fires, which could have transported more non-hygroscopic
- 630 fine particles.
- 631 Fine particles have been observed to exhibit decreasing asymmetry factors with increasing
- 632 wavelength (Bergstrom et al., 2003). This trend is observed in all the months for the monthly
- 633 median asymmetry factors (Fig. 3e) suggesting the predominance of smaller aerosol particles.
- 634 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
- 636 particles are the probable dominant particles during this time. They are usually composed of
- 637 hygroscopic inorganics, non-hygroscopic soot, and relative non-hygroscopic organic fractions
- 638 (Petters et al., 2009). Knowing the composition of biomass burning particles over the study
- 639 region will help in the understanding of hygroscopicity and its impacts on radiation.

640 3.2.5 Refractive Index

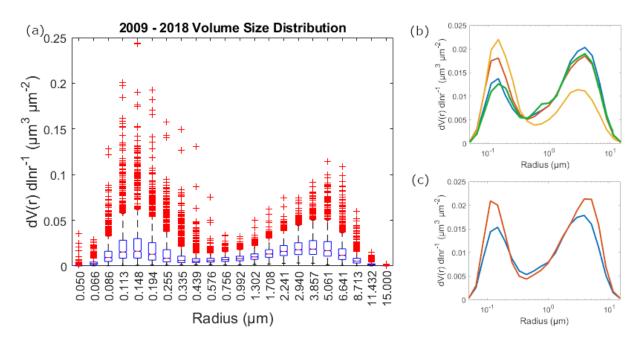
- 641 Refractive index is an intrinsic parameter as it does not depend on the mass or the size of
- 642 particles, and thus can be used to infer aerosol particle composition (Schuster et al., 2016). For
- 643 the case of the AERONET data, which include refractive index values that are insensitive to
- 644 <u>coarse particles (Sinyuk et al., 2020), the focus of the discussion will be for fine mode particles</u>
- 645 <u>and may be limited when coarse particles are involved</u>. Refractive index measurements are
- 646 complex since they include real and imaginary parts related to light scattering and absorption,
- 647 respectively. All aerosol particles scatter light but only certain types absorb light significantly.
- 648 The most prominent particle absorbers in the atmosphere are soot carbon, brown carbon (organic 649 carbon that absorbs light), and free iron from dust (hematite and goethite in the ultraviolet to
- 650 mid-visible) (Schuster et al., 2016). For this study, we examine refractive index values at 440 nm
- 651 wavelength because this is the wavelength used to calculate SSA (Andrews et al., 2017). For this
- study, we examine refractive index values at 440 nm wavelength. Pure sources of soot carbon
- have the highest real refractive index values (\sim 1.85) as well as the highest imaginary refractive
- 654 index (~0.71), both independent of wavelength (Koven and Fung, 2006; Van Beelen et al.,
- 655 2014). Brown carbon and dust have relatively lower real refractive index values at 440 nm
- $(\sim 1.57 \text{ and } \sim 1.54)$ and imaginary refractive index values (~ 0.063 and ~ 0.008) that decrease with
- 657 increasing wavelength (Xie et al., 2017).
- In this study the range of the monthly median real refractive index values (440 nm) was from
- 659 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 streagener
- indices over the Manila Observatory can be due to the presence of more water in the atmospherein 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

- 667 important contributor. The greatest change in the monthly median real refractive index with
- 668 increasing wavelength also was observed in December (Fig. 3h), possibly due the increased
- 669 fractional contribution of constituents other than soot carbon (because the real refractive index of
- 670 soot carbon is invariant with wavelength). Noteworthy as well is the month of August (Fig. 3f),
- 671 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 isalso the month when long-range biomass burning emissions are observed to be highest, and
- 675 when the real refractive index values would otherwise be expected to be highest.
- when the real refractive index values would otherwise be expected to be nighest.
- 676 Water content seems to play a significant role in the real refractive index values in Manila.
- 677 March, when the monthly median real refractive index values are highest (Fig. 3f), is when
- 678 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)
- 680 were observed (Fig. 3f). March was when there was a relatively small change in real refractive
- 681 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.,
- 683 2014). Looking more closely at the imaginary refractive index values will help elucidate this
- 684 issue.
- 685 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 686 687 mentioned earlier because of the mixed nature of the sampling site with contributions from 688 brown carbon and dust. The highest imaginary refractive index values in September and 689 December suggest the greatest fractional contribution of soot because the highest imaginary 690 refractive index values are associated with soot. These are also similar in magnitude to biomass 691 burning particles in the Amazon (0.013) (Guyon et al., 2003). The key distinction between soot 692 carbon and other major absorbers (brown carbon and dust) is that its imaginary refractive index 693 is invariant with wavelength. Both brown carbon and dust exhibit a decrease in the imaginary 694 refractive index with increasing wavelength (Xie et al., 2017). The ratios of imaginary refractive 695 index values (440 nm to average of 670-1020 nm) (Fig. 3h) show a relative invariance with 696 wavelength (ranging from 0.88 to 1.4), which indicates the dominance of soot as the major 697 absorber in the region (Eck et al., 2003). While observed wavelength invariance points to high 698 soot contributions, the size of the particles can help distinguish between brown carbon, which 699 reside mainly in the fine mode, and dust sources, which yield more coarse particles (Schuster et 700 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 701 702 index varied most with wavelength (1.4 ratio of the imaginary refractive index at 440 nm and the 703 imaginary refractive index average for 670 nm to 1020 nm in Fig. 3h) possibly with greater 704 absolute contributions from brown carbon, even with the highest soot carbon fractional 705 contributions. Brown carbon has been observed both from primary and aged aerosol particle 706 emissions from biomass burning (Saleh et al., 2013). As noted earlier, December also had the 707 highest imaginary refractive index values as well as relatively coarser particles, possibly due to 708 larger dust absolute contributions even with the highest soot carbon fraction contributions. The 709 lowest monthly median imaginary refractive index values in June, on the other hand, when fine 710 mode particles prevail suggest highest fractional contributions of brown carbon relative to other 711 months (Fig. 3h).

