Inferring Polluted Asian Absorbing Aerosol Properties Using Decadal Scale AERONET Measurements and a MIE Model

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Abstract Absorbing aerosols uniquely impact radiation, aerosol transport, and meteorology. This paper quantifies black carbon core and sulphate shell size and mass using decadal measurements of multi-spectral aerosol optical depth, single scattering albedo, and angstrom exponent from Aerosol Robotic Network stations located throughout East, Southeast, and South Asia, in connection with a MIE model. All sites are uniquely characterized into four types: urban, biomass burning, long-range transport, and clean. Unique size and mass probability distributions of both the core and shell are calculated within each classification. Well known urban, biomass burning, and clean sites are all properly identified. Furthermore, two unique sites previously thought to not have multiple characteristics are identified, with urban and biomass burning significant in Beijing and long-range transport significant in the otherwise clean South China Sea at Taiping Island. It is hoped that these results will allow for advances in attribution and radiative forcing studies.

Plain Language Summary Black Carbon strongly absorbs visible radiation, leading to unique impacts on atmospheric radiation, climate, the water cycle, and PM2.5. This work attributes different aerosol source characteristics, and further specifies the size distribution and concentration of aerosol black carbon cores and refractive shells. This work uses measurements of aerosol optical depth, single scatter albedo, and angstrom exponent, across multiple different wavelengths of light, in combination with statistics and a MIE model (physical model of aerosol/radiation interaction) using a Core-Shell approximation. The results show that aerosols observed in East, Southeast, and South Asia can be uniquely classified into four types: urban, biomass burning, long-range transport, and clean. These results are consistent in terms of aerosol size and mass at each site within each type of characterization. Furthermore, two unique sites are identified in which a second characteristic occurs some significant fraction of every year, which otherwise was not known or previously identified in the literature. These results are expected to help enhance the understanding of attribution of aerosols, as well as provide specific size and mass details of the aerosols useful to improve radiative forcing models and aerosol impacts on climate change.

1. Introduction

Aerosol has been identified as one of the largest contributors to the uncertainty in our understanding of a diverse set of environmental issues including: the earth’s radiation balance and climate change (Chung & Seinfeld, 2005; Jacobson, 2001; Ramanathan & Carmichael, 2008), surface loadings of air pollution (Langmann et al., 2009; Rosenfeld, 1999), and acid rain (Charlson & Wigley, 1994; Wang, 2013; Zhang et al., 2004), among others. Furthermore, aerosols indirectly impact the lifetime, size, phase, and other properties of clouds, which in turn further impact the Earth’s radiative balance, precipitation, and consequently the lifetime of aerosols themselves (Rosenfeld et al., 2014; Tao et al., 2012). The major sources of absorbing aerosols come from anthropogenic emissions including: the combustion of fossil fuels (Crippa et al., 2018; Reuter et al., 2014; Tang et al., 2013), biomass burning and forest fires (Kim et al., 2008; Ming et al., 2010; Wang et al., 2020), industrial emissions (Wang, 2013); as well as from natural sources such as mineral dust (Kim et al., 2008; Wang et al., 2009) and volcanoes (Mills et al., 2016) etc., with significant changes in aerosols associated with changes in economics, population, energy consumption, human activities and climate change (Bauer et al., 2016; Cohen & Wang, 2014; Flato et al., 2013). Changes in aerosols occur in terms...
of the concentration, spatial-temporal variation, and chemical, physical, and optical properties, especially as different regions undergo different pathways of development (Cohen et al., 2017; Lin, Cohen, Wang, & Lan, 2020; Lin, Cohen, Wang, Lan, & Deng, 2020; Wang et al., 2020).

