

Combining Multi-Wavelength AERONET SSA Retrievals with a MIE Model to Quantify the Size of Absorbing Aerosols and the In-Situ Lifetime of Sulfate

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Abstract

Energy, transport, urbanization and burning are responsible for changes in atmospheric BC. This work uses direct solar atmospheric column measurements of single scatter albedo [SSA] retrieved at multiple wavelengths from AERONET at 68 Asian sites over 17 years. A MIE model is solved across the wavelengths using a core-shell mixing approximation to invert the probabilistic BC, shell size, and UV SSA. Orthogonal patterns are obtained for urban, biomass burning [BB], and long-range transport [LRT] conditions, which are used to analyze and attribute source types of BC across the region. Large urban areas (thought to be dominated by urban BC) are observations during targeted times (shorter than seasonally) to yield significant contributions from non-urban BC. BB and LRT are observed to dominate Beijing and Hong Kong 2 months a year. LRT is observed during the clean Asian Monsoon season in both Nepal and Hong Kong, with sources identified from thousands of kilometers away. Computing the shift in shell size required to constrain the results approximates secondary aerosol growth in-situ, and subsequently aerosol lifetime, which is found to range from 11 days to a month, implying both a significant amount of BC above the boundary layer, and that BC generally has a longer lifetime than PM_{2.5}. These findings are outside of the range of most modeling studies focusing on PM_{2.5}, but are consistent with independent measurements from SP2 and modeling studies of BC that use core-shell mixing together with high BC emissions.

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Key Points:

- Urban sites throughout Asia are impacted not only by local sources but also biomass burning and sources conveyed long-range transport.
- Quantifies the age and attributes the sources as a function of time based on multiple wavelength analysis.
- Uncertainty analysis indicates that the number of very small particles and total absorption are underestimated by surface measurements.

Abstract

Energy, transport, urbanization and burning are responsible for changes in atmospheric BC. This work uses direct solar atmospheric column measurements of single scatter albedo [SSA] retrieved at multiple wavelengths from AERONET at 68 Asian sites over 17 years. A MIE model is solved across the wavelengths using a core-shell mixing approximation to invert the probabilistic BC, shell size, and UV SSA. Orthogonal patterns are obtained for urban, biomass burning [BB], and long-range transport [LRT] conditions, which are used to analyze and attribute source types of BC across the region. Large urban areas (thought to be dominated by urban BC) are observations during targeted times (shorter than seasonally) to yield significant contributions from non-urban BC. BB and LRT are observed to dominate Beijing and Hong Kong 2 months a year. LRT is observed during the clean Asian Monsoon season in both Nepal and Hong Kong, with sources identified from thousands of kilometers away. Computing the shift in shell size required to constrain the results approximates secondary aerosol growth in-situ, and subsequently aerosol lifetime, which is found to range from 11 days to a month, implying both a significant amount of BC above the boundary layer, and that BC generally has a longer lifetime than PM_{2.5}. These findings are outside of the range of most modeling studies focusing on PM_{2.5}, but are consistent with independent measurements from SP2 and modeling studies of BC that use core-shell mixing together with high BC emissions.

Plain Language Summary

Energy, transport, urbanization and burning are responsible for significant changes in atmospheric Black Carbon aerosol [BC]. This study analyzes atmospheric column measurements

using different parts of the solar spectrum with a physical model, to describe the solution set of BC and coating sizes in the atmosphere. This information is then used to make different aerosol types, including urban, biomass burning, and aerosols which have been in the atmosphere for a very long time. These type profiles are applied throughout Asia to analyze the sources of BC. While many sites are as expected, most large urban areas are observed to have both biomass burning and long-range transport types found when looking over short time periods which occur annually. Next, even during heavy Monsoon rains, urban areas are found to have significant amounts of long-range transported BC. The lifetime of the BC is also obtained from the growth in the shell size, demonstrating that BC stays in the atmosphere longer than current studies focusing on PM_{2.5} can observe. Focusing models more specifically on BC, formulating different controls during different periods of the year, and better studies of BC emissions could all yield improved understanding of BC and its effects.

1 Introduction

Black carbon (BC), organic carbon (OC) and dust (or Absorbing Aerosols - AA) can strongly interact with solar radiation, leading to impacts on the atmospheric radiation budget, climate, water cycle, and more (Jacobson, 2001; Menon et al., 2002; Wu et al., 2008). Black Carbon can absorb solar radiation strongly in the visible and infrared. Small changes in BC loadings can cause a significant change in the local solar radiation absorption and hence altering the regional and global climate (Ramanathan et al., 2001a, 2001b). This interaction critically relies on the particle size distribution, chemical composition and mixing state in-situ (Jacobson, 2001). The two most significant difficulties in estimation of the climate effects of black carbon are the variable physical-chemical properties of BC-containing particles (Miyakawa et al., 2017) and the heterogeneous distribution (Bollasina et al., 2013; Shen Z et al., 2020; Cooke et al., 2002; Lu et al., 2011). While BC itself is water-insoluble once it has been in the atmosphere for a long enough time, it will become coated (Zhang et al., 2016; Cohen and Wang, 2014) in turn becoming hygroscopic (McMeeking et al., 2011) and therefore having the potential to impact climate via wet and ice clouds (Penner et al., 1992; Penner et al., 2001; Kuwata et al., 2007). Due to its long lifetime in the atmosphere, BC has also been known to travel very long distances, impacting regions thousands of km away from the source region, and undergoing in-situ processing such that its absorbance and impact on clouds may both be enhanced, further impacting the radiative balance over large scales (Liu et al., 2011; Zhang et al., 2015; Prasad et al., 2018; Guha et al., 2015; Wang et al., 2009). Taking all of these factors into consideration, the current range of radiative forcing estimates of BC (at TOA) range from +0.13 to +1.4 W/m² (Haywood and Shine, 1995; Haywood et al., 1997; Haywood and Ramaswamy, 1998; Myhre et al., 1998; Penner et al., 1998; Cooke et al., 1999; Jacobson, 2000; Wang, 2004; Wang et al., 2014; Jacobson et al., 2001; Bond et al., 2013; Cohen et al., 2011).

Attempts have been made to elucidate particle size, mixing state, emissions, and aging over highly polluted regions in Asia using different models and methods (Salam et al., 2012; Quinn et al., 2004; Kompalli et al., 2021; Han et al., 2013; Streets et al., 2003; Kühn et al., 2014; Xu et al., 2015; Song et al., 1999). However, at the present time there are few if any studies that have comprehensively focused on the long-term properties of the particle size and source (i.e. distance to a biomass burning or urban source) and aging (i.e. how long the aerosol has undergone processing and deposition in-situ). Studies which have analyzed aerosols using remote sensing have tended to focus on the visible and IR radiative bands, with measurements from the UV band scarcely used (Corrigan et al., 2006). In specific, many present remote sensing

based studies merely focus on a fine mode and coarse mode (Eck et al., 2010; Koulouri et al., 2008; Zhang et al., 2017; Kleidman et al., 2005). Attempts have also been made via field observations. Schwarz et al (2008) used an airborne single-particle soot photometer to quantify BC aerosol microphysical state in fresh urban and BB emissions. These differences contribute to significant impacts on the BC size distribution and concentration, when observed using measurements based on particulate absorption (Sato et al., 2003). However, results such as these from both remote sensing and field studies are not considered in most chemical transport and aerosol models today, which tend to not assume a Core-Shell approximation which in turn tends to underestimate the overall absorption profile, requiring scaling to match optical properties such as SSA, AAOD and size information (Myhre et al, 1998; Cooke et al., 1999; Jacobson et al., 2000; Wang et al., 2009; Cohen et al 2011). Consequently, these models tend to underestimate ultra-long-range transport and non-local polluted conditions, frequently induced by vertical transport and aging associated with underestimated absorption (Wang et al., 2020). It has been demonstrated that a Core-Shell model fits best with aerosol properties in heavily polluted region such as East, Southeast and South Asia where sulfate or nitrate can coat on BC in a short time after it has been emitted, consistent with the fact that in these parts of the world biomass burning sources and urbanization are located in close proximity to each other (Cohen and Wang, 2014; Peng et al., 2016; Zhang et al., 2016; Cohen et al., 2017).