712 3.2.6 Volume Size Distributions

713 The volume size distribution (VSD) is another way to be able to more deeply characterize 714 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 715 716 VSD for the entire dataset (Fig. 5a). The fine mode median values peaked in the accumulation 717 mode at 0.148 μ m particle radius while the coarse mode median values peaked at 3.857 μ m-718 (Fig. 5a and Table S1). The median coarse mode amplitudes and volume concentrations were 719 higher than the fine mode amplitudes and volume concentrations for most of the year (DJF, 720 MAM, and SON, Fig. 5b and Table S1), except during the southwest monsoon (JJA) when the 721 fine mode amplitude and volume concentration was higher. This is consistent with observations 722 earlier of fine mode prevalence during the southwest monsoon. Median VSD amplitudes (Fig. 723 5c) were greater in the afternoon, with higher peaks and volume concentrations for both the fine 724 and coarse modes, compared to the morning. There was a slightly larger coarse median 725 amplitude and volume concentration, compared to the accumulation mode median amplitude and 726 volume concentration, for both the morning and afternoon size distributions. While the VSDs 727 confirm several observations based on the analysis of the aerosol particle parameters presented 728 earlier, not much further information is gained especially regarding chemical composition. Size 729 distributions are a result of contributions from multiple sources, and thus being able to 730 discriminate the sources based on their characteristic size distributions will help identify relevant 731 sources.

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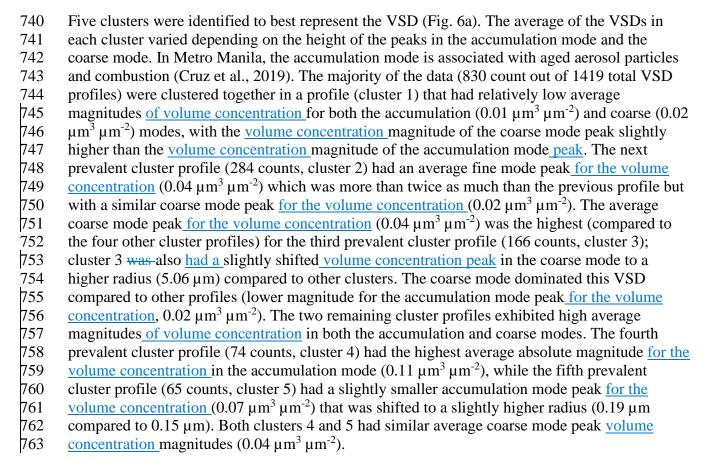
January 2009 and October 2018. Median VSDs over the study period based on (b) season (blue:

737

738 **3.3 Clusters**

739 3.3.1 VSD Cluster Profiles

⁷³⁶ DJF, red: MAM, orange: JJA, green: SON) and (c) time of day (blue: AM, red: PM).





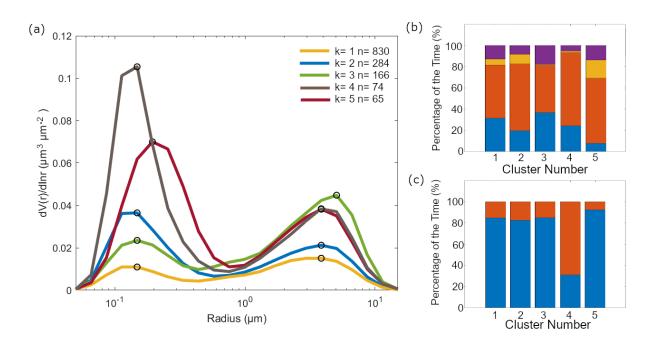
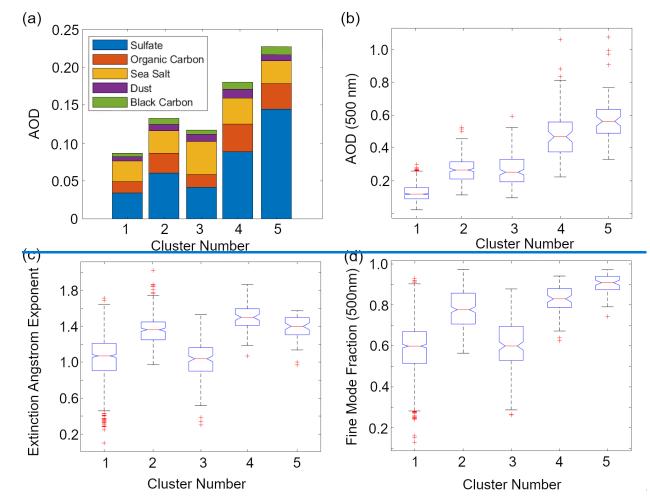


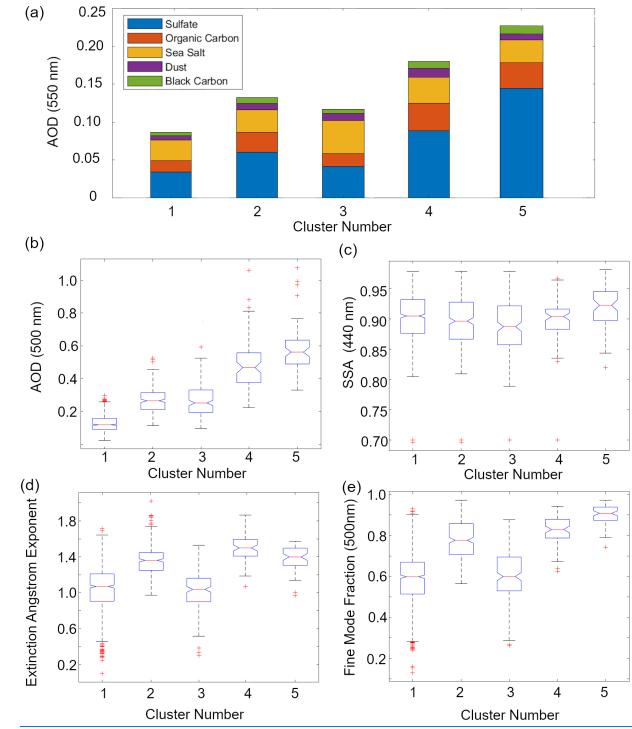


Figure 6: (a) Cluster analysis of VSD data yielding five characteristic and averaged VSDs with the number of points per cluster shown in the legend. The black circles on the curves show the

- peak locations in the submicrometer (<1 μ m) and coarse (≥1 μ m) 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.
- 778 Median total (AERONET) AOD values (Fig. 7b) were lowest (0.12) for cluster 1, though it had
- the second highest sea salt fractional contributions (31%) (Fig. 7a) to total AOD (MERRA-2)
- 780 (31%)-among all the clusters. Cluster 2 had relatively mid-range median total AOD values (0.27) that along with glusters 4 and 5 were demined a hyperfield state of a second state (4.0%) = 1.20%)
- 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
- 783 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
- 785 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
- 787 properties can help associate the clusters to air masses.

