

Monsoonal variations in aerosol optical properties and estimation of aerosol optical depth using ground-based meteorological and air quality data in Peninsular Malaysia

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Abstract

Obtaining continuous AOD measurements is a difficult task due to cloud cover problem. An AOD-predicting model with the main motivation to overcome this problem is proposed. In this study, the optical properties of aerosols in Penang, Malaysia were analyzed for four monsoonal seasons (northeast monsoon, pre-monsoon, southwest monsoon, and post-monsoon) based on data from the AEROSOL ROBOTIC NETWORK (AERONET) from February 2012 to November 2013. The aerosol distribution patterns in Penang for each monsoonal period were quantitatively identified according to the scattering plots of the Angstrom exponent against the aerosol optical depth (AOD). A new empirical algorithm was proposed to predict the AOD data. Ground-based measurements (i.e., visibility and air pollutant index) were used in the model as predictor data to retrieve the missing AOD data from AERONET due to frequent cloud formation in the equatorial region. The model coefficients were determined through multiple regression analysis using selected data set from in situ data. The calibrated model coefficients have a coefficient of determination, R^2 , of 0.72. The predicted AOD of the model was generated based on these calibrated coefficients and compared against the measured data through standard statistical tests, yielding a R^2 of 0.68 as validation accuracy. The error in weighted mean absolute percentage error (wMAPE) was less than 0.40 % compared with the real data. The results revealed that the proposed model efficiently predicted the AOD data. Performance of our model was compared against selