Analysis of Aerosol Optical Depth and Angstrom Exponent Number over Singapore 2007 – 2014

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ABSTRACT: South-East Asia is one of the fastest growing economic regions. As a consequence, emissions of anthropogenic aerosols, such as from industrial pollution and biomass mass burning, have increased. Singapore, being located at the crossroads of known biomass burning emission sources, is greatly affected by this annual phenomena. To understand and study such emission sources, the Centre for Remote Imaging Sensing and Processing (CRISP) in Singapore has deployed a permanent automatic Sun-tracking Photometer which is part of the Aerosol Robotic Network, AERONET*. This instrument performs daily direct sun measurements and all data products are cloud screened and validated by AERONET protocols. The principal products obtained are the aerosol optical depth (AOD), the derived Angstrom Exponent (AE), as well as fine mode and coarse mode part of the aerosol size distribution. Previous local and regional studies focused on the investigation of haze/smoke events, particularly between the months of August and October due to trans-boundary smoke transport, mainly caused by forest and agriculture burning in Sumatra and Borneo (Indonesia). In this study, we try to answer the question of whether there is an inter-annual pattern and whether there is a noticeable trend within the AOD and AE data sets over the last 7 years. AERONET level 1.0 data sets are further processed and exclusively used for this work. For locating and back-trajectory tracking of potential aerosol sources the HYSPLIT** model is used.

1. INTRODUCTION

Singapore located at 1.3˚N 103.8˚E in the tropical rainforest climate (Köppen: Af) (Schultz, 2002) is influenced by the rain-causing North-East monsoon from December till March and the South-West monsoon in the dry season from June until September. Singapore lies at the southern tip of the Malay Peninsula, situated in the center of South East Asia. Economic growth has led to increased emissions of anthropogenic aerosols and their precursors (Reid et al., 2013) from local sources and the surrounding countries. Between August and October the "slash and burn" practice for clearing plantations in Indonesia frequently causes haze events in Singapore due to trans-boundary smoke transport.

From a scientific, as well as from a Geo-political perspective, atmospheric aerosol pollution and its global distribution is gaining increased interest due to its importance to global climate change (e.g. IPCC 2013), and also due to its influence on air quality and its effects on human health (Behera et al., 2014; Streets et al., 2009). Aerosols are primary emitted as liquids or solids from natural and/or anthropogenic sources or can be secondarily formed through gas-to-particle conversion via reaction with oxidants such as O_3 and OH radicals (Seinfeld, 2006). In this study we focus on particles of the accumulation mode with radii < 0.5µm, such as from fossil fuels or biomass-burning. The two major chemical components of the latter source are black carbon (BC), which primarily absorbs, and organic carbon (OC), which primarily scatters solar radiation (IPCC 2013; Seinfeld, 2006), but also includes nitrogen oxides (NO_x) and methane (CH_4). Depending on the presence of hydrophilic sites or water soluble compounds in the organic aerosol particle, it can be wetted, water can condense on it, and the aerosol may act as Cloud Condensation Nuclei (CCN) (e.g. IPCC 2013, Hobbs et al., 2006, Seinfeld, 2006, Sun et al., 2006). CCN are particles that in the presence of supersaturated water vapor, as through adiabatic cooling by ascending air masses, activate to become cloud and fog droplets (Seinfeld, 2006). As a consequence aerosols can indirectly impact local weather conditions, monsoon cycles, and precipitation patterns (Lau et al., 2006).

Many recent studies focus on the regional aerosol environment, especially on smoke events caused by biomass burning. Such studies describe aerosol properties and transport patterns, particularly in the so called "fire season" (e.g. Atwood et al., 2013; Chew et al., 2013; Reid et al., 2013, Salinas et al., 2013). For Singapore, to our knowledge, there are no studies investigating the inter-annual pattern of aerosol optical properties and possible long-term evolution and transport trends. Inter-annual patterns are linked to recurring weather phenomena, and cyclic

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[*] http://aeronet.gsfc.nasa.gov (22.07.2014)

[**] https://ready.arl.noaa.gov/HYSPLIT_traj.php (22.07.2014)

anthropogenic and biological activity, like seasonal agricultural practices. Long term trends can be attributed to land surface changes due to climate change and increased industrial pollution due to economic growth (Zhang et al., 2010).