Properties of aerosol particles in-situ (including their concentration, size, composition, optical properties, and other microphysical characteristics) allow for a quantitative relationship to be made between aerosols, the environment and climate change (Cohen, 2014; Cohen & Wang, 2014; Lin et al., 2014). Some of the largest uncertainties connecting aerosols and radiation depend on the current lack of knowledge of the optical properties, size, and mixing state of aerosols in the atmosphere (Cai et al., 2020; Wang et al., 2020). The mixing state of aerosol particles impacts upon the amount of direct and scattered radiative streams, atmosphere-radiative equilibrium, condensation of secondary species, cloud condensation and ice nuclei, water uptake, particle acidity, and aerosol chemistry (Bondy et al., 2018; Chew et al., 2013; Schutgens & Stier, 2014). Therefore, it is important to identify the source (urban, biomass burning, long-range transport), size (nucleation, fine, coarse, etc.) and mixing state, including internal, external, and Core-Shell mixing, of existing aerosol particles in order to accurately assess their direct and indirect effects on the atmosphere (i.e., Cohen & Wang, 2014; Kim et al., 2008; Kodros et al., 2018; Wang et al., 2021).

Absorbing aerosols are dark particles (consisting of Black Carbon [BC], dust and certain other organics) that uniquely both scatter and absorb solar radiation in the atmosphere (Bond & Bergstrom, 2006; Kahn et al., 2010; Kim et al., 2008). These aerosols in turn have been measured to both significantly scatter and absorb radiation over specific parts of the globe, in turn leading to measurable cooling at the surface and heating in the middle of the atmosphere (Holben et al., 1998; Satheesh & Ramanathan, 2000; Seinfeld & Pandis, 1998). When absorbing aerosol has a high loading in the atmosphere, which is frequently observed due to its heterogeneity, they may exert a strong positive radiative forcing at the top of the atmosphere (Cohen et al., 2011; Grandey & Wang, 2019; Lau & Kim, 2006). This in turn provides a positive forcing leading to a warmer overall climate state, and further impacting the Earth's atmospheric circulation, surface vegetation and cloud and precipitation formation (Chung & Seinfeld, 2005; Guo et al., 2019; Jacobson, 2012; Ming et al., 2010; Wang, 2013). Therefore, work to further elucidate the size, mixing state, and absorbing characteristics of these aerosols is essential to further understanding.

Accurate simulation of the aerosol mixing state and in-situ composition is an open question currently being addressed by the scientific community (Andreae & Gelencsér, 2006; Kahnert et al., 2013). One such approach is to represent aerosol particles as a set of concentric spherical Core-Shell layers (where the core consists of BC, and the shell consists of sulphate and/or nitrate). This system is both physically reasonable, and can be made to represent scattering and absorbing properties ranging those measured in the real world. In specific, it has been determined that a Core-Shell model usually produces a better match with aerosol properties observed in heavily polluted region, such as found in East, Southeast, and South Asia, where BC once discharged into the atmosphere is coated with sulphate or nitrogen oxides in a very short time (Cohen & Wang, 2014; Peng et al., 2016; Zhang et al., 2016). Most current chemical transport models (CTMs), general circulation models and reanalysis products (such as WRF-Chem, GEOS-Chem, CESM-3, MERRA, NCEP, etc.) assume a non-Core-Shell approximation (Ichoku & Ellison, 2014; Kim et al., 2015; Nordmann et al., 2014; Yu et al., 2012), meaning that while CTMs can resolve the atmospheric composition of unmixed aerosols reasonably well, there are intrinsic problems in terms of being able to predict the composition of aerosols under polluted conditions or which have undergone long-range transport, and further tend to underestimate their overall absorption of radiation (Cohen et al., 2011; Kajino et al., 2012; Wang et al., 2021). The use of the Core-Shell model in this work is intended to aid elucidating how changes in measured atmospheric absorption and overall radiative properties of aerosols of different sizes and mixing ratios, can provide more insight into the sources and in-situ processing of observed BC.

The aerosol measurements are measured from the Aerosol Robotic Network (AERONET) network from 1997 through the present, using a Core-Shell approximation approach (Dubovik et al., 2002). A detailed analysis of the decadal AERONET optical measurements reveals important changes in the loadings, size and chemical composition of aerosols throughout most areas in East, South and Southeast Asia, due to rapid economic development, increasing urbanization, and increased population, among other factors. Furthermore, work to characterize the size of freshly emitted BC particles from biomass combustion is a complex function of the burning efficiency, fuel properties, moisture, and the surrounding atmospheric and
land-surface environment (Hyer et al., 2012; Lolli et al., 2018). Three different approaches all show that this size should fall somewhere in the range from 50 to 300 nm, as demonstrated in an idealized laboratory environment (Bond et al., 2004; Seinfeld et al., 2003), an inter-model comparison study (Lee et al., 2013; Regayre et al., 2018), and an in-situ measurement campaign (Kodros et al., 2018).