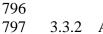
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790

Figure 7: (a) Average compositional contributions to aerosol optical depth (AOD at 550 nm)
from MERRA-2 per identified cluster. (counts per cluster from 1 to 5: 830, 284, 166, 74, 65).
Boxplots of AERONET (b) total AOD (500 nm), (c) single scattering albedo (SSA at 440 nm).
(d) extinction angstrom exponent (EAE, at 440 nm - 870 nm total), and (de) fine mode fraction
(FMF, at 500 nm) per cluster.



3.3.2 Air Mass Types

798 Air masses have been classified in previous studies based on their AOD, EAE, FMF, and

- 799 **FMFSSA** values (e.g., Lee et al., 2010 and Aldhaif et al., 2021). The criteria from different
- 800 studies (Table 2) were applied per cluster. Median total AOD of cluster 1 (0.12) was less than 0.2
- 801 (Fig. 7b), which is the threshold for sea salt sources (Kaskaoutis et al., 2007; Kaskaoutis et al., 802
- 2009). Half of the data points in cluster 1 also fall below the threshold for clean environments 803 (AOD < 0.1) (Sorooshian et al., 2013). Based on its median FMF (0.60) and EAE (1.07, where
- 804 EAE < 1 is coarse and EAE >1 is fine) and FMF (0.60) values (Fig. $\frac{7e7}{d}$ and $\frac{7d7}{d}$), cluster 1 is
- 805 a mixture of fine and coarse particles. The fine Cluster 1 is the only cluster with a median that
- 806 meets that threshold value for clean marine sources (AOD < 0.2), and we know from Sect. 3.3.1
- 807 that its average VSD magnitude was greater for the coarse fraction and that its sea salt
- 808 contribution to total AOD was second greatest among the clusters. Thus, we can say that most
- 809 probably, cluster 1 is a background clean marine source, since it also is predominant throughout
- 810 the seasons (Fig. 6b). This makes sense given the proximity of the ocean to Metro Manila from
- 811 both the east and the west. The median SSA (0.90 at 440 nm) for cluster 1 (Fig. 7c), however,
- 812 suggests the presence of absorbing particles most probably due to high black carbon in the local
- 813 source (Cruz et al., 2019) that is mixed in with this generally clean marine source.
- 814 Most of the data from the other clusters all fall in the polluted category (Table 2), based on their
- 815 median total AODs (>0.1) (Fig. 7b). Cluster 2 has a median FMF value of 0.78 (Fig. 7e7e),
- 816 which suggests that most of the particles in this air mass are in the fine fraction. They are,
- 817 however, not sufficiently dominant in the aerosol for them to be typical of urban/industrial
- 818 sources. The average VSDs (Fig. 6a) of cluster 2 similarly suggest that their relative
- 819 accumulation mode magnitude is higher than the coarse magnitude, but not much higher. Like
- 820 cluster 1, cluster 2 is also more evenly distributed across the seasons (Fig. 6b)). The median SSA 821
- for cluster 2 (0.90 at 440 nm) is also similar to the SSA of cluster 1 (Fig. 7c) where the local and
- 822 background particles are mixed. Cluster 2 could be a fine polluted background source 823
- superimposed on the dominant marine source. Metro Manila is a megacity with continuous and 824
- large amounts of sources that could be, due to its proximity to the ocean, interacting with the
- 825 background.
- 826 Based on its median EAE value (1.04) (Fig. 7d), cluster 3 is mixed but mostly in the coarse
- 827 fraction, consistent with its VSD profile (Fig. 6a) which has the highest coarse magnitude (FMF
- 828 = 0.60) compared to the other clusters. The contribution of data from September to February is
- 829 greatest in cluster 3, consistent with expected coarser particles during this period when the winds
- 830 are initially shifting from the southwest before becoming more northeasterly, as previously
- 831 noted. This air mass can be a mixed polluted air mass, which is possibly transported due to the
- 832 large sea salt contribution to total AOD (Sect. 3.3.1). Median SSA (0.89 at 440 nm) was lowest
- 833 for cluster 3 (Fig. 7c), this and the relatively high coarse particle contribution suggests cluster 3
- 834 as a possible dust source based on past studies (Lee et al., 2010). This air mass can be a mixture
- 835 of local sources and transported dust air masses, the large sea salt contribution (~37%) to total
- 836 AOD (Sect. 3.3.1) can be related to long-range transport.
- 837 Both clusters 4 and 5 have median total FMF (0.83 and 0.91) (Fig. 7e7e) values exceeding the
- 838 mark (> 0.8, Table 2) for urban/industrial air masses. Combining this and results from the
- 839 previous sections confirms that cluster 4 can be an urban/industrial source given that it had the
- 840 highest median accumulated mode peak and organic carbon contribution ($\sim 20\%$) to total AOD
- 841 among the clusters. The median SSA for cluster 4 (0.90 at 440 nm) was similar to the median

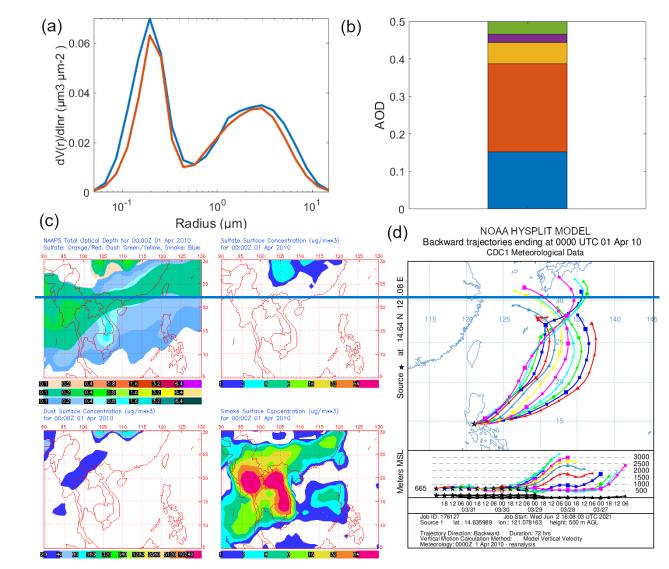
- 842 <u>SSA of clusters 1 and 2 (Fig. 7c), but the maximum SSA value for this cluster was lowest in</u>
- general among all the clusters suggesting cluster 4 has the net most absorptive effect. The cluster
- 4 air mass is probably from local sources and transported biomass burning emissions. The high
- 845 median EAE (1.40, Fig. 7d) may be associated with aerosol particles due to biomass burning
- 846 (<u>Deep et al., 2021).</u>

Cluster 5 had the highest median total AOD (0.56) and FMF (0.91) values (Fig. 7e7b and 7e). It

- also had the highest (Fig. 7a)-sulfate contribution (~64%) to total AOD as well as (Fig. 7a), the
- highest median SSA (0.92 at 440 nm, thus most reflective particles among the clusters) (Fig. 7c),
- 850 and a shifted accumulation mode peak (Fig. 6a). These characteristics suggest that cluster 5 is a
- 851 possible cloud processing air mass (Eck et al., 2012). The larger peak in the accumulation mode 852 is possibly the cloud signature. Previous studies have attributed this larger mode to cloud
- as possibly the cloud signature. Previous studies have attributed this larger mode to cloud
 processing due to the conversion of SO₂ to sulfate (Hoppel et al., 1994). Cloud processing is a
- major source of sulfate (Barth et al., 2000).
- 855 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<u>dust</u> (12%), urban/industrial (5%), and cloud
- processing (5%) air masses contribute 22% all together<u>altogether</u>. We can investigate more
- deeply and look at specific case studies that can better describe the air masses identified here.
- 860

861 **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
 bis bas VSD construction magnitudes
- 865 higher VSD <u>concentration</u> magnitudes.
- 866
- 867 3.4.1 Long Range Transport of Smoke
- 868 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
- 870 contributor to AOD (Fig. 8b and 9b) for both long-range transport cases. The first of two events
- 871 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
- 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 in the area (Fig. 80). Though the absolute black carbon 875 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
- 878 (Reid et al., 2005).



Apr 1 02:30:46 2010 NRL/Monterey Aerosol Modeling

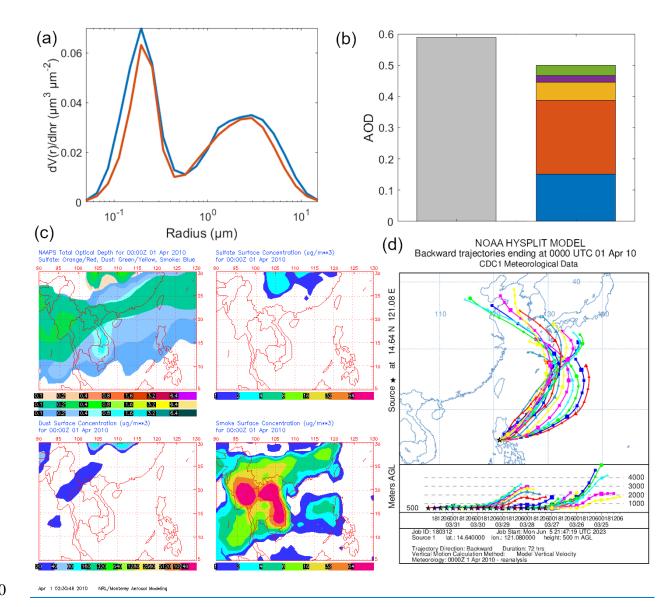




Figure 8: Case study of long-range transport (smoke – East Asia) around 1 April 2010. (a)
AERONET VSDs at (blue) 00:01 and (red) 00:26 UTC, (b) <u>AOD from AERONET (gray:</u>
median AOD at 500 nm) and MERRA-2 hourly (green: black carbon, violet: dust, yellow: sea
salt, orange: organic carbon, blue: sulfate) compositional contributions to AOD (550 nm) 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 threeseven-day back-trajectories arriving at Manila Observatory
 at 00:00 UTC.
- 889 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
- 891 was greater (reaching 0.8) especially in Peninsular Southeast Asia. Smoke surface concentrations
- 892 were also widespread (Fig. 8c) with greatest concentrations in East Asia that reached the
- 893 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

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

900 direction of the Malaysia and Indonesia. NAAPS maps likewise show elevated AOD,

901 specifically smoke contribution to AOD (Fig. 9c), as well as enhanced smoke surface

902 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

904 Philippines in the same direction of the back-trajectories. There were fires in the lowland (peat)

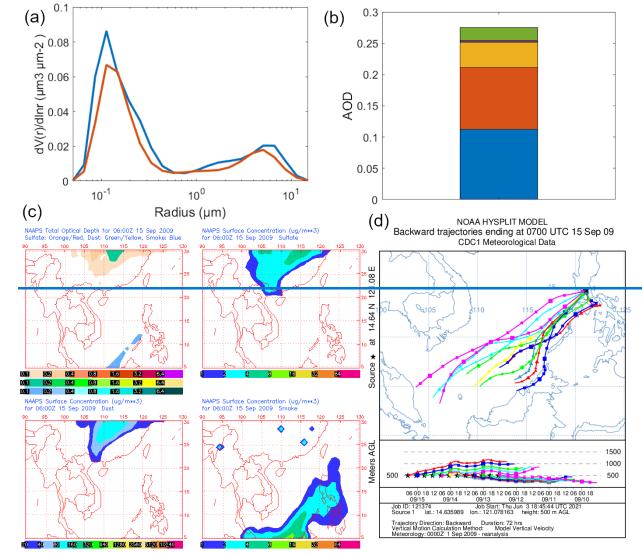
forests of Borneo around this time (NASA, 2009). MERRA-2 AOD contributions for this case were greatest due to organic carbon as well as sulfate (Fig. 9b), and the absolute black carbon