To achieve a better understanding of AA and its impact on the atmosphere, this work incorporates multi-spectral atmospheric column high frequency measurements and inversion products from AERONET including AOD and SSA, as well as mixing state and size measurements from ground station, in connection with a inverse constrained variance maximization approach to reveal size and absorption properties of aerosols in Asia. A specific focus is made on the added value of using measurements in the UV bands which provides deeper support for and quantitative insight into the loadings and sizes of smaller particles, allowing for better characterization of both aging as well as fresher particles. The inversely constrained particulate core and shell sizes based on the SSA values from the Mie model and constrained by AERONET, allow for a new categorization of aerosols based on whether they are emitted locally in Urban areas [Urban], have undergone Long-Range Transport [LRT], or are emitted locally by Biomass Burning [BB]. The approach also allows a statistical solution space to indicate the range of mixtures of these source types which would also be capable of being physically realistic, taking into the specific particle size (both core and shell) distribution. This allows the use of typical and well classified sites to be used to improve understanding of different sources and their contributions to the atmospheric loading, as well as impacts on radiative forcing and subsequently on climate change. Since these are based on column measurements of the entire atmosphere, they provide important support to further promote improvement of models, in terms of capturing the total atmospheric loading, attribution of extreme or abnormal events, and impacts of high-frequency changes on the overall performance of models, all of which are cited as important weaknesses of current aerosol modeling systems. Some specific scientific points of interest discovered include quantification of significant events in which mixing occurs between an expected and an unexpected source, detection of missing/previously unidentified sources, details about changes in the optical and size properties of aerosols which are otherwise obscured when only looking at a PM_{2.5} or Fine Mode type of aerosol constraint, and quantifying of different aging rates occurring between BB and Urban cases. It is finally hoped that ongoing calculations incorporating the quantitative approach explicitly employed here will allow for further expansion and insights to be gained and for more

multi-wavelength measurement platforms of aerosol absorption to be further incorporated in the future, allowing for further understanding, advances in attribution, and improvement of models and impacts on people, health, and the climate system.

2 Data and Methods

2.1 AERONET Data

This work uses Aerosol Optical Depth (AOD) and Single Scattering Albedo (SSA) from March 1997 to May 2017 using ground-based Aerosol Robotic Network (AERONET) observations. AERONET uses CET318-TS9 instruments to provide information about the spectral sun irradiance and sky radiances at multi-bands including at wavelengths of 340 and 380 (AOD only), and 440, 670, 870 and 1020nm (both AOD and SSA) (https://aeronet.gsfc.nasa.gov/new_web/data_description_AOD_V2.html). This work uses level 2.0 cloud screened and quality assured data (Holben et al., 2006), focusing on the time variations and loading/magnitude of the daily mean measurements over the entirety of the time series which have three or more individual measurements available within the time of one calendar day. To avoid the case where there is a weak signal and ensure sites have sufficient data to capture the feature of each site, all AERONET sites were filtered previous to classification, only retaining sites that have 100 or more days of data in addition to at least one of three conditions: high mean AOD, high extreme event AOD, or highly variable AOD. If a site has more than 300 days of total data (Wang et al., 2021; Dubovik and King et al., 2000; Dubovik et al., 2002), then the site is also used, even if it does not meet one of the three conditions. After filtering 68 sites remain for further analysis throughout East Asia, Southeast Asia, South Asia, Eastern Russia, and Australia. Furthermore, other than sites located in Western India and Western China, the remainder of the sites in this analysis do not generally have a significant dust loading, and therefore the major source of absorption is assumed to be black carbon (BC), allowing this work to assume a direct relationship between BC and SSA (Cohen and Wang, 2014).

2.2 Mie Model

A Mie model is used to connect the radiative measurements and observations from AERONET with approximations of the various optical, mixing state, and implied chemical properties of the aerosols. Mie Theory solves the aerosol optical properties based on a given size and optical distribution of particles, following Bohren and Huffman (1983). It assumed the time variation of the field is $\exp^{-i\omega t}$, leading exclusively to positive imaginary parts of the refractive index corresponding to absorbing media. Although externally, internally and core-shell mixtures are frequently used to characterize particle morphologies and chemical compositions (Cheng et al., 2008; Lin et al., 2013; Tao et al., 2019; Zhang et al., 2017), this work assumes that all the particles have a Core-Shell structure, which has been demonstrated to be reasonable in urban areas in Asia based on both modeling and measurement studies (Cohen et al., 2011; Cohen and Wang, 2014; Zamora et al., 2019), as well as being compatible with the AERONET inversion technique (Dubovik, 2002).

Using particle size and refractive indices, this work computes the extinction coefficient (Q_{ext}), scattering coefficient (Q_{sca}) and absorbing coefficient (Q_{abs}). The resulting single-scattering albedo (SSA) is calculated based on the equation (1) (Hansen et al., 2009).

$$SSA = \frac{SCATTERING}{EXTINCTION} = \frac{Q_{sca}}{Q_{ext}} \quad (1)$$

Since AERONET does not report the SSA in the UV band, this work computes the SSA field using the MIE model at 340nm using a core-shell approximation where the aerosols contain a pure BC core with a scattering (i.e. Sulfate or Nitrate) coating, where the core refractive index is $2.0+1.0i$ (Schuster et al., 2005) and the shell refractive index is $1.52-5*10^{-4}i$ (B Aouierats et al., 2010). This is physically consistent with the facts that many primary aerosols in these regions are organic and that they age rapidly as a function of their time in-situ (Cohen and Wang, 2014; May et al., 2015; Song et al., 1999). The model computes particle sizes of the core in steps of $0.01\mu\text{m}$ from $0.05\mu\text{m}$ to $0.50\mu\text{m}$, and the shell in steps of $0.01\mu\text{m}$ from $0.01\mu\text{m}$ to $0.80\mu\text{m}$.

2.3 Copernicus Atmosphere Monitoring Service (CAMS) SO_2 data

CAMS global reanalysis product provides monthly average SO_2 at $0.75^\circ \times 0.75^\circ$ resolution (<https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-reanalysis-eac4-monthly?tab=overview>). This study uses data from 2019 data over 7 vertical levels from the surface to 700hPa. Additional measurements of SO_2 at the surface are obtained over China from the Ministry of Ecology and Environment of the People's Republic of China.

2.4 Single Particle Soot Photometer (SP2) Measurement

SP2 measures individual aerosol particles. These measurements are capable of determining the optical size and characteristics by scattering and absorption using 1040nm radiation, as well as induced laser incandescence of pure carbon-containing cores. In the real atmosphere at these wavelengths, the major source of absorption corresponds to BC (Bond, T.C., et al., 2013; Bond and Bergstrom, 2006; Bond, 2001; Penner, 1994), which can then be converted into a core diameter (D_c) using a mass equivalent diameter ($\rho=1.8\text{g/cm}^3$ for atmospheric BC) and assuming that BC is spherical (Martins et al., 1998; Smith and Grainger, 2014; Jacobson and M.Z., 2000, 2001). The optical diameter of the non-BC portions of the particle (D_p) is calculated using Mie theory, and presented as (D_p/D_c).

This study used SP2 data measured by the Institute of Atmospheric Physics Beijing ($39.58'28''\text{N}$, $116.22'16''\text{E}$) observed from 2016.11.10 to 2017.09.15. This specific location is located very close to an AERONET station in Beijing (39.977N , 116.381E) which has measurements in 2001 (March to May), 2015 (September to December) and 2016 (January to July). There is an overlap between these two datasets from 2016.11.10 to 2017.01.12, which hereafter is used for the comparison between D_p/D_c inverted from AERONET and measured by SP2.

2.5 In-situ lifetime calculation

Considering that a minimum concentration of sulfuric (or nitric) acid is required before BC can age, the lifetime of SO_2 provides an excellent proxy to approximate the time in-situ. This can be quantified following Equation 2 where M is the SO_2 concentration, and k is the rate constant.

$$\frac{D[M]}{Dt} = -k[M] \quad (2)$$

The solution of Equation 2, after rearrangement of the terms is given in Equation 3 where k equals to $1/\tau$, t is the in-situ time the particle spent in the atmosphere, τ is the e-folding time of SO_2 in-situ, $M(\text{env.})$ is the mean environmental concentration of SO_2 encountered by the particle, and $M(\text{growth})$ is calculated based on the [BB] Profile (more details in 4.1), based on the average atmospheric concentration of sulfate in the region studied being about $2.9\mu\text{g/m}^3$.

$$t = \frac{-\ln \frac{M(growth)}{M(env.)}}{\tau} \#(3)$$

In the background case, the assumed lifetime of SO₂ is 1.56 days (Pham et al., 1995; Chuang et al., 1997; Chin et al., 1996; Croft et al., 2014; Restad et al., 1998). In the urban case, the lifetime is expected to be shorter, since the loading of OH is higher and conversion to H₂SO₄ gas and subsequent gas to particle conversion is faster (Cohen et al., 2011; Liu et al., 2018; Saiz-Lopez et al., 2017; Saxena et al., 1987). This work recalculates the e-folding time assuming that Beijing is a representative site for urban chemistry. Based on the Beijing high time profile obtained by extracting the size value in the peak period (25 days in total) and the ground measurements of SO₂ from the China EPA Beijing station, the urban lifetime is computed to be approximately 0.98 days.