Our goal is to find an objective, clear, and simple way to systematically analyze the regional characteristics of aerosols, their sources, and in-situ environmental evolution. By connecting this approach with ground observations at known idealized urban, biomass burning, long-range transport, and other sites, a probabilistic set of source conditions can be quantified. This is then applied to other such sites around the world, allowing the contributions to the measured absorption to be better understood. This may lead to better controls on the loadings of particulate matter, especially when different techniques for source control or impact mitigation should be made at the same geographic location under different atmospheric, climatological, and other environmental conditions. The results can further aid in understanding the contributions to human health, air quality, and the climate system, since the new relationship helps provide information both to the local user, as well as other users upwind or downwind.

2. Methods and Data

Measurements from AERONET are used in conjunction with different thresholds to allow for classifications of the aerosol type to be made with respect to the Aerosol Optical Depth (AOD), Angstrom Exponent (AE), and Single Scatter Albedo (SSA). In addition, variance maximization and statistical fitting are also applied to further refine the source types following the approach of (Lin, Cohen, Wang, & Lan, 2020). A MIE model is then used to compute the SSA with respect to the size and mixing state of the aerosols simultaneously across multiple different wavelengths in the visible and near infrared (VIS/NIR). These results are then used with statistical-analytical techniques to constrain the size, and mixing state properties associated with the theoretical SSA and constrained by the measured SSA. This constraint is applied across all of the wavelengths simultaneously, allowing a solution space to be derived that probabilistically describes the set of mixing states and particle size distributions consistent with all of the observations.

2.1. AERONET Measurements

Daily measurements of AOD at 440, 675, 870, and 1020 nm were obtained from all AERONET stations in East Asia, South Asia, and Southeast Asia, using version 3, level 2 aerosol inversion products (Holben et al., 1998) from March 1997 to May 2017. As a measure of the amount of absorbing aerosol, Version 3 inversion products corresponding to SSA at 440, 675, 870, and 1020 nm were also obtained when AOD > 0.4 (Cohen & Wang, 2014; Dubovik et al., 2000). AE was computed based on AOD measurements at 440 and 675 nm, using only particles with an AE < 0.3 (due to possible cloud contamination).

2.2. Filtering and Grouping Highly Polluted Regions

To ensure all sites have a sufficient amount of AERONET inversion data so as to group the findings using variance maximization (following the method of Lin, Cohen, Wang, & Lan, 2020), each site must have either 100 or more than days of data with one of the three following conditions: high mean AOD (average AOD > 0.4), high extreme event AOD (one day of AOD > 1.0), highly variable AOD (standard deviation [STD] of AOD > 0.2), or more than 300 days of total measurements. This combination ensures that regions which have occasional extreme events such as biomass burning or long-range transport, are also considered alongside those regions which are more constantly polluted, such as urban areas, leaving 67 sites for further analysis.

Determination coefficients ($R^2$, $P > 95\%$) between all combinations of AOD, AE, and SSA (at 440 nm), absolute mean values of AOD (at 440 nm), the ratio of standard deviation of AOD to AOD (at 440 nm), and the 90th percentile and the 50th percentile of the ratio of standard deviation of AOD to AOD (at 440 nm) are used to categorize the 67 sites into four orthogonal groupings. The biomass burning group has increasingly smaller particle sizes as AOD increases (AE and AOD increase simultaneously), as well as a mean AOD greater than 0.5. Clean sites have a lower mean AOD (less than 0.5) and a lower absolute standard deviation.
ratio of AOD (less than 0.3). Urban sites have larger and less absorbing particles as AOD increases or larger particles as AOD increases, due to rapid deposition of secondary nitrate, sulphate, and low vapor pressure Volatile Organic Compounds (VOCs). This should be coupled with a not too large difference in extremes, here defined as the ratio between the 90th percentile and the 50th percentile of the Probability Density Function (PDF) of the standard deviation ratio (ratio less than 2.65). Finally, long-range transport has more absorbing aerosols as AOD increases or larger particle size as AOD increases, due to high amounts of small absorbing aerosols emitted from the fires, which slowly age and oxidize as they are transported far downwind; combined with a very high standard deviation (the ratio of the 90th to 50th percentile of the standard deviation of AOD to AOD ratio is more than 2.65), as consistent with the growth of aerosol particles in-situ for an extended period (Cohen & Wang, 2014; Cohen et al., 2011), as well as statistics of measurements downwind from fires as observed by Cohen et al. (2017), Deng et al. (2020), Lin, Cohen, Wang, Lan, and Deng (2020), Wang et al. (2020).

2.3. Radiative Properties

A MIE model is applied to compute the optical properties (including absorption and scattering) of hundreds of different combinations of sizes of individual Core-Shell mixed aerosols, using a BC core and a sulphate or nitrate shell of varying thicknesses following (Haywood & Shine, 1995). The vertical integral of these optical properties through the atmospheric column corresponds to the radiative properties. In this work, the thickness of the BC core \( R_{\text{core}} \) and shell \( R_{\text{shell}} \) are varied in steps of 10 nm each from 0.05 to 0.50 \( \mu m \) and 0.01 to 0.80 \( \mu m \) respectively, using best estimates of the respective real part \( a \) (2.0 for BC and 1.426 for sulphate) and imaginary part \( b^* \) (1.0 for BC and 0.0 for sulphate) (Kim et al., 2008; Schuster et al., 2005). This range of sizes allows for the full range of the fine mode fraction to be explored (Kahn et al., 2016; Van Donkelaar et al., 2016; Weagle et al., 2018). The resulting computed values of extinction efficiency \( Q_{\text{ext}} \) and absorption efficiency \( Q_{\text{abs}} \) are used to compute the SSA (Zhang et al., 2008).

2.4. Model and Measurement Inter-Comparison

A new metric is computed to constrain the modeled SSA simultaneous measurements across all four of the wavelength-dependent measurements from AERONET. First, the central 80% of the PDF of measured SSA is computed independently at each wavelength \( ii \), hereafter referred to as \( \int_{0.1}^{0.9} \text{SSA}_{440} \). Second, we take the intersection of the solution space given by \( \int_{0.0}^{0.8} \text{SSA}_{440} \) for each of the four wavelengths to constrain the values computed by the MIE model, herein called the Merged SSA (MSSA), as detailed in Equation 1.

\[
\text{MSSA} = \left( \int_{0.0}^{0.8} \text{SSA}_{440} \right) \cap \left( \int_{0.0}^{0.8} \text{SSA}_{675} \right) \cap \left( \int_{0.0}^{0.8} \text{SSA}_{870} \right) \cap \left( \int_{0.0}^{0.8} \text{SSA}_{1020} \right)
\]  
(1)

The MSSA is subsequently used to constrain the modeled SSA as a function of core and shell size. The resulting set of solutions \( [ R_{\text{core}}, R_{\text{shell}} ] \) (radius of BC core and refractive shell respectively in nm) form a solution space spanning at most a few hundred combinations. This space is hereby analyzed with two additional indices: The Aerosol Size Ratio (ASR) and the Aerosol Mass Ratio (AMR), which are respectively computed by Equations 2a and 2b (in which \( R_{\text{total}} \) is the total particle radius, \( \rho_{\text{shell}} \) and \( \rho_{\text{core}} \) are the density of the sulphate/nitrate shell and BC core, while \( M_{\text{shell}} \) and \( M_{\text{core}} \) are the mass of the sulphate/nitrate shell and BC core respectively), and represent the individual size and mass ratios over the MSSA solution space.

\[
\text{ASR} = \frac{R_{\text{shell}}}{R_{\text{core}}} = \frac{R_{\text{total}} - R_{\text{core}}}{R_{\text{core}}}
\]  
(2a)

\[
\text{AMR} = \frac{M_{\text{shell}}}{M_{\text{core}}} = \frac{\rho_{\text{shell}} * \left( R_{\text{total}}^3 - R_{\text{core}}^3 \right)}{\rho_{\text{core}} * R_{\text{core}}^3}
\]  
(2b)

Some sites classified as urban have a significant minority of days that also contain characteristics of biomass burning or long-range transport. To address this complexity, we separate extremely polluted days point-by-point and re-analyze the separated data set. The extremes are separated by first computing the mean and the standard deviation of the AOD at 440 nm and second separating all data greater than the mean plus one standard deviation. This procedure is repeated three times, merging each iteration of extremes with the previous extremes. This method has been shown to work well for extremes of Ozone Monitoring Instrument NO2 and Multi-angle Imaging SpectroRadiometer AOD measurements (Deng et al., 2020).
3. Results

The new method results in all stations being uniquely sorted into four groups: 13 biomass burning, 25 clean, 14 urban, and 15 long-range transport. In addition, we find two sites obviously are a mixture of more than one characteristic group occurring on a minority of days, and the other group occurring overall.

3.1. Grouping and Classification

The sites belonging to the biomass burning classification are given in Figure 1a, and are shown to have an AOD time series with most of the year being relatively clean and a short but intense period which is extremely polluted, with the difference ranging from a factor of 2.3–5.7. At each individual site these peaks occur annually with a similar magnitude, start time and end time every year. However, these magnitudes, start times and end times vary greatly between different sites. In addition, at all biomass burning sites, the correlation coefficient between the AOD and AE is always positive, with a maximum correlation coefficient of 0.43, consistent with a smaller particle size at higher pollution loadings which then undergo in-situ condensational growth.

Chiang Mai has the most overall data among these biomass burning sites (Figure 1b), and every year the extreme events occur exclusively from February to April. Furthermore, the mean AOD value is 0.73, and correlation coefficient between the AOD and AE is 0.18. It is also well known as a typical biomass burning site (Cohen et al., 2017; Lin, Cohen, Wang, & Lan, 2020). For these reasons, Chiang Mai is hereafter used as the representative biomass burning site.

Unlike biomass burning regions, the clean regions do not have any obvious or recurring annual peak, have both an overall low absolute AOD value and variance. Among the sites in the clean group, Bandung is the most representative (Figure 1c). This side has the vast majority of its AOD lower than 0.5, and the ratio of variance of AOD to AOD is also concentrated around and below 0.5.

Urban regions have emissions that tend to be “quasi steady-state” over space and time, and hence the AOD time series is expected to have a relatively higher mean and lower variability when compared with other classifications (Cohen & Prinn, 2011), with the major source of variance being associated with local meteorology. The sites classified as urban all have an increasing SSA with AOD (maximum and average of correlation of 0.48 and 0.34 respectively, Figure 1g). Additionally, urban sites all have a decreasing AE with increasing AOD (range of correlation from −0.46 to −0.06, Figure 1h). These results are consistent with pollution events enhanced by atmospheric stagnation, high concentration oxidant conditions, and higher secondary aerosol formation, leading to more coating of existing particles and an overall larger particle size and more scattering radiative profile. The statistically most representative urban site is Taihu (Figure 1d), which hereafter will be used as the representative urban site.

Those sites classified as long-range transport, when compared with urban sites have an average and standard deviation of AOD which is both higher and less stable (0.52 for AOD, and a factor of 3.3 for the 90th percentile of the Standard Deviation of AOD to AOD ratio). These results are consistent with an initial BC emission source that include no subsequent source of BC, while all secondary growth all occurs via expansion of the shell slowly in-situ. In this case, more radiation is reflected by the larger shell, while the thicker shell reduces overall lensing, leading to less efficient secondary absorption by the BC core (Lack & Cappa, 2010). The most representative site with this classification is Palangkaraya (central Indonesia, Figures 1e and 1f), with an AOD value of 0.41, a standard deviation of 0.44, and a very negative correlation coefficient of −0.32 between AOD and AE. The time series of decreasing AE does not occur randomly, but instead occurs only during certain times of the year when the wind is coming from the westward. Further, the AOD time series at Palangkaraya has some peaks that are higher than the mean plus variability, with these special peaks only occurring in certain years. When they do occur, they are always from late Winter through Spring time and vary strongly in magnitude from event to event.

This explanation is consistent with highly aged black carbon from far away biomass burning sources know to occur in Sumatra or Borneo (Cohen, 2014) or elsewhere in Southeast Asia (Deng et al., 2020) being transported after undergoing significant aging, but are still significantly more absorbing than the local sources. The same phenomenon also occurs in other long-range transport regions identified here, including Taiwan.
Figure 1. (a) Geographic distribution of all four different site types; (b) Correlation between Aerosol Optical Depth (AOD) and Angstrom Exponent (AE) of Chiang Mai; (c) PDF of AOD and STD/AOD of Taihu; (d) PDF of AOD and STD/AOD of Bandung; (e) Correlation between AOD and SSA of Palangkaraya; (f) Correlation between AOD and SSA of Palangkaraya; (g) Correlation between AOD and SSA of Taihu; (h) Correlation between AOD and AE of Taihu.
and Northeast China, South Korea, and Nepal. A specific example of the vertically integrated wind field from 650 to 950 mb during the high AOD days from March 13th to March 18th, 2016 (Figure S1a) clearly shows the long-range transport observed in Hong Kong and Lulin is consistent with sources on the Southern edge of the Himalayas from Eastern India through Northern Vietnam, while on the remainder of the days in February and March 2016 (Figure S1b), the wind direction cannot transport eastward of Guangxi and instead blows into the Eastern Indian Ocean.

3.2. Aerosol Physical Properties

The results of the measurement-constrained modeled SSA at Chiang Mai is given in Figure 2a. The solution space of the particle sizes is relatively thin (32.3 nm core size, 80 nm shell size) and the slope is relatively small. It is worth noting that first, there are almost no solutions available when the core size and shell size are greater than 220 and 600 nm respectively, and second the total absorption of the particles is pretty high (SSA < 0.83). In specific at smaller particle sizes, the SSA of the net aerosol particles is found to gradually increase, although the median and maximum are found to respectively be 0.90 and 0.93.

The ASR of Chiang Mai is found to have a median of 2.85, and a range from 2.25 to 4. Assuming that the actual density of BC has an uncertainty range from 1.8 g/cm$^3$ to 2.3 g/cm$^3$ (Cohen & Wang, 2014), the corresponding AMR minimum ranges from 4.52 to 5.77, while the corresponding maximum ranges from 27.4 to 35. The solution spaces for core size, shell size, the ASR and the AMR are found to be uniform across all biomass burning sites.

The results of the measurement-constrained modeled SSA at Taihu is given in Figure 2b. The solution space of the particles is relatively broad (50.5 nm core size, 131.7 nm shell size) and the slope is relatively larger than Chiang Mai. It is noteworthy to see that the absorption of the particles is moderate (SSA > 0.86). In specific at smaller particle sizes, the SSA of the particles is found to gradually increase, although the median and maximum are found to respectively be 0.92 and 0.95.

The ASR of Taihu is found to have quite a wide range, with a median of 3.64, and a range from 2.80 to 4.75. The corresponding AMR is also broader with a range from 9.11 to 59.0. Not all sites classified as urban have as similar an ASR and AMR as the sites classified as biomass burning. This is mainly due to the fact that while some sources of urban aerosol particulates are continuous (i.e., power plants), others may vary (i.e., transportation and household burning), while the meteorology and photochemistry are both also more variable during the emissions times.

The results of the measurement-constrained modeled SSA at Bandung is given in Figure 2c. The solution space of the particles is relatively broad in terms of both core and shell, and the slope is relatively less useful to interpret the results than in Taihu. It is noteworthy to see that the absorption of the particles is low (SSA > 0.90). Overall, this site has a lower AOD, and a slightly wider range of ASR (from 2.81 to 5.25) and AMR (9.24–79.8).