906 were greatest due to organic carbon as well as sulfate (Fig. 9b), and the absolute black carbon 907 contributions were greatest compared to other cases. The VSD of this smoke case from Southeast

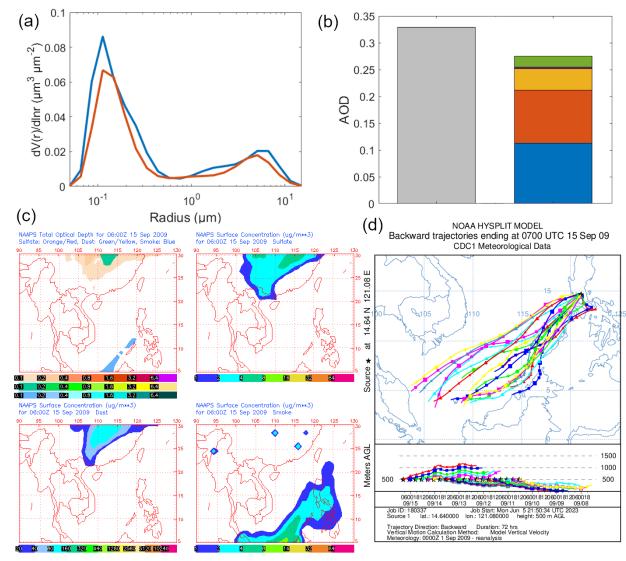
907 Controlutions were greatest compared to other cases. The VSD of this shoke case from Southeast 908 Asia (Fig. 9a) resembled that from long-range transported smoke from East Asia (Fig. 8a) and

909 the urban/industrial air mass (cluster 4, Fig. 6a). This case occurred in the afternoon, which was

910 the prevalent time that the urban/industrial air mass was observed (Fig. 6c).



Sep 15 08:31:38 2009 NRL/Monterey Aerosol Modeling



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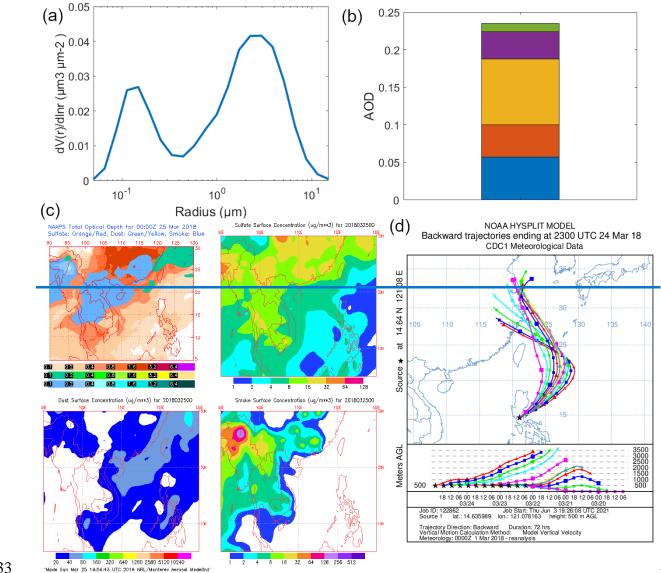
2. Sep 15 08:31:38 2009 NRL/Monterey Aerosol Modeling

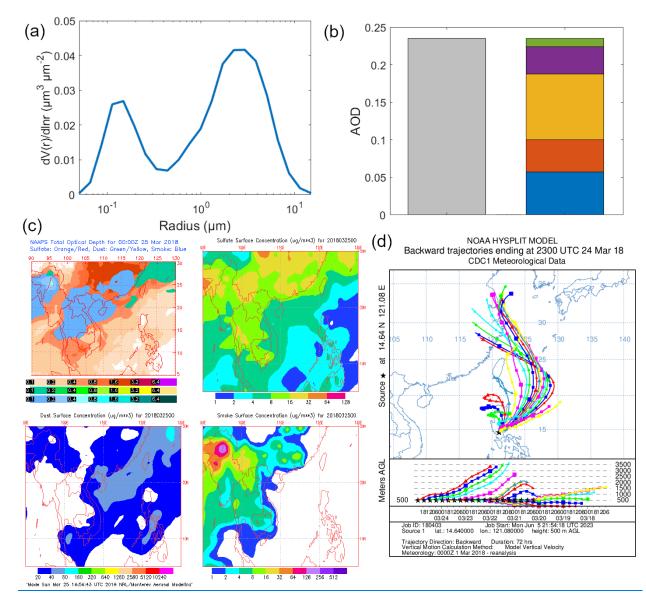
- 913 **Figure 9:** Case study of long-range transport (smoke Southeast Asia) around 15 September
- 2009. (a) AERONET VSDs at (blue) 07:27 and (red) 07:52 UTC, (b) <u>AOD from AERONET</u>
- 915 (gray: median AOD at 500 nm) and MERRA-2 hourly (green: black carbon, violet: dust, yellow:
- 916 sea salt, orange: organic carbon, blue: sulfate) compositional contributions to AOD (550 nm)
- 917 closest in time to 07:27 UTC, (c) NAAPS maps of total and compositional hourly AOD
- 918 (orange/red: sulfate, green/yellow: dust, blue: smoke) and sulfate, dust, and smoke surface
- 919 concentrations at 06:00 UTC, and (d) HYSPLIT threeseven-day back-trajectories arriving at
- 920 Manila Observatory at 07:00 UTC.
- 921
- 922 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
- 924 polluted<u>dust</u> cluster (cluster 3), which had a mixed size distribution but a more dominant coarse
- 925 contribution. This is consistent with the most dominant contribution to AOD in the area, which
- 926 was sea salt and dust (Fig. 10b). The back-trajectories were from East Asia around the same
- 927 latitude as Taiwan (Fig. 10d). That area, at that time, had increased AOD in general from sulfate

928 and dust (Fig. 10c). The AOD from both AERONET and MERRA-2 (Fig. 10b) are lower than

929 0.3 (the AOD threshold for dust in other studies, Table 2) because of the long distance from the
 930 source (thousands of kilometers). The dust and sulfate seemed to have been transported to Metro
 931 Manila from East Asia based on the NAAPS sulfate and dust surface concentrations (Fig. 10c).