2. INSTRUMENTATION, DATA PROCESSING AND METHODOLOGY

Since 2006, the National University of Singapore host a multi-spectral CIMEL 318 Sun-photometer (Salinas, 2009) which is part of the AERONET (Holben, 1998) network. This instrument performs daily direct Sun measurements are used to obtain AOD and other related by-products such as AE and particle size distribution. For this study, we used level 1.0 raw photometer data of the aerosol optical depth (AOD or τ_α). The raw data is used, because it assures that potential smoke-contaminated data aren't accidentally removed by AERONETs automated cloud screening. Furthermore, we used the Angstrom Exponent Number (AE or α) and the so called fine mode fraction[***] (FMF or η) for the analysis (e.g. O'Neill et al., 2001). To extract these parameters we applied the spectral deconvolution algorithm (SDA) according to O'Neill (O'Neill et al., 2001). This procedure also serves as cloud screening through constrains developed by Salinas (Salinas et. al 2013). The AE contains information about the particle size distribution (PSD) (O'Neill et al., 2001). Values of $\alpha > 1$ are classified as aerosol regimes dominated by fine mode particles (radius < 0.5µm), $\alpha < 1$ by coarse mode (radius $r > 0.5$µm) particles (Salinas et al., 2009).
First we compared the monthly averaged SDA corrected level 1.0 AOD data with the AERONET level 2.0 (cloud screened and quality assured) to assess its usability. Month with less than 30 data points and less than 8 days of measurement within a month are classified as invalid sample and are excluded from the further analysis.

For the initial trend analysis the month August, September and October are not used to avoid a bias from the annual data set due to smoke of biomass burning. Later, for a further reduction of the influence of inter-seasonal variability and inter-seasonal trends, a boxcar averaging with a window width of 3 month is applied to the whole data set (Zhang et al., 2010; Weatherhead et al., 1998). This means that a monthly data point is replaced by a 3 month average of this month and the two neighboring month. Linear regression is applied by computing the chi-square error statistics and evaluated on its significance. The significance of a non-zero regression coefficient is tested by application of a t-test according to Bahrenberg/Giese/Nipper 1999. Possible changes in frequency and intensity of AOD readings over time is studied through the comparison of AOD histograms.

Furthermore, we investigated the development of AE and FMF over the same time period to reveal whether a change in the fine mode part of the PSD is detectable. High AOD values ($\tau_\alpha > 0.8$) together with high AE ($\alpha > 1$) values are classified as data, which potentially contains smoke from biomass burning. Those events detected outside the known fire season are analyzed on potential fire sources in the surrounding regions. For this the HYSPLIT2 model (Hybrid Single Particle Lagrangian Integrated Trajectory Model) is used to generate backward trajectories of air parcels, starting from the measurement site at heights 100m, 500m and 1500m above ground respectively. We used back-trajectories not older than 96 hours since the residence time for smoke aerosols is not more than 3 – 5 days (Atwood et al., 2013b; Hobbs, 2000). This is the average time a smoke particle stays in the atmosphere before it gets removed by dry deposition, which means transfer to the earth surface without the help of precipitation like sedimentation, or wet deposition by processes like rain or through cloud droplets (Hobbs, 2000; Seinfeld, 2006). These backward-trajectories are overlaid on top of MODIS fire hotspots graphs in order to identify potential pollution sources that can account for the observed increased AOD and AE readings over Singapore.