2.6 Statistics and analytics used in this work

Considering that many sites in the region are undergoing rapid change and might have different characteristics during different time periods, a method is derived to look at extremely polluted days separately from the non-polluted days. This selection of heavily polluted days is made by defining the peaks of the data greater than the mean plus one standard deviation, removing these, and re-iterating 3 times. The net aggregation of those days is herein considered heavily polluted.

3 Analyses of selected regions

3.1 Time series of selected cases

The temporal characteristics of daily average AERONET AOD (340nm) over different times are given in Figure 1, with non-polluted values shown in green, while those considered heavily polluted are uniquely represented as blue, black, and red respectively based on being filtered during the first, second, and third passes respectively. Common features observed include relatively regular signals of an annual peak and trough with a roughly similar start time, end time and duration. Some sites occasionally have large values and otherwise are relatively low or moderate in terms of AOD, but without any clear pattern (these patterns do not have any clear start or end time, duration, magnitude, or extremum), leading to the changes being associated with some process that locally varies significantly from time to time, consistent with long-range transport based events. In these cases, the average AOD during the non-peak times tends to be not high. In addition to the above situations, sometimes the distribution of AOD is completely irregular, with no obvious pattern during either the cleaner periods or the peak periods, and no obvious pattern in terms of high or low magnitude. In this case, the source region is more likely to be urban in nature, because urban emissions exist year-round without a large amount of variation, although small changes due to the day of the week, the season of the year, and local meteorology combine to provide temporal variability.

Chiangmai is the biggest city in northern Thailand and is the only location in Northern Thailand, Myanmar, or Laos with an industrial presence (population of 127,000). However, based on previous work (Lin and Cohen et al., 2020; Wang and Cohen et al., 2020; Cohen et al., 2017), this region generally has industrial and urban signals of emissions which are very small, instead having a signal dominated by biomass burning. Based on Figure 1a, the time series of

Chiangmai has a very unique feature with most of the year being relatively clean, and a short but intense time during which it is extremely polluted. The peak always occurs during roughly the same time (From February through 19 April) every year over the entirety of the 10 years of measurements, and furthermore a peak never occurs outside of this time. Given this and the previous works identified above, Chiangmai is considered the most representative region with respect to biomass burning.

Taihu is a large lake located partially in Wuxi (7 million people) and partially in Suzhou (11 million people), cities which have the second and third highest per capita PPP in China. The lake is located in the middle of the Shanghai, Nanjing, Hangzhou conurbation. For this reason, the region has a huge amount of individual vehicles, factories, manufacturing and other economic activity, yet is not adjacent to any specific road or factory. As such, it is quite representative of a region in which the major source of pollution is urban, as has been shown by previous works (Huang et al., 2021; An et al., 2021; Logan et al., 2013; Chen et al., 2020). As shown in Fig. 1(b), a time series of the AOD tends to be high on average, and contains a large amount of variability that appears both random and not intensely concentrated during any specific periods of time.

Palangkaraya is a small city located in southwestern Borneo, which is surrounded by forests and peatlands. The city itself is not a major source of emissions, and while the surrounding land had been frequently burned in the past, today it consists of plantations without any more burning. The data available at this site ranges from 2012 through 2016, during which the location region was already transformed into plantation (nearly no local sources), but which was surrounded by generally high amounts of biomass burning hundreds to thousands of kilometers away coming from elsewhere in the Maritime Continent in 2012 and 2015, as demonstrated by the time series data itself, and previous studies (Deng et al., 2020; Cohen, 2014). The relatively low mean and high standard deviation of AOD show a consistent pattern whereby the region is impacted by long-range transport of biomass burning from other regions. Consistently there is observed to be a variable start and end time changing year to year, starting as early as early August and ending as late as early November. Otherwise, this region tends to have very low levels of BC and generally is clean. This combination of conditions leads to it being an excellent candidate to represent long-range transport.

Beijing as a typical megacity, with a population more than 18 million, and considerable sources from high tech, industry, and government. Based on the total available three year's data in 2001, 2015 and 2016 the features in general appear to be like Taihu, consistent with the understanding that Beijing is also urban. The value of AOD during the highest period is about 3.2, while most of the second pass and third pass data fall in the range of 1.5~3, with the concept that the temporal occurrence of peak events is random but tending to be of similar magnitude. In addition, there is a considerable amount of the total data (37%) which behaves like long range transport. This tends to occur during spring, but only occurring in specific years (Han et al., 2015; Wang et al., 2015; Wang et al., 2004). This is consistent with the fact that transport from western China (Shanxi, Shaanxi, Ningxia, etc.) and its large amount of energy and downstream energy emissions sources, or northeastern China (and its heavy biomass burning and industrial emissions) does occur at times when spillover occurs over the mountains surrounding the edges of Hebei and Beijing (Li et al., 2022; Qin et al., 2022; Wang et al., 2020).

The temporal characteristics of AOD in the megacity of Hong Kong form another very interesting case, as observed in Figure 1(e). The value tends to have a relatively high mean with most measurements between 1.0 and 1.5, and seemingly occurring randomly throughout most of

the year. This result is consistent with its large industrial, transportation, and shipping sectors, and located in the middle of the highly populated and heavily industrialized Pearl River Delta region. However, there also is a recurring increase in the overall AOD (between 1.5 and 2.6) that occurs in October. These results are unexpected in Hong Kong when compared with many local studies (Tan et al., 2016; Fang et al., 2018; Xu et al., 2022), however they are consistent with recent studies which have focused on the changes in the Monsoon (Yang et al., 2006; Yao et al., 2008; Lau et al., 1997) and biomass burning increases throughout Northern Southeast Asia (Lin et al. 2020; Wang et al., 2021). A major difference is that these studies which have observed these changes are whole-atmospheric studies, not specifically related to measurements at the surface.

Lumbini in South Asia is a rapidly developing urban area in Nepal. The signal corresponds strongly with known urbanization, a high population density, and many small and randomized sources including rubbish and trash burning, small brick and cement kilns, and a growing proliferation of combustion-based transport options. The strength of the red signal varies in intensity and magnitude from an observed relative maximum in the middle and end of 2014 as development rapidly accelerated (Rupakheti et al., 2020).

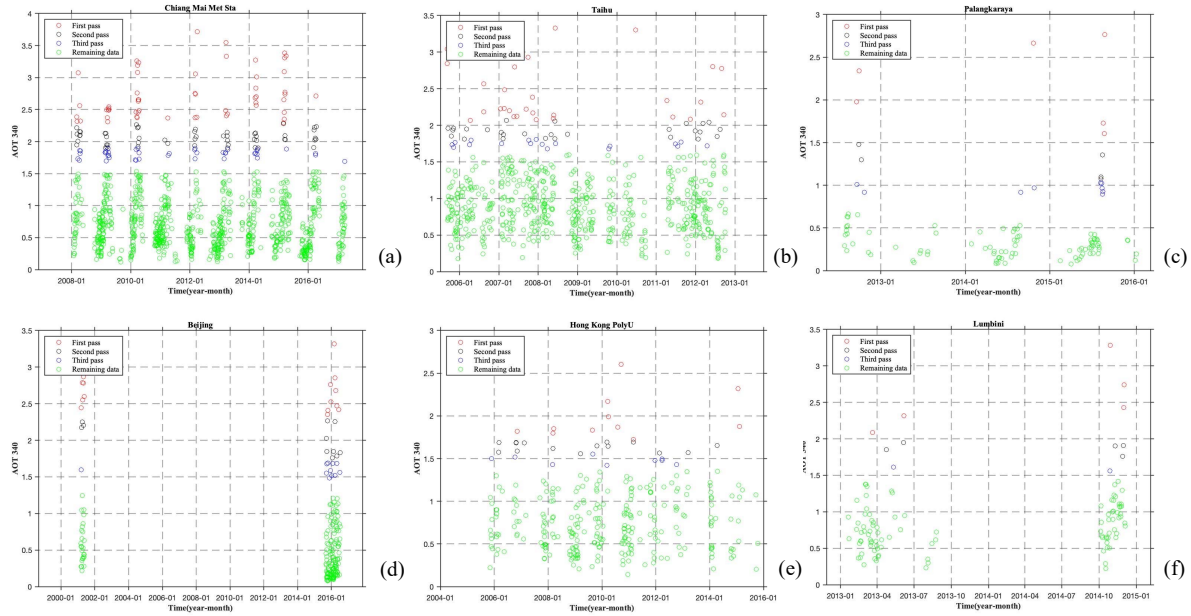


Figure 1. Time series of AOD at 340nm of different sites. (a) Chiang Mai Met Sta; (b) Taihu; (c) Palangkaraya; (d) Beijing; (e) Hong Kong PolyU; (f) Lumbini