932





934

Figure 10: Case study of long-range transport (dust) around 24-25 March <u>20092018</u>. (a)

AERONET VSD at (blue) 23:23 UTC, (b) AOD from AERONET (gray: AOD at 500 nm) and

937 MERRA-2 hourly (green: black carbon, violet: dust, yellow: sea salt, orange: organic carbon,

blue: sulfate) compositional contributions to AOD (550 nm) closest in time to 23:23 UTC, (c)

- 939 NAAPS maps of total and compositional hourly AOD (orange/red: sulfate, green/yellow: dust,
- 940 blue: smoke) and sulfate, dust, and smoke surface concentrations at 00:00 UTC on March 25,
- and (d) HYSPLIT threeseven-day back-trajectories arriving at Manila Observatory at 23:00
- 942 UTC.
- 943

944 <u>3.53.4.3</u> Cloud Processing

Sulfate dominated the AOD (Fig. 11b) for this case on 26 August 2009 in the area around Metro

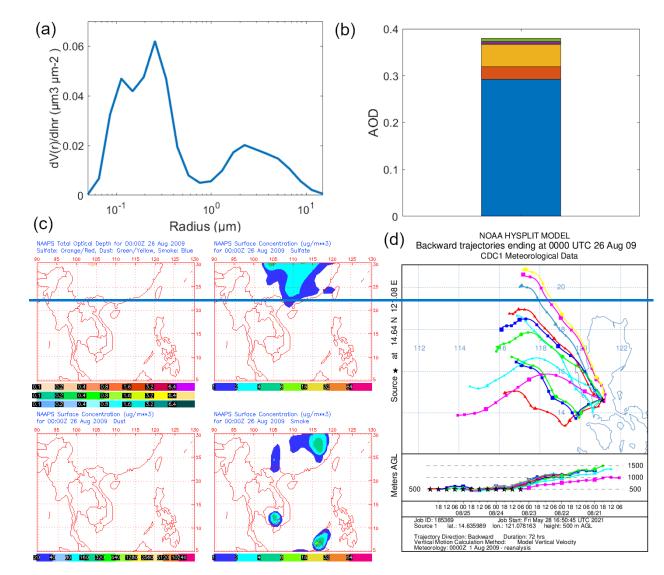
946 Manila. This along with its VSD exhibiting a second peak (Fig. 11a) in the accumulation mode

947 make it very similar to the cloud processing cluster (cluster 5). Sulfate has been known to be

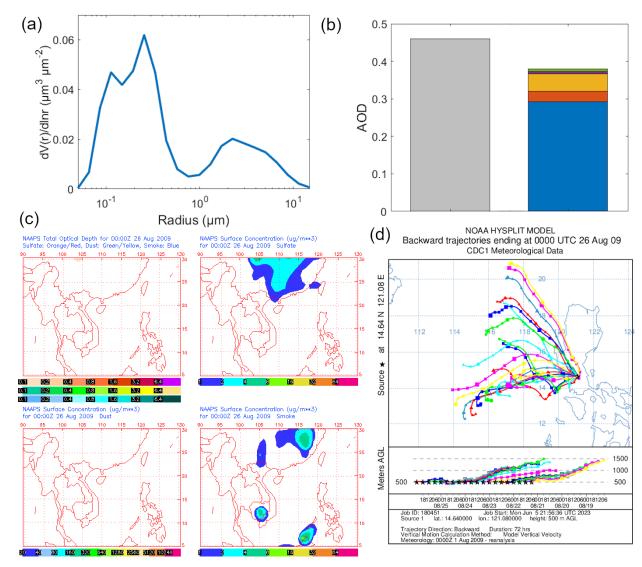
- 948 enhanced through chemical productions in clouds and is used as a signature for cloud processing
- 949 (Barth et al., 2000; Ervens et al., 2018). Aqueous production of sulfate is significant in areas with

- 950 sources and clouds (Barth et al., 2000), and this case study has both. Aside from the high sulfate
- 951 contribution to AOD, the cloud fraction (Aqua/MODIS, Terra/MODIS, Fig. S3) is very high
- 952 (~100%) in the area of the back-trajectories (Aqua/MODIS, Terra/MODIS, Fig. S211d).
- 953 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
- 955 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
- 959 (Jamora et al., 2020).

960



Aug 25 08:38:20 2009 NRL/Monterey Aerosol Modeling



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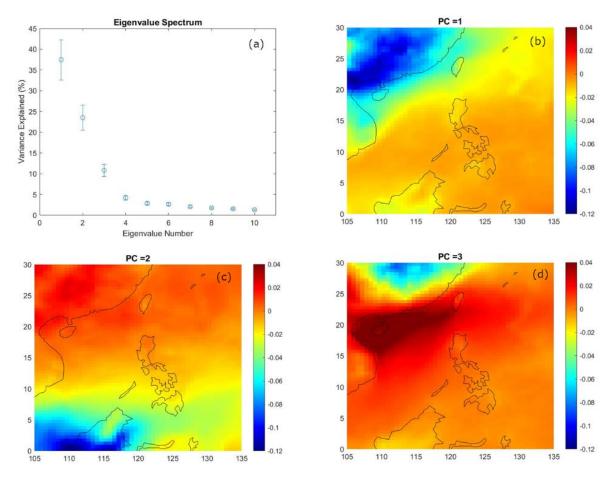
Figure 11: Case study of cloud processing on 26 August 2009. (a) AERONET VSDs at 00:18
UTC, (b) <u>AOD from AERONET (gray: median AOD at 500 nm) and MERRA-2</u> hourly (green:
black carbon, violet: dust, yellow: sea salt, orange: organic carbon, blue: sulfate) compositional
contributions to AOD (550 nm) closest in time to 00:18 UTC, (c) NAAPS maps of total and
compositional hourly AOD and contributions and smoke surface concentrations at 00:00 UTC,
and (d) HYSPLIT threeseven-day back-trajectories arriving at Manila Observatory at 00:00
UTC.