The results of the measurement-constrained modeled SSA at Palangkaraya is given in Figure 2d. This site has the highest SSA value (>0.92), AMR (3.31–6.00) and ASR (15.3–120.) among the sites in this group. These results are consistent with long-range transport, wherein the aerosols were in the air longest and underwent the most amount of physical and chemical processing. The timing of the peaks in AOD is also consistent with the timing of the burning season in western Indonesia as well as super long-range transported burning from northern Southeast Asia. Similarly, the timing of the peaks in AOD at another long-range transport site (Chen Kung university) are found to occur at the same time as fire sources in Eastern India, Myanmar, and Northern Thailand, and therefore provides a second consistent story (Lin, Cohen, Wang, Lan, & Deng, 2020; Wang et al., 2020). Other sites identified as long-range transport sites are found in South Korea and Northeast China.

PDFs of the core size, shell size and total particle size of each site are given in (Figures 3a–3d, 3g and 3h). For BC, the proportion larger than 200 nm in Chiang Mai (43%), which is much higher than elsewhere. Taihu has the second largest percent of BC larger than 200 nm (26%). The remaining sites have 7% or less BC larger than 200 nm (Figure 3g). These results are consistent with biomass burning and urban sources, where larger particles of BC tend to be produced. In terms of the shell size, Chiang Mai, Bandung and
Figure 2. The MSSA computed (Colorbar is SSA) using a Core-Shell MIE model constrained by 4 wavelengths of AERONET measurements at the six most representative sites: (a) Chiang Mai; (b) Taihu; (c) Bandung; (d) Palangkaraya; (e) Beijing; (f) Taiping.
Figure 3. PDF statistical analysis of core size, shell size and total aerosol particle size for all six most representative sites: (a) Chiang Mai; (b) Taihu; (c) Bandung; (d) Palangkaraya; (e) Beijing; (f) Taiping; (g) PDF statistical analysis of core BC size; (h) PDF statistical analysis of shell sulphate/nitrogen size.
Palangkaraya all have a significant proportion of sulphate less than 400 nm, ranging 22%, 26% and 27% respectively (Figure 3h), consistent with the source regions not having a large amount of nitrate or sulphate to condense, and also consistent with measured field results in other ultra-long-range transport environments (Kodros et al., 2018). In Taihu, the proportion of sulphate less than 400 nm is only 9%, while the proportion between 600 and 800 nm is the largest at 63%, which is consistent with large amounts of locally emitted and secondarily produced nitrate and sulphate.

3.3. Special Cases—Impactful Urban and Long-Range Transport

Some sites have different set of characteristics during a subset of the total time series from the overall set of characteristics, allowing multiple categorizations as a function of the times of the year. In particular, some sites generally classified as urban or clean also have a significant but small fraction of their total days that behave either like biomass burning or long-range transport. The two most obvious such cases are Beijing and Taiping Island.

The Beijing site is geographically located in an urban area; however, the classification is frequently found to have a biomass burning characteristic. In specific the mean value of AOD in Beijing is higher than Chiang Mai, and the 50th percentile of the STD of AOD is slightly lower than in Chiang Mai. This shows that Beijing has a higher average AOD than in Chiang Mai and less overall variation, consistent with its being an urban site. Similarly, the results of the MIE model show that Beijing generally matches well with Taihu (Figure 2e). However, the larger amount of variability of AOD, wider SSA range leading to lower SSA values than observed at Taihu, all consistent with the wider size of BC core and shell, and in specific possessing a set of larger core sizes and smaller shell sizes than Taihu, yet consistent with the range formed due to a linear combination of urban and biomass burning characteristics. In particular, the solution space of the particle is found to be the widest of all of the sites in this paper with a 61.3 nm core size range and 154.4 nm shell size range. More similar to the urban characteristic, at smaller particle sizes the SSA of is found to gradually increase, with the median and maximum of the solution space respectively 0.91 and 0.95. The ASR of Beijing is also found to have a wide range, with a median of 3.5, and a range from 2.5 to 4.8. The corresponding AMR is also broad with a range from 6.4 to 59. The result of the probability density function of Beijing shows in Figure 3e. For BC core, the proportion of core larger than 200 nm is 68%, while the shell has 38% of measurements between 400 and 600 nm and 50% between 600 and 800 nm.