3. RESULT AND DISCUSSIONS

3.1 Comparison SDA corrected level 1.0 and AERONET level 2.0 data

In Figure 1 and for comparative purposes only we show a monthly averaged AERONET level 1.0 data and level 2.0 data for the period between 2006 and 2014. As stated in section 2.0, we strictly use AERONET level 1.0 data with SDA applied as a form of cloud screening (Salinas et. al 2013). We do this due to the fact that the stringent cloud screening protocols applied in AERONET level 2.0 data effectively removes elevated levels of AOD, such as those from heavy pollution events, by misidentifying them as possible clouds. The SDA algorithm effectively removes the coarse mode part (clouds) of the size distribution and preserves the fine mode part of the distribution. Inter-annual level 1.0 and 2.0 AOD trends are shown in Figure 1. Both describe a similar pattern with minimum averaged AOD values occurring during the December months which coincide with periods of highest annual rainfall (NEA, 2014). On the other hand, AOD maxima coincide with the so-called "fire season" which typically occurs between the months of August and October. Moreover, a comparative differential trend line shows a systematically positive bias of level 1.0 over level 2.0 by a total average of 20.70% approximately. Frequently occurring cirrus clouds, which are endemic in the tropics (Chew et al., 2011), might contribute to the observed bias by increasing AOD values while lowering FMF and the AE value (Salinas et al., 2013). However, widespread cirrus cloud intrusion is minimal as SDA has

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[***] The FMF is the fraction of the total AOD, which is attributable to fine mode particles, to the total AOD.

shown to be an effective cloud screening algorithm (Salinas et al., 2013). Peak discrepancies can be found in February 2008, October 2010 and September 2011 and 2012 (cf. Figure 1 bottom). Comparison with the monthly average AE and FMF values for theses month as shown in Table 1 reveal the dominance of fine mode particles. Much of the observed peak discrepancies do coincide with the region's dry periods in which seasonal biomass burning increases substantially. We interpret this as evidence that perfectly valid fine mode particle events have been removed from the level 2.0 data set.

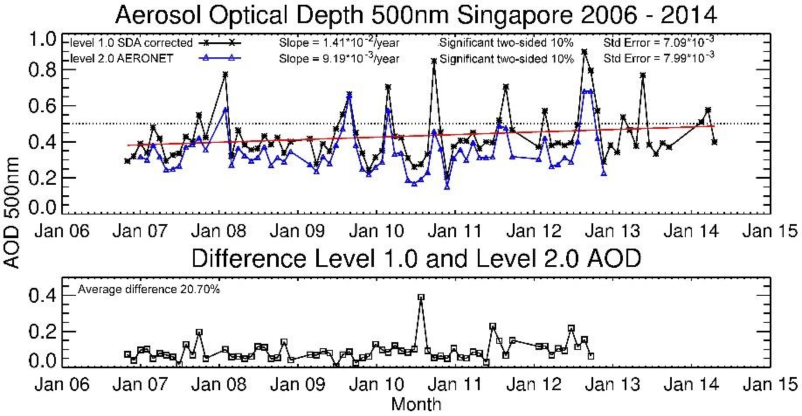

Figure 1 Comparison SDA corrected level 1.0 with AERONET level 2.0 data 2006 -2014 (top) and the difference of both data-sets (bottom)

Table 1 Monthly average AOD, AE and FMF for month with peak discrepancies between level 2.0 and level 1.0 data

Date	AOD	AE	FMF
FEB 2008	0.7742	0.9226	0.6197
OCT 2010	0.8477	1.3251	0.7351
SEP 2011	0.7052	1.2744	0.7720
SEP 2012	0.8992	1.2467	0.8385

3.2 Trend analysis

Figure 2 shows a seven year AOD trend without the fire season months of August, September and October. Linear regression reveals an AOD trend increase of the order of AOD $1.27*10^{-2}$/year (standard error se = $6.48*10^{-3}$), which is lower than the observed AOD trend that include the fire months (Figure 1). However, removing the seasonal fire months of August-to-October, which usually coincide with the regional South-West monsoon, has induced an indirect bias towards trans-boundary pollution generated by fires that occur during the North-East monsoon over the Indo-China region (and seen as elevated AOD peaks in Figure 2). To reduce possible impacts of these inter-seasonal effects on the trend analysis, box car averaging with a ± 3 month window is applied to the data-set including both monsoon seasons (Figure 4) according to Zhang et al. 2010. This results in a higher, significant increase of AOD $1.34*10^{-2}$/year (se = $4.53*10^{-3}$) compared to the trend without the summer fire-season. The trend can be a result of increased frequency of high AOD events or of increased intensity or both.