3.2 Typical Features of Well-Defined Source Regions

To calculate the typical characteristics of biomass burning, the distribution of solutions of size pairs of core and shell radius corresponding to the intersection across all different wavebands of the measured SSA in Chiangmai during the high season will subsequently be used, hereafter called [BB], see Fig.2a. Since the burning in Chiangmai is local, the profile may be used to represent other regions which also have a significant local biomass burning source. If a region has a good match with the biomass profile, it implies that the local aerosol profile is influenced by local biomass burning. When a region has a good match with the biomass profile after being shifted upwards, it is consistent with a region that is impacted by biomass burning

which has undergone growth in-situ while undergoing transport between its source and where it was ultimately measured. In this case, an approximation of the age of the biomass burning can be obtained indirectly by considering how much upward shift is required to make the best fit, where the shift quantifies the amount of shell growth while in-situ.

Similarly, the distribution from Palangkaraya is used to compute the conditions under which long-range transport occurs, subsequently called [LRT] (Wang et al., 2021), see Fig.2b. The AOD pattern of Palangkaraya shows a combination of relatively low mean and a high standard deviation of AOD, which starts and ends around the same time every year. Local measurements of CO from the long-term WMO station in the same city also has a similar signal (Aouizerats et al., 2014). Basically, the local environment is nearly pollution free, except for when biomass burning occurs on Sumatra, Borneo, and other islands in the region, which then transport to the site. For these reasons, measurements at this site are an excellent candidate to represent sites typically impacted by long-range transport.

Finally, the distribution of solutions of size pairs of core and shell radius corresponding to the measured SSA in Taihu referred to as [UR], see Fig.2c. Since Taihu is near the center of the world's largest conurbation, yet located in the middle of a lake, highways and other immediate point sources are reduced, making the profile highly representative of a generalized urban area.

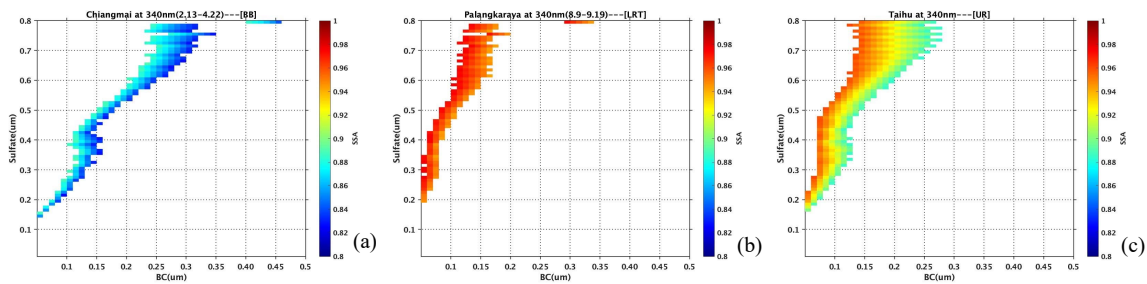


Figure 2. BC core and Sulfate shell size distributions, with corresponding SSA (color) at the three well-defined source sites: Chiangmai [BB], Palangkaraya [LRT], and Taihu [UR].

3.3 Quantifying New Spatial and Temporal Relationships in BC Properties Induced by Various Natural and Anthropogenic Forcings

The climatologically constrained SSA distribution in the 126 million population Greater Bay Conurbation (hereby defined based on measurements from the Hong Kong PolyU site) is given in Fig.3a. The climatological core and shell size distributions show mostly urban characteristics, as expected from the high population density, large and growing economic footprint in this region. However, the geographical location of Hong Kong places it in a region influenced by the Asian monsoon, in a dense urban area surrounded by significant agriculture, and upwind from the rapidly developing and transforming nations in ASEAN.

As expected, the overall climatology of aerosol properties in Hong Kong, based on the magnitude of the SSA (i.e., color) and size distribution of both the core and shell (i.e., shape of the SSA distribution) derived over this station is mostly dominated by [Urban], with very small amounts of [LRT] and [BB] (Fig 3a). However, both inter-annual and intra-annual variability are observed to be very important in this region.

A first case is observed in Fig 3b-c. Monsoon theory indicates that there is a strong Southeast wind occurring in July and August, leading to flow from the clean South China Sea to the Chinese Mainland, with Hong Kong being one of the first points of contact (Yang et al., 2014;

Lawrence and Lelieveld, 2010; Webster and Yang, 1992; Zhu et al., 2012). It has generally implied that during this time, the air should be extremely clean. And when analysis is done using all days from July and August as given in Fig 3b, the local signal is observed along with an LRT signal. However, a look at only the August data reveals a pure LRT signal (Fig.3c). Since the winds are not changing during this period of time, therefore the results are consistent with a significant change in the aerosol loading on the southern edge of the South China Sea. This is consistent with known massive fire events which occur starting in August on a nearly annual basis (Deng et al., 2021; Cohen, 2014; Cohen et al., 2017; Wang et al., 2020; Li et al., 2020; Chen et al., 2017). This data offers a first independent characterization of significant long range transport of biomass burning absorbing aerosol over 2000km from Borneo to Hong Kong.

Furthermore, it can be seen in Figs 3d-g that in 2006 the [Urban] source occupies the vast majority of the September/October/November signal, with a very small amount of BB mixed in, while the September/October/November signal in 2014 is found to mostly consist of LRT with only a small amount of Urban. Similarly, the January signals in 2014 and 2006 are also vastly different from each other, with 2014 being mostly Urban in nature, and 2006 having an extremely strong BB signal. All of these results are consistent with the fact that 2006 was a strong El-Nino and 2014 was a mixture of a budding La-Nina occurring at the same time as a negative IOD (Logan et al., 2008; Deng et al., 2021; Cohen, 2014; van der Werf et al., 2008; Rinsland et al., 2008; Chi et al., 2019; Xie and Fang, 2019).

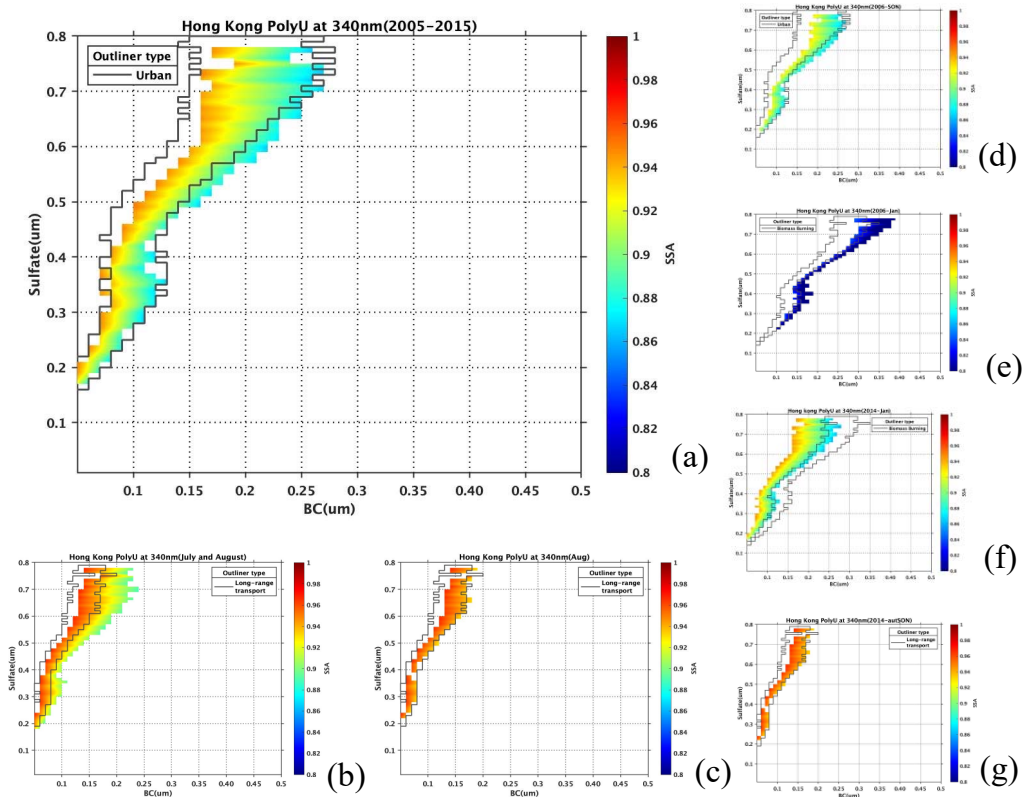


Figure 3. Constrained distribution of SSA from model result of Hong Kong PloyU during different special time periods with urban, Biomass Burning and Long-range transport profile.