970

971 3.63.5 EOF Analysis of AOD in Southeast Asia

- 972 To contextualize the analysis of aerosol particle
- 973 The air masses in Metro Manila, major are influenced by regional sources of aerosol particles in
- 974 Southeast Asiawhich were identified based on the dominant principal components fromthrough
- 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

977 probably the major distinct aerosol particle sources in the region. They will be the focus of the 978 subsequent discussion.



979

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.

983 The first PC explains 37.46% of the data variance (Fig. 12a) and, based on the map of the 984 regression coefficients (Fig. 12b), separates mainland East Asia from the Philippines and 985 Indonesia. East Asia is a globally recognized source for high AOD (Li et al., 2013), and its 986 contribution to particles in Southeast Asia possibly corresponds to the first PC. The second PC 987 explains 25.51% of the data variance (Fig. 12a) and separates the southern Southeast Asia from 988 northern Southeast Asia at around 15°N (Fig. 12c). Southern Southeast Asia is a known regional 989 source of aerosol particles due to biomass burning (Cohen et al., 2017) and could be associated 990 with the second PC. The third PC explains 10.80% of the data variance (Fig. 12a) and separates 991 northern East Asia from southern East Asia mainland and the rest of Southeast Asia (Fig. 12d).

992 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.

995 The correlation maps of the first PC and the sulfate contribution to AOD (Fig. 13a and 13d) 996 show high and statistically significant correlations (gray areas) in mainland East Asia and 997 Taiwan, parts of western Philippines and Borneo, which are the probable sulfate sources. Clues 998 from the mean monthly wind vector maps in April (Fig. 14a and 14d) and mean monthly AOD in 999 either March or April (Fig. S3c or S3e) most resembling the features of regression map of the 1000 first PC (Fig. 12b) and the PC time series peaking in March (Fig. S4) together suggest that the 1001 first PC may be associated with air masses that are present around March or April. Emissions 1002 sources and meteorology that are dominant during the peak dates in the PC time series offer 1003 clues to the attribution of each PC. The Southeast Asia region and the Philippines is influenced 1004 by the monsoon systems (Coronas, 1920; Matsumoto et al., 2020) and February to March is the 1005 time when the winds are transitioning from the northeasterly to easterly. The first PC could be 1006 affected by the easterly winds, which are dominant around March when its PC values peaked. 1007 The higher-level winds (free troposphere) (Fig. 14a) in April are from the west in mainland East 1008 Asia and are from the east in the Philippines and it is possible that the different wind regimes are 1009 distinguishing the sulfate sources in East Asia and the Philippines and beyond. Sulfate is a 1010 known product of industry in East Asia (Smith et al., 2011; Li et al., 2017) while the West Luzon

1011 and West Visayas islands have large power plants (Jamora et al., 2020).

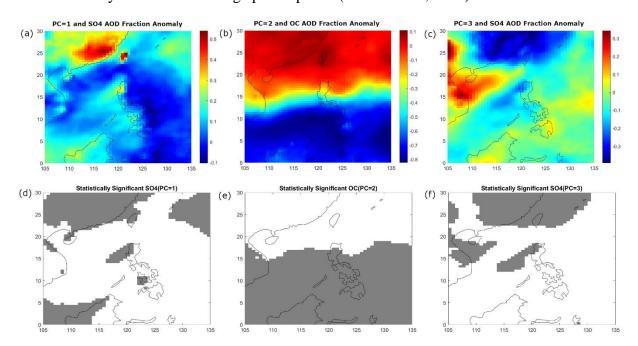




Figure 13: Correlation coefficients of principal components with (a/c) sulfate AOD fraction and (b) organic carbon AOD fraction. Statistically significant (90%, d-f) areas are shaded gray.

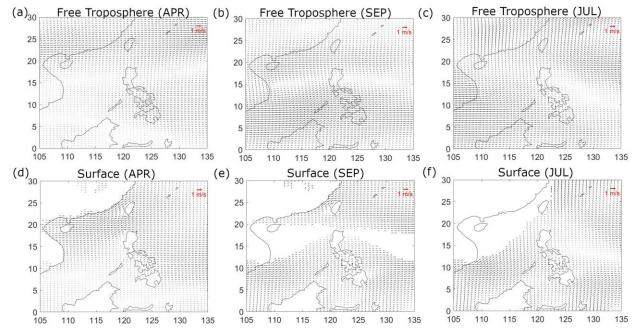


Figure 14: Monthly averaged winds for (a & d) April, (b & e) September, and (c & f) July from MERRA-2 at (725 hPa, a-c) the free troposphere approximate and at (1000 hPa, d-f) the surface.

1015

1018 The correlation maps of the second PC and the OC contribution to AOD (Fig. 13b and 13e) show 1019 high and statistically significant correlations from $0^{\circ}N$ to $15^{\circ}N$. The large magnitude of the 1020 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 1021 1022 burning during its fire season (Glover and Jessup, 1998), and thus the local significance 1023 established in the southern Southeast Asia is most likely due to the Indonesia biomass burning 1024 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 1025 1026 PC in the time series (Fig. S4). Winds are usually from the southwest and west due to the 1027 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 1028 1029 troposphere mean monthly winds (Fig. 14b and 14e) are from the southwest (in the general 1030 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 1031 1032 fires. Of interest is the line of separation of the northern and southern Southeast Asia in the 1033 principal component that is within the area of the monsoon trough (Wang et al., 2007). This line 1034 is also evident in the surface and the free troposphere maps where the southwest winds from the 1035 area of Indonesia meet the easterlies in north Southeast Asia (Fig. 14b and 14e) and which thus 1036 appears to be limiting the dispersion of the biomass burning emissions to southern Southeast 1037 Asia.