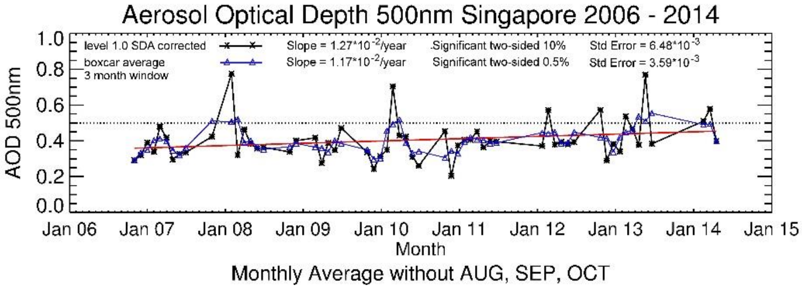

Figure 2 Seven year trend AOD and trend with box car averaging see text

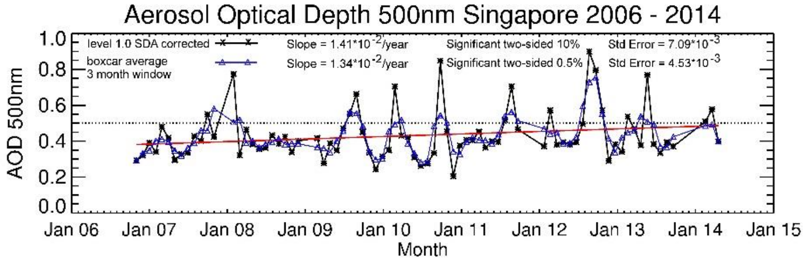

Figure 4 AOD 2006 – 2014 including both monsoon seasons

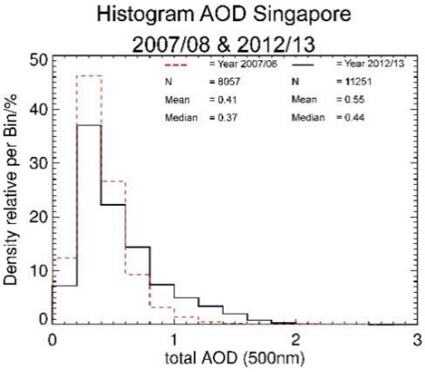

Figure 3 Comparison AOD histogram 2007-2008 and 2012-2013

The comparison of the normalized AOD histograms from the years 2007-2008 with those from 2012-2013 show a shift of the latter to high AOD range, indicating an increase in total aerosol loading (Figure 3). The mean AOD values show an increase for period 2007-2008 from mean $\acute{x} = 0.41$ (median M = 0.37) to $\acute{x} = 0.55$ (M = 0.44) in 2012-2013. For 2012-2013, a decrease of AOD in the range $\tau_{\alpha} < 0.6$ can be observed, whereas the frequency of occurrence of

AOD values higher than 0.6 is increasing. Higher frequency and intensity can be caused by anthropogenic activities as well as increased natural emissions. Analysis of the Angstrom Exponent number and the fine mode AOD, together with total AOD gives us some further insight about particle size and family type. Figure 5 shows the trend of the AE with both monsoon seasons with an increase of AE $1.46*10^{-2}$/year with a 90% degree of confidence. Accounting for inter-seasonal variability the box car averaged reveals a similar increase of the AE $1.45*10^{-2}$/year. Since high AE is associated with small particles, the increase of AOD is likely to be due to a higher aerosol loading of small particles. However, the fine mode AOD (FAOD) in Figure 6 shows a smaller positive slope of FAOD $8.86*10^{-3}$/year (se = $6.04*10^{-3}$). To put this into perspective, the FAOD increase would indicate a near constant or a slightly increase on the influx of fine mode particles into the environment. This influx is supported by a constant increase of the AE number and hence of smaller particle sizes.