Lumbini is located in Nepal and represents the densely populated Southern Slopes of the Himalayas, a region impacted by biomass burning, the Monsoon, and very large variance in aerosol loadings from ultra clean to some of the most polluted conditions on the planet. Fig 4 shows the constrained SSA distribution during three seasons where there are vastly different conditions observed.

First during the monsoon season (JJAS) the majority of the sources are influenced by long range transport, consistent with the squeezing of air from the Continent into the foothills of the Himalayas (Dumka et al., 2015; Rupakheti et al., 2017; Rupakheti et al., 2020; Chen et al., 2020). During this time, a mixture of long-range transport and urban aerosols are observed, with the solution covering a very wide range of particle radii from 210nm up to 1030nm. This result is consistent with the site being impacted by air masses both of a local origin as well as those having been transported very far away from regions with a very high loading. These results are consistent with the rapid onset in terms of wind speeds, and consistent direction from south to north. Localized fires, coupled with urban areas in Eastern India and Bangladesh are all upwind from Lumbini during this time. It is important to note that while heavy rain removes aerosols during this time of the year, that it also does not rain 24 hours a day 7 days a week over the entire domain, and therefore a significant amount of aerosols will be transported and not fully removed, consistent with findings by (Lee and Wang, 2015; Takeishi and Wang, 2021). This finding is further supported by the observed higher concentration of sulfate to BC ratio, requiring a smaller amount of overall freshly-emitted BC particles, and a longer time in-situ, both of which allow the surviving particles more time and greater opportunity to grow. It is also consistent with the highly humid atmosphere providing plenty of water to facilitate the growth of a shell. As demonstrated in Fig.4(a) the BC core size is less than 220nm, while the remaining 50 to 170nm is found in the shell.

Second, during the latter part of the cold season (Fig4c-d)(specifically JF) there is observed to be a large amount of locally dominated aerosol sources (both [Urban] and local [BB] associated with economic activity, solid fuel used for cooking, etc.), which in turn fit well with the observed types (Dumka et al., 2008; Kumar et al., 2015). However, during the earlier part of the cold season, also termed the harvest season (ND), the majority of the sources are influenced by local biomass burning, with the results showing that there is a very absorbing fraction of biomass burning type solely observed during this period of time (Rupakheti et al., 2017). The impact of crops being harvested and some straw being left to dry can be observed in both of these seasons, and is ultimately burned to clear the land, or stored and later burned for heating later in the winter, completely consistent with the observed mixture of [Urban] and [BB] profile match in JF (Engling et al., 2011; Lee and Wang, 2017; Duc et al., 2021). This result is further consistent with works by Rupakheti (2019) who demonstrated that aerosols during the post-monsoon season are mostly associated with the biomass burning. Wan et al (2017) who demonstrated the presence of biomass burning organic aerosol tracers is highest in winter and late autumn compared to other seasons of the year, and Liu and Cohen (2022) who demonstrated that areas in Northeastern India at the same time have a significant underestimate in NO_x emissions associated with biomass burning.

Overall, these conditions match well with the extensive use of local brick kilns and rubbish burning occurring in the winter, transport from India and Bangladesh during the monsoon, and burning of straw and other agricultural waste during the harvest season. However, given that the cold period has 2 different phases, further work should be clear about which of the two phases is being indicated when analyzing the impacts of biomass burning in this part of the world.

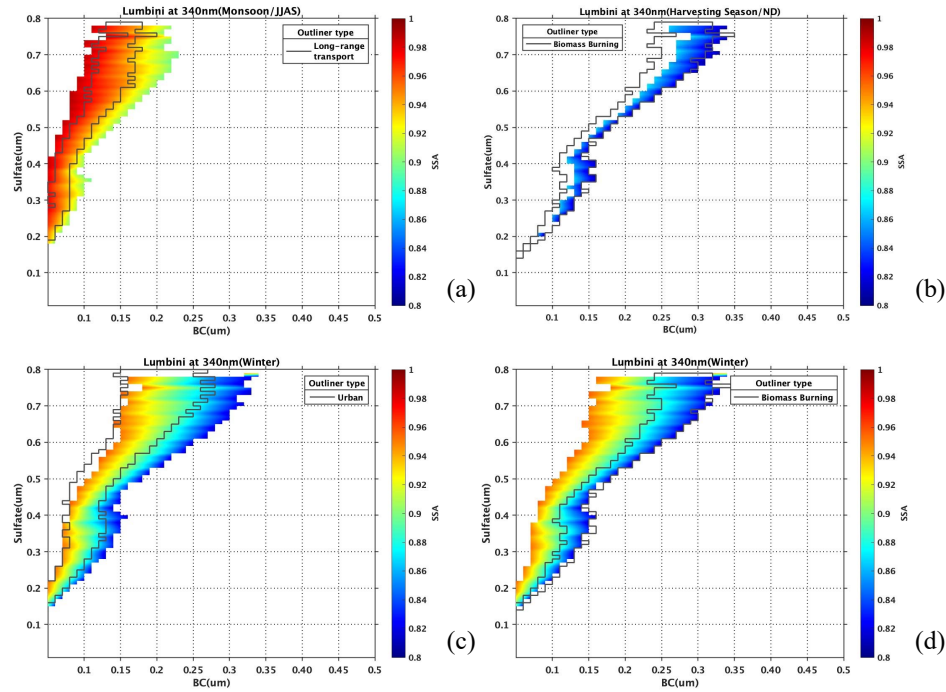


Figure 4. Constrained distribution of SSA from model result of Lumbini during winter(DJF), Harvest season and Monsoon with urban, Biomass Burning and Long-range transport profile.

Beijing is a highly dense urban area with a very large population, extensive private transport and without much heavy industry, indicating that an urban profile would generally be expected to dominate. Figure 5 demonstrates that over 9 months of the year, that there is a very strong urban signal in the data, when analyzed on a month-by-month basis. However, Beijing is also located at the foothills of mountainous regions to the West and North, and therefore can be subject to biomass burning and dust transport due to its unique geography.

A deeper investigation of the month-by-month properties show a clear intra-annual pattern which coherently and smoothly changes on a month-by-month basis, transitioning from one aerosol type to another, a very different finding from the hundreds of studies grouping Beijing aerosol properties on a seasonal basis (Liu et al., 2019; Wang et al., 2016; Han et al., 2014; Sun et al., 2015; Hu et al., 2017; Zhang et al., 2013; Zheng et al., 2005). November and December demonstrate a strong, narrow, and pure [BB] signal, while January has a strong [BB] signal, but one which is slightly less narrow. February continues this transition to smaller BC core sizes and higher SSA values, with the entire pattern from January shifted to the left by 0.03um, demonstrating a mix of both [BB] and [Urban] signals. March continues this leftward shift and thickening of the solution space, with nearly all of the results falling in the [Urban] range, while also comprehensively filling the breadth of the [Urban] signal. Both April and May reveal a pure [Urban] signal, without [BB] or [LRT] being observed. This is also observed in September, but in this case, it is transitioning in terms of larger BC cores in the opposite direction from [LRT] back towards [BB]. In June, the signal shifts further left, with a loss of some [Urban] signal and a growing signal of [LRT]. In July, the maximum [LRT] contribution is obtained, with the [LRT] contributing roughly 50% and the [Urban] contributing the remainder. August starts to shift in the opposite direction, with the BC core size growing, and the fraction of [LRT] decreasing to 40% and the fraction of [Urban] increasing. Finally, in October, the signal is thinner on average

and consists of mostly [Urban] signal and a small amount of [BB]. These results are indicative of the fact that while Beijing is an urban area, and the measurement site is located in the northern part of the urban core, that the sources of aerosols are highly variable from month-to-month, including biomass burning from near-by, and long-range-transported sources from beyond the JingJinJi area. It is hoped that further projects analyzing sources and profiles of absorbing aerosols in Beijing will look at a higher time frequency and move beyond the season-to-season approach currently adapted by the community. Some specific examples are that the biomass burning seems to cut between both the late Autumn and early Winter periods, and that the large sources of long-range transport seem to mostly exist in early summer, but over a shorter period than the summer as a whole. The fact that each month except for November and December is unique, indicates that there are far more complex forces at work.