1038 The third PC was also well correlated to the sulfate AOD fraction though, compared to the first 1039 PC correlation maps, there were distinctions between the northern and southern East Asia 1040 regions (Fig. 13c and 13f). The local Philippine source still came out in the correlation maps as a 1041 significant source. It was not clear from the PC time series (Fig. S4), which showed peaks in the 1042 third PC in February, how the dates were related to the PC profile. The free troposphere winds in 1043 July (Fig. 14c), as well as the AOD monthly mean map in July (Fig. 14c), however, showed 1044 more similarities to the third PC regression map. Both showed a delineation between the 1045 northern East Asia and southern East Asia (including Hong Kong) features. Mean winds (Fig. 1046 14c) in the free troposphere are from the west, due to the southwest monsoon, in the area around 1047 the Philippines, and they were from the northeast in north Southeast Asia. The interface of the 1048 winds is within the approximate location of the monsoon trough in July (Wang et al., 2007), and 1049 it is thus possible that the monsoon trough is causing the separation of the sulfate sources. This 1050 could be investigated further. The monsoon trough has been noted to scavenge aerosol particles 1051 from southern Southeast Asia (Reid et al., 2013). It is evident from the analysis that meteorology 1052 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., 1053 2019; Braun et al., 2020; Hilario et al., 2020b; Hilario et al., 2022).

1054

1055

1056 4. Conclusion

1057 Metro Manila has both urban and industrial local sources known to contribute to the dominance

1058 of fine mode particles in its air (Cruz et al., 2019). Ten years of AERONET data in Manila

1059 Observatory suggest that aerosol particles in Metro Manila were mixed in size but with a

1060 prevalent fine mode fraction (>50% FMF) throughout the year. Background clean marine aerosol 1061 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 1062

1063 cluster analysis of volume size distributions. The proximity of Metro Manila to the sea, both in

1064 the east and west, along with local sources, transportation being the most prominent, together

contribute to the prevalence of the marine and fine particles. The prevalence of marine particles 1065 1066 could explain the relatively small AOD values in Metro Manila compared to other Southeast

1067 Asian megacities (Reid et al., 2013).

1068 Regional sources and meteorology also impact monthly aerosol optical depth trends in Metro 1069 Manila from EOF analysis. Biomass burning from Borneo and Sumatra emerged in the study as

1070 the second most prevalent regional anthropogenic aerosol particle source in Southeast Asia. 1071 Though the monsoon trough limits the dispersion of aerosol particles throughout the entire

1072 Southeast Asia, biomass burning emissions impact southern Southeast Asia including Metro

1073 Manila during the southwest monsoon (July to September). The monsoon winds facilitate the

1074 transport of fine particles during the peak burning season in Borneo and Sumatra (August-

1075 September). This is experienced in Metro Manila as higher than usual aerosol particle loadings

1076 duringaround the same period- (August to October). Climatologically, August iswas also when

1077 aerosol optical depth peaked over Metro Manila, concurrent there were particles with the greatest

1078 fine mode fractions that were relatively absorbing and non-hygroscopic possibly due to increased

1079 organic and elemental carbon fractional contributions. Though not as strong a source as the 1080 Borneo and Sumatra case, the peninsular Southeast Asia burning season (March-April) also

1081 contributed to extreme aerosol particle concentrations over Metro Manila.

1082 High aerosol particle loadings due to transported dust, probably from East Asia, were observed

1083 in Metro Manila during the transition period between the southwest and northeast monsoons and

1084 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 1085

1086 northeast monsoon when annual wind speeds were highest. Particles then were observed to be

- 1087 largest in diameter, with the greatest coarse fraction contribution, relatively high absorptivity,
- and most hygrosocopicity, compared to other months of the year. This is probably due to
- 1089 constituents other than soot, especially aged dust (Kim and Park, 2012; Geng et al., 2014) and
- 1090 sea salt which the northeast winds appear to be bringing in from the general direction of the
- 1091 Luzon Island and the Philippine Sea (West Pacific Ocean).

1092 Cloud processing is one of the cases that were linked to very high aerosol particle loading in

- 1093 Metro Manila. This is associated with sulfate sources, which appear more localized in nature
- 1094 because of a power plant nearby. This sulfate source seems to be distinct from the industrial
- 1095 sulfate air mass from East Asia, which is the most dominant regional aerosol particle source in
- 1096 Southeast Asia (Li et al., 2013). Winds appear to limit the mixing of this notable East Asia air
- 1097 mass with local industrial sources in the region including the Philippines and Indonesia.
- 1098 The formation of cloud systems in Southeast Asia is complex due to intersecting large- and
- 1099 small-scale mechanisms. Additionally, the interaction of particles and clouds in Southeast Asia is
- 1100 not yet well understood. In Metro Manila, both topography and meteorology affect aerosol
- 1101 particle distribution (Cruz et al., 2023). This baseline study on the aerosol particle characteristics
- 1102 in Metro Manila and in regional Southeast Asia shows how meteorology impacts varied aerosol
- 1103 particle sources (e.g., sulfate, elemental carbon, and organic carbon) and their distribution in the 1104 region. This can help in mitigating aerosol particle sources in the region and in the deepening of
- region. This can help in mitigating aerosol particle sources in the region and in the deep the understanding of the relationship of aerosol particles, meteorology, and clouds.
- 1105

1107 Data availability

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- 1137

1138 Author contributions

- 1139 GRL and AS designed the experiment. NL, SNU, GRL, GFG, HJO, JBS, and MTC, carried out
- 1/140 various aspects of the data collection. GRL, AS, JBS, MOC, MRH, <u>CC</u>, and <u>CCLDG</u> 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
- 1143 led the management and funding acquisition. All authors approved the final version of the 1144 manuscript.
- 1144 m 1145

1146 **Competing interests**

- 1147 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.
- 1150

1151 Acknowledgements

- 1152 The authors acknowledge support from NASA grant 80NSSC18K0148 in support of the NASA
- 1153 CAMP²Ex project, in addition to ONR grant N00014-21-1-2115. We acknowledge the US Naval
- 1154 Research Laboratory for providing the AERONET instrument. We acknowledge the use of
- 1155 imagery from the NASA Worldview application (https://worldview.earthdata.nasa.gov), part of
- 1156 the NASA Earth Observing System Data and Information System (EOSDIS).
- 1157

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