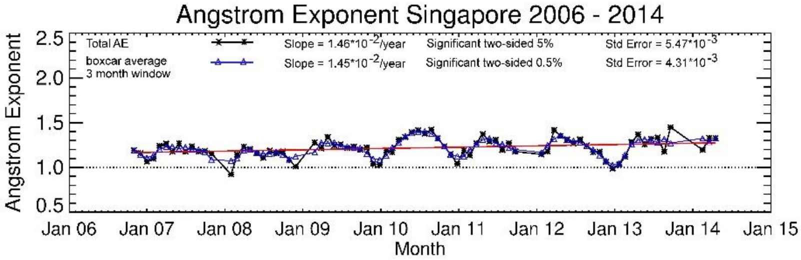

Figure 5 Angstrom Exponent Number 2006 - 2014

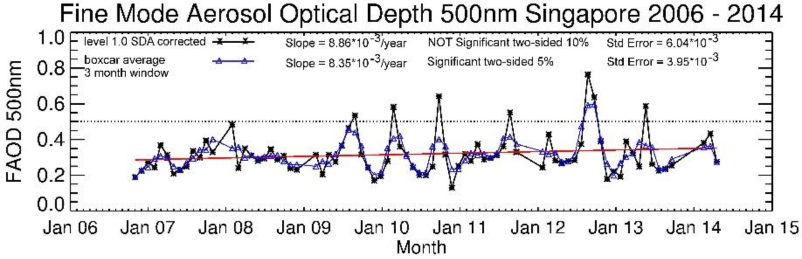

Figure 6 Fine Mode AOD 2006 – 2014

Table 2
Northern-hemispherical spring month with high monthly average AOD, AE and FMF

Date	AOD	AE	FMF
MAR 2007	0.4797	1.2436	0.7549
FEB 2008	0.7742	0.9226	0.6197
MAR 2010	0.7045	1.1719	0.7991
MAR 2012	0.5721	1.1787	0.7277
MAR 2013	0.5370	1.1192	0.7026
MAR 2014	0.5099	1.1957	0.7388

3.3 A Case study of off-season fire event from South-China Sea

Average high AOD and AE values together with high FMF can be seen in Figure 2 and Table 2, especially during the month of March. This suggest that Singapore, besides the typical South-West monsoon season fires, is affected by trans-boundary smoke transported during the transition period from North-East to South-West monsoon with prevailing weather conditions, causing increased abundance of fine mode particles from Indo-China and South-China Sea. To study this with more detail, we choose a case study that describes this transition period.

8 March 2010

March 2010 shows regular daily pattern of combined increase of AOD and AE values from morning until late afternoon (Figure 7). On 8th of March the values are rising from 1am till 9:30am UTC, passing the thresholds for smoke at 4:30am UTC (Figure 7 bottom). Figure 8 shows that the air parcels reaching Singapore in 100, 500 and 1500m above ground come from north-east. The overlay with fire hot-spots show fires on the north-east coast of Malaysia and the north coast of Sumatra. When the 1500m air parcel reaches the Malay Peninsula in around 700m height at 5pm UTC on the 7th of March, it passes over this location where a convective uplift happens before it reaches our receptor site in Singapore. The 100m and 500m air parcels do not coincide directly with the fire hot-spots. But the comparison with ensemble backward-trajectories for 250m and 1500m, which calculate 27 trajectories with slightly different starting conditions in all-possible offsets, suggest that aerosols/smoke can be transported to Singapore via convective up-lifting and/or vertical wind shear as suggested by Chew, 2013 and Atwood, 2013a (Figure 9).

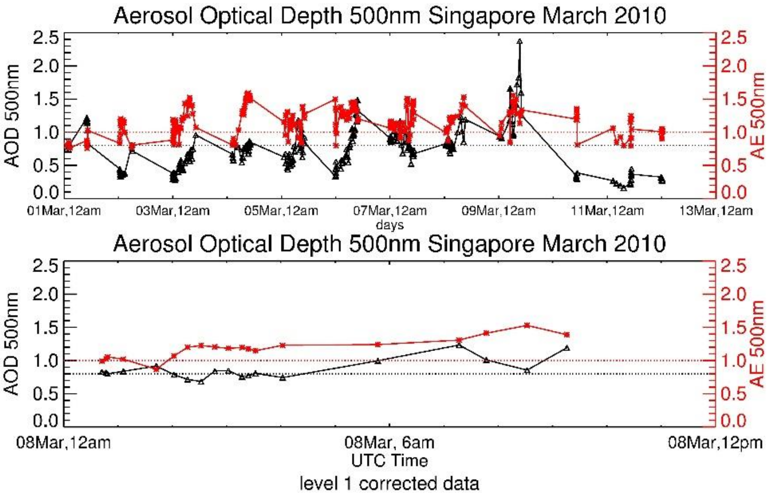

Figure 7 AOD and AE March 2010 (top) AOD and AE 08th March 2010 (bottom)