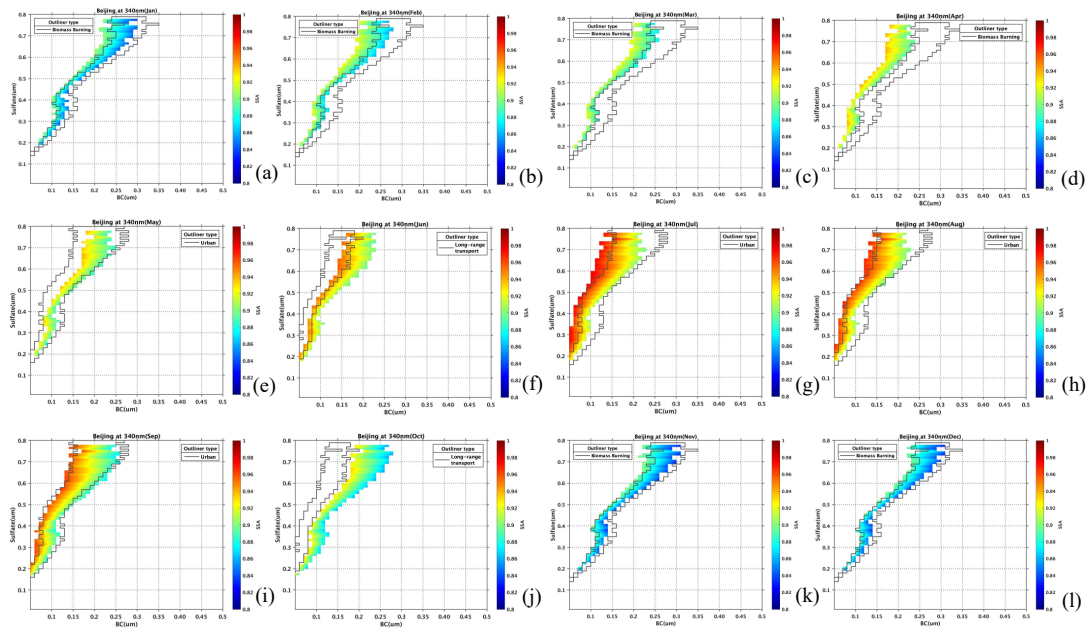


Figure 5. Constrained distribution of SSA month-by-month, demonstrating Urban, BB, LRT profiles all during different times of the year.

The resulting BC and shell size information during only the high periods of AOD in time (as displayed in Fig.1) are shown in Fig.6. This focus on when the AOD is highest offers the perspective of the properties of the BC and shell size when the loadings are most significant. There is a clear long range transport path observed at both Dongsha Island and Chen Kung University, and a very small probability of long-range transport identified in Hong Kong, all of which are consistent with the high biomass burning time periods found throughout Continental Southeast Asia (Lin et al., 2014; Chan et al., 2003; Lee et al., 2017; Sahu et al., 2014). These results are found to match very well with the standard [LRT] signal observed in Chiangmai, but shifted vertically upwards. This result is consistent with the source being biomass burning from Chiangmai and other surrounding areas (Huang et al., 2013; Fu et al., 2012; Chuang et al., 2015), which later transports to Dongsha and Chen Kung. While in-route, there is continued in-situ growth due to SO_2 , NO_2 , and VOCs from urban areas, and DMS from the South China Sea. The high time first appears in Hong kong from February 12 through April 13, with only 2% of the total solution being [LRT], which given the local magnitude of emissions during the dry season

in the GBA area (Chen et al., 2020; Duc et al., 2021), may still offer a somewhat significant impact. The contribution in Dongsha is observed from February 19 to April 22 with a far larger percentage of [LRT] at 42%, due to the fact that Dongsha does not have local sources (except for shipping), but it is still close enough to the GBA that it may contain some not-so-aged transported aerosols. Finally, the contribution in Chen Kung is observed from March 3 to May 8 and has 62% of [LRT], indicating that long range transport dominates the total loading there, consistent with the fact that there are very few local sources.

Under the assumption that Chiangmai is the source of the biomass burning, the particles will travel from west to east, first arriving in Hong Kong, next arriving at Dongsha, and finally at Chen Kung, assuming that the eastern wind in the lower free troposphere is relatively steady (Wang et al., 2013; Liu et al., 1999). Various one-off experiments and model studies have been conducted to study the impacts of the annually varying biomass burning events occurring in Continental Southeast Asia on surrounding areas. While there are many such studies which have demonstrated the plume in the near-downwind areas including Vietnam, Guangxi and Hainan (Huang et al., 2020; Ding et al., 2004; Li et al., 2007; Ding et al., 2021; Ding et al., 2013), there are very few which have been able to successfully transport as far as we have observed here (Cohen, 2014; Wang et al., 2021). Furthermore, while modeling studies of individual single fire events transporting in such a manner have been found (Aouizerats et al., 2015; Yen et al., 2013; Cohen et al., 2017; Wang et al., 2021), there has been no clear observational study, using data over multiple years (12 at Chen Kung, 10 at Hong Kong, and 6 at Dongsha), in which such a finding has been made. Furthermore, these findings are not limited in time to a single season or a single transport event, and provide a comprehensive, fully measurement based analysis.

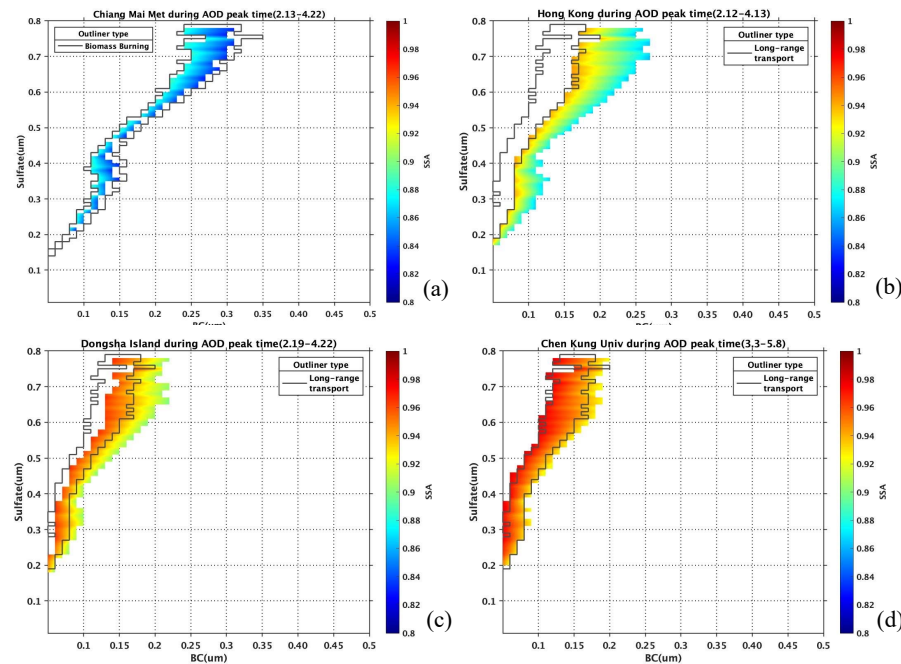


Figure 6. Constrained distribution of SSA from model result of sites that located along possible transport path.

4 Applications of Results

4.1 In-situ Lifetime of Sulfur Dioxide

The computed e-folding time is generally long, consistent with the fact that the air parcels have spent a significant amount of time being transported through the free troposphere, as demonstrated in Table 1. These e-folding times are approximated around the idea of DMS and/or anthropogenic SO₂ decaying into SO₄⁻ which then condenses onto the shell. The results are not strongly influenced by the quick removal of SO₂ and conversion into H₂SO₄ that occurs in the boundary layer near emissions sources under very high OH, wet and warm conditions found in heavily polluted conditions in this part of the world. Instead, the aging is mainly controlled by gas phase oxidation process happening in cooler and lower pressure air, which is also more likely to be exposed to UV radiation since it is found above the cloud layer. The results therefore should be consistent with relatively higher amounts of OH (closer to urban than background), due in part to the extra amount of UV radiation available above the cloud layer. Additionally, the results should show slower growth due to the lower pressure and cooler conditions. Based on equation (3) the maximum and minimum lifetime of SO₂ in two different chemical background conditions (the background case results are given in the columns of 4 and 5; the urban case results are given in the columns of 6 and 7) are calculated in this manner and listed in Table 1 and the possible uncertainty have been evaluated.