Taiping Island is classified as clean, which is consistent with its location in South China Sea more than 100 km from other land. However, if analyzing the data from February and March separately, a new classification of long-range transport is determined. The results of the MIE model show that while the overall characteristics of the solution space of aerosol properties at Taiping Island is similar to Palangkaraya (Figure 2f), overall, the particle size is wider than both Palangkaraya and Bandung (55.4 nm core size, 204.2 nm shell size). Furthermore, the total absorption has the lowest value of all of the sites analyzed (SSA>0.93). In specific at smaller particle sizes, the SSA of the net aerosol particles is found to gradually increase, although the median and maximum are found to respectively be 0.96 and 0.99. The corresponding ASR and AMR at Taiping Island both have a wide range, with a median of 4.6, and a range from 3.38 to 6 for the ASR and a range from 16.4 to 555 for the AMR, consistent with the range of particle sizes spanning both clean and long-range transport types. The result of the probability density function at Taiping Island (Figure 3f) show clearly that the BC core sizes are relatively small (the proportion of core larger than 200 nm is 0%) and a very thick shell (with 34% between 400 and 600 nm and 37% between 600 and 800 nm) respectively. These results are consistent with the air flow from Eastern India and Northern Southeast Asia containing large amounts of fires which are then strongly advected in the middle troposphere to the east annually during February and March.

4. Conclusions

This work as demonstrated an objective, clear, and simple way to systematically analyze the regional characteristics of aerosol size and mixing state based on measurements from AERONET sites located throughout East Asia, Southeast Asia and South Asia. In specific, use of multi-wavelength measurements of AOD, SSA and AE, as well as the standard deviation of AOD and the ratio of the standard deviation of AOD to
AOD reveal that aerosols in Asia have at least 4 different fundamental characteristics, corresponding respectively to: urban, biomass burning, long-range transport, and clean.

Applying a MIE model with a Core-Shell assumption, and constraining the results with the 80% central range of measured SSA across multiple wavelengths, allows for a solution set of core and shell sizes to be obtained for each AERONET site. These solutions require both the core and shell sizes, as well as other aerosol properties including the mixing state to be consistent with all of the measurements. The constrained aerosol properties are further found to be consistent within each of the four categories of aerosol types, and are not otherwise mixed during the same place and time between the different categories of types, showing that these classifications are robust. It is found that the largest BC core sizes are associated with biomass burning regions (the AMR ranges from 4.5 to 35 in Chiang Mai), and the thickest shell sizes are found for long-range transport regions (the AMR in Palangkaraya ranges from 15.3 to 120), consistent with primary aerosols which have undergone significant in-situ processing and coating by reflective sulphate and other species. Furthermore, aerosols at biomass burning sites are more high absorbing, due to the higher fraction of black carbon to total aerosol emissions and little time for in-situ processing.

Two locations were found which had characteristics of different aerosol types, and can be only successfully separated during different times of the year. Heavily urban Beijing is found to be consistent with both urban and biomass burning properties (the overall size range is far wider than in Taihu, with 61.3 nm core size range and 154.4 nm shell size range). Very remote Taiping Island is found to have both clean and long-range transport characteristics (AMR ranges from 16.4 to 555). The number of sites with such characteristics is increasing due to the increasing diversity of aerosol sources in terms of both space and magnitude. These current results confirm that considerable new information can be gleaned from existing AERONET measurements, as well as more evidence for the effects of changes in the attribution of biomass burning, urbanization, and long-range transport of sources, all of which are known to be rapidly changing and not well characterized throughout Asia.

Conflict of Interest
The authors declare no conflicts of interest relevant to this study.

Data Availability Statement
All processed data and codes are freely available for download at https://doi.org/10.6084/m9.figshare.14077169.

Acknowledgments
We would like to acknowledge the PIs of the AERONET instruments for providing the remote sensing measurements be downloaded from https://aeronet.gsfc.nasa.gov/cgi-bin/webtool_inv_v3. The work was supported by the Chinese National Young Thousand Talents Program (Project 41180002), the Chinese National Natural Science Foundation (Project 41030028; Project 42075147), and the Guangdong Provincial Young Talent Support Fund (Project 42150003).

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