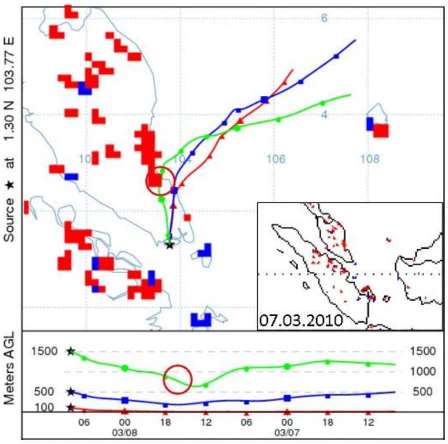

Figure 8 HYSPLIT backward-trajectories 48hrs, ending 0800 UTC 08 Mar 10. Overlay MODIS fire hotspots (red square: AQUA, blue square TERRA)

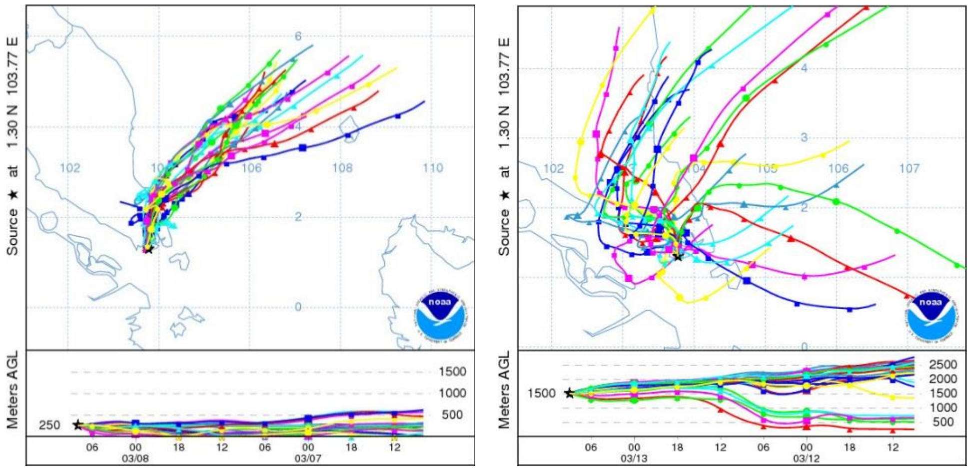

Figure 9 HYSPLIT ensemble backward-trajectories 48hrs, ending 0800 UTC 08 Mar 10. Start above ground level: 250m (left), 1500m (right).

SUMMARY AND FUTURE PROSPECTS

The inter-comparison between SDA corrected level 1.0 with AERONET level 2.0 data suggest that legitimate smoke data is effectively removed from level 2.0 AERONET product, thus affecting the study of individual smoke events as well as long time AOD trends.

A multi-year trend analyses show a positive slope increase of average monthly AOD by a rate of $1.34*10^{-2}$/year (se = $4.53*10^{-3}$). To reduce possible impacts of inter-seasonal effects on the trend analysis, box car averaging with a ± 3 month window was applied. Comparison of 2007-2008 and 2012-2013 histograms show a positive shift to higher AOD values with increasing frequency of occurrence. Applied linear regression analysis to the average monthly AE values show a positive slope, suggesting that a higher contribution of fine mode particles to the particle size distribution is the source for the increased AOD values.

An individual case study shows that during northern-hemispherical spring under dry conditions in the monsoon transition month of March, Singapore is likely to be affected by trans-boundary smoke transport. Sources are mostly found in Malaysia, but most back-trajectories indicate aerosol sources to be from the South-China Sea. Furthermore, a multi-layered aerosol vertical distribution system is present over Singapore, each layer might be from different sources and containing aerosols at different aging stages.

Detailed study of the vertical distribution, physical and chemical properties, and the interactions of anthropogenic aerosols of different origin with local aerosol sources need to be further investigated.

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