The results are consistent with the lifetime of SO₂ being longer as compared to the lifetime in the boundary layer where the temperature and recycling of OH are both high. In the free troposphere, although the OH may be high due to stronger UV, the lack of water vapor and VOCs lead to OH regeneration being very slow. Since the lifetime in urban areas is faster, it would make sense that by efficiently reducing the SO₂ emitted from urban areas, the longer timescale the aging of BC and sulfate is reduced (Cohen et al., 2011) for the BC emitted from the biomass burning and subsequently transported downwind over urban areas. Overall, while these lifetimes are long, they are not so long as to be ruled out based on the amount of time it takes to transport from the source region to the measured destination.

Testing the values of the e-folding time under background OH conditions yield a maximum lifetime with a value ranging from 21 to 32 days and a minimum ranging from 16 to 20 days. This increase in the amount of time, in particular for the minimum number of days from 10 to 16 leads to a set of solutions which are likely not possible. The amount of time required to grow would be more than the amount of time that it would take to be transported. Overall, the breadth of the solution range for lifetime is different between the minimum and maximum cases, consistent with the fact that the decay of atmospheric SO₂ into sulfate is slower than expected, but still reasonable in the case of a higher OH concentration, with the difference from 0 days to 9 days. The results require that a significant amount of the total BC is observed above the boundary layer.

There are other possible methods and assumptions which could be made to decrease the aging lifetime, which have not been considered herein due to the lack of data. First, higher levels of SO₂ that provided by CAMS, possibly due to increased vertical transport of SO₂ from urban areas along the path of transport, or from heightened DMS emissions or from faster chemical processing of DMS could lead to higher M(env.) in equation 3. Second, this work didn't consider the NO₂+OH reaction to calculate HNO₃, which is optically the same as H₂SO₄ and has the same effect on the result, which will make the in-situ lifetime shorter. Third, this work didn't consider reactions which may occur on the liquid water existing on the coated BC surface (e.g. potentially

sulfate aerosol production via aqueous phase oxidation), which would also make the in-situ lifetime shorter. If all these processes are taken into consideration, it would make the in-situ lifetime shorter, but not so short that the background OH cases would then be too short to match with the transport lifetimes. In general, the difference between the max lifetime of background OH and the min lifetime of urban OH allow the real world observed transport lifetime to be matched, based on the assumption of whether the oxidation and growth is purely based on the gas-phase oxidation and equation 1, or whether these other processes are also important.

Site name	Longitude	Latitude	Max(BG,days)	Min(BG,days)	Max(UR,days)	Min(UR,days)
ARM_Darwin	130.891	-12.425	27	25	17	15
ARM_Nainital	79.458	29.359	23	19	15	12
Anmyon	126.33	36.539	25	19	16	13
Bac_Giang	106.225	21.291	26	20	17	12
Bac_Lieu	105.73	9.28	22	16	14	10
Baengnyeong	124.63	37.966	28	22	17	14
Bandung	107.61	-6.888	26	21	16	13
Beijing	116.381	39.977	30	26	21	17
Beijing-CAMS	116.317	39.933	28	24	18	15
Chen-Kung Univ	120.217	23	24	18	15	12
Chiang_Mai Met Sta	98.972	18.771	—	—	—	—
Chiayi	120.496	23.496	26	20	16	13
Darwin	130.892	-12.424	—	—	—	—
Dhaka_University	90.398	23.728	29	24	19	15
Doi_Ang Khang	99.045	19.932	29	23	19	14
Dongsha_Island	116.729	20.699	23	17	14	11
EPA-NCU	121.185	24.968	26	21	17	13
Fukuoka	130.475	33.524	30	21	19	13
Gandhi_College	84.128	25.871	31	23	19	14
Gangneung_WNU	128.867	37.771	26	20	16	12
Gosan_SNU	126.162	33.292	21	18	13	12
Gual_Pahari	77.15	28.426	31	25	20	15
Gwangju_GIST	126.843	35.228	26	20	16	12
Hankuk_UFS	127.266	37.339	29	22	18	14
Hokkaido_University	141.341	43.075	26	21	16	13
Hong_Kong PolyU	114.18	22.303	31	22	19	14
Jabiru	132.893	-12.661	27	23	17	14
Jaipur	75.806	26.906	28	21	18	13
Jomsom	83.714	28.778	25	21	16	13
KORUS_Kyungpook_NU	128.606	35.89	26	21	16	13
KORUS_UNIST Ulsan	129.19	35.582	25	22	15	14
Kaashidhoo	73.466	4.965	26	19	16	12

Kanpur	80.232	26.513	31	24	19	15
Kathmandu-Bode	85.39	27.68	—	—	—	—
Kathmandu Univ	85.538	27.601	31	26	19	16
Lahore	74.325	31.542	31	22	19	14
Luang Namtha	101.416	20.931	30	23	16	14
Lumbini	83.28	27.49	—	—	—	—
Mukdahan	104.676	16.607	26	19	16	12
NCU Taiwan	121.192	24.967	26	21	17	13
NGHIA DO	105.8	21.048	27	20	17	13
Nainital	79.458	29.359	28	22	18	14
Nong Khai	102.717	17.877	29	21	18	13
Noto	137.137	37.334	26	19	16	12
Osaka	135.591	34.651	24	20	15	12
Palangkaraya	113.946	-2.228	—	—	—	—
Pimai	102.564	15.182	26	20	16	12
Pokhara	83.971	28.151	27	26	17	16
Pune	73.805	18.537	31	27	20	17
Pusan NU	129.083	35.235	28	21	17	13
SACOL	104.137	35.946	32	26	20	16
Seoul SNU	126.951	37.458	28	21	18	13
Shirahama	135.357	33.693	24	18	15	11
Silpakorn Univ	100.041	13.819	29	21	18	13
Singapore	103.78	1.298	23	18	15	11
Son La	103.905	21.332	30	20	19	13
Songkhla Met Sta	100.605	7.184	24	17	15	11
Taihu	120.215	31.421	—	—	—	—
Taipei CWB	121.5	25.03	26	21	17	13
Taiping	114.362	10.376	—	—	—	—
USM Penang	100.302	5.358	23	17	14	11
Ubon Ratchathani	104.871	15.246	25	18	16	11
Ussuriysk	132.163	43.7	30	23	19	15
Vientiane	102.57	17.992	26	20	17	12
XiangHe	116.962	39.754	30	23	19	14
Xinglong	117.578	40.396	30	22	19	14
Yonsei University	126.935	37.564	24	22	15	14
Yulin	109.717	38.283	30	29	19	18

4.2 Sensitivity of Solutions to Different AERONET Data Assumptions

The AERONET measurements contain some data which is of lower or questionable quality as compared to the bulk of the data. This is frequently due to either not enough absorption to successfully measure the SSA (i.e., when the $AOD < 0.4$), non-successfully cleared thin clouds, or other such phenomenon. To account for this uncertainty, this work has previously focused solely on the middle 80% of the total SSA measurements for each given dataset at each wavelength and time period studied. By discarding the top and bottom 10%, the reliability of the remaining measurements is much higher, and therefore the model results are more believable. The fact that differences on a monthly-basis are already clearly observed is proof that this is a reasonable approach.

However, selecting the central 80% is also arbitrary. So, a further sensitivity study is done to consider the effects of choosing the central 90% and 70% of data respectively, and quantify the impact of this cutoff on the computed BC and SO_4 size distributions. The results are given in Fig.7. A respective use of 70% and 90% of the data lead to a slightly biased change, although not leading a significant overall difference. In the case of 70% of data, the SSA distribution reduces by 32%, 27% and 30% respectively in the urban, LRT and BB cases. As observed, there is a slight bias with the decrease in urban being a further separation from LRT, and the decrease in BB being a move towards urban. In the case of 90% of data, the SSA distribution increases by 23%, 25.7% and 50% respectively in the urban, LRT and BB cases. As observed, there is a slight bias in the LRT case, in which case it extends further from the urban and in the BB case, in which case it also extends further from the urban. However, in all cases, the biases do not significantly alter the shape, and if anything, make it easier to distinguish the different underlying factors from each other.

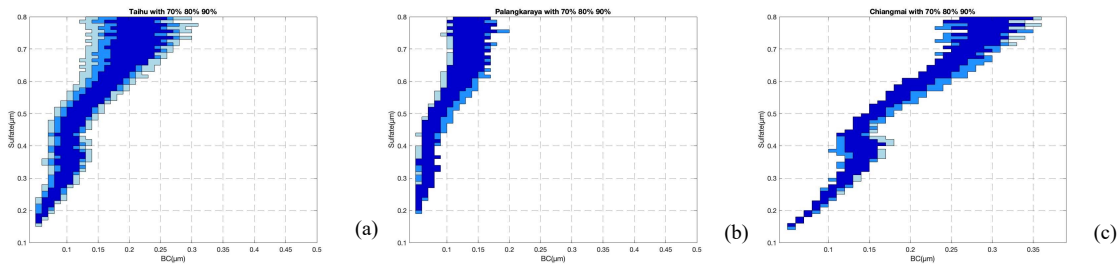


Figure.7 SSA distribution of three profiles with different amount of AERONET measurement data used to constrain the MIE inversion. When the central 70% of the measurements are used, the results are given in dark blue, when the standard 80% of the measurements are used the results are given in medium blue, and when the central 90% of the measurements are used the results are given in light blue. The plot on the left is for Urban, the plot in the center is for LRT, and the plot on the right is for BB.

4.3 Comparison with Independent Measurements

The MIE model inverted core and shell sizes in Beijing overlap at the same time with measurements taken by SP2 in Beijing from 2016.11.10 to 2017.01.12, as shown in Fig.8a. The inverted ratio of D_p/D_c from the MIE model approach and as observed from SP2 are shown in Fig.8b. This comparison assumes that the refractive index of core and shell are the same between the surface SP2 measurements and the column inverted Mie outputs. In general, the two methods

compare well with each other, with nearly all of the SP2 measurements (the 99.5% larger than 1.2) falling within the range of the MIE model inverted results. Next, the bulk of the SP2 data occurs with a ratio of 1.4-1.6, which also happens to be the mode of the range of outputs of the MIE model inversion. The only discrepancy is that the MIE model inversion has a range of values with D_p/D_c larger than 2, which is not observed in the SP2 data.

There are a few known possibilities for this discrepancy. First, is that the SP2 may fail to capture very small particles of BC, which may disaggregate or disintegrate when being hit by the laser associated with the SP2 detector. Similarly, some of the sulfate shell may be evaporated due to the heating of the BC, leading to a smaller than actual shell being measured. A third possible source of disagreement is that the SP2 only measures particles near the ground, while AERONET measures particles throughout the troposphere, and particles which are aloft as pointed out already, are more likely to have been in the air longer, and therefore be more significantly aged, leading to a larger value of D_p/D_c . It is possible that the ageing scale or transport range observed at the surface urban site may not pick up the fact that there are some biomass burning aerosols which are mixed into the total column but not necessarily at the surface, consistent with the fact that 28% of the observed solution space is BB while 72% is Urban.

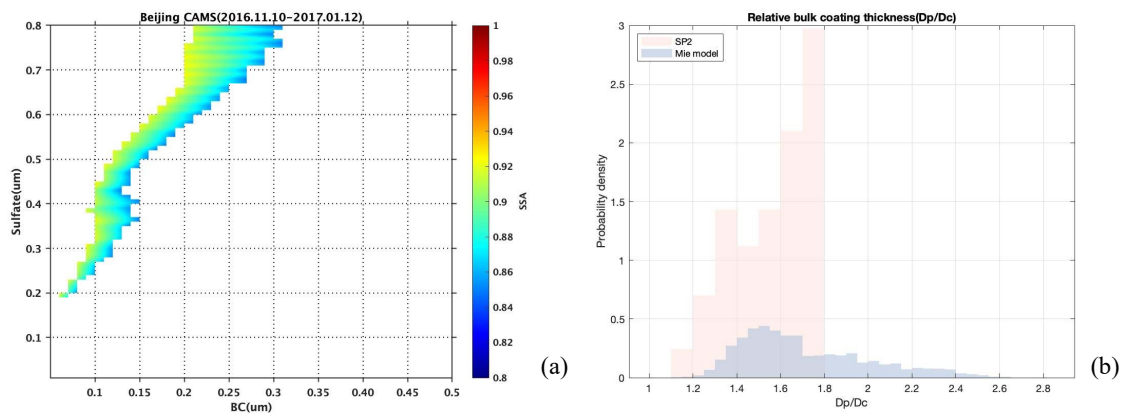


Figure.8 Size distribution during overlapping time period between AERONET and SP2 measurement in Beijing(left) plot. Probability density comparison of D_c and D_p/D_c of two datasets.

5 Conclusions

Applying a Mie-model with a core-shell approximation over multiple UV bands of the observed clear sky AERONET SSA has successfully allowed for a new inversion of BC and shell size and mixing state information to be obtained. These results are observed to group very well at typical geographical locations with well characterized BC properties. These results help deepen the understanding of first order chemical aging and particulate growth, and how the distribution of particle size and absorbing optical characteristics behave under these different classifications. By narrowly focusing on the size of BC, the size of the refractive shell, and the mixing state, results are found to be consistent with advanced modeling studies and independent measurements. The results conform very well over regions that are known to be heavily polluted, both in terms of urban, biomass burning, long-range transport and mixed sources.

Deeper analysis of the temporal and spatial variance, allows for disentanglement of overlapping source types, clearly identifying multiple characteristics as a function of space and time. Some of these characteristics are mixed in time with others, showing that multiple source types are possible to quantify in a probabilistic manner. These results are consistent with the atmospheric column quite reasonably having different sources located near the surface from those observed at the same time in the free troposphere. During specific times, individual sites are observed to rapidly transition from one dominant characteristic state to a different one, indicating significant different sources, atmospheric properties, and driving factors behind the aerosol distribution, even within a single geographic place. Frequently these changes occur at time scales with short temporal duration, although consistently across many different years, indicating new definitions of climate, season, and dynamical driving patterns may be more appropriate to use, rather than standard climatological definitions.

The work demonstrates that these types can be successfully divided into three distinct and unique size distributions for [Urban], [BB] and [LRT] conditions. These findings are applied across many different regions in terms of geography, economic development levels, and national boundaries, all of which have their own unique air pollution control rules, environmental goals, and air pollution emissions characterizations. Three excellent examples are observed in Hong Kong, Kathmandu and Beijing. These sites clearly exhibit local sources as expected, but also have periods of time where intense biomass burning, long range transport, and mixtures of these dominate. However, such shifts frequently occur only during specific months, although these patterns are found to hold consistently over many years. In each case, there is an unexpected scientific result: Hong Kong is found to have long-range transport during the Asian Monsoon (usually thought to be clean), associated with LRT transport from Southeast Asia; Nepal is found to have LRT during the monsoon rapidly switching to BB post Asian Monsoon, even when the region experiences the Monsoon rains; and Beijing is exhibited to have all different types slowing shifting from one to another and back, but only observed on a month-by-month basis, vastly different from the typical analyses which group Beijing season-by-season and do not identify these regular occurring and shifting patterns.

Based on how these best fit distributions change in time, in specific being shifted from a standard distribution to one with more shell growth, allows for an indirect way to quantify in-situ lifetime. This is clearly observed across many of the sites, allowing for a new observational constraint on the lifetime of aerosols and a quantitative way to constrain the mixing between local and non-local sources. The in-situ age of aerosols at different sites reveals a vast difference in in-situ lifetime, ranging from a week and a half up to a month, based on assumptions of the time of year and pure gas-phase aging and growth. Since these changes are observed throughout the total column, they are reflective of lifetime on a climatological scale, and provide support to many studies which have indicated that absorbing aerosols can transport significant distances, such as across the Pacific Ocean and to the three poles.

This work demonstrates that control strategies can be strategically developed for each individual location and adapted during different times of the year. Locations which have a single type would adopt a single strategy, while those with multiple times or time varying types would develop strategies which are multi-perspective and vary from time to time. This approach may allow for significant cost savings and/or more efficient control efforts. Locations with special events could opt for control of different types and amounts during different periods of the year. This flexibility will provide new opportunities to enhance the local atmospheric environment.

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Author contributions

The first draft of the review was prepared by **Xinying Wang**: Conceptualization, Writing - original draft, Data curation, Writing - review & editing, Investigation, Formal analysis, Software. **Jason Blake Cohen**: Conceptualization, Data curation, Writing - original draft, Writing - review & editing, Formal analysis, Funding Acquisition, Supervision. **Shuo Wang** software and visualization.

Data Availability Statement

The AERONET datasets used in this study are available at https://aeronet.gsfc.nasa.gov/new_web/data_description_AOD_V2.html. The CAMS global reanalysis product EAC4 is available at <https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-reanalysis-eac4-monthly?tab=overview>. The SP2 measurements in Beijing are available at <https://data.ceda.ac.uk/badc/aphh/data/beijing/man-sp2>. All of the data and underlying figures are available for download at <https://figshare.com/s/a8a98c6dbb7d7b50dabc>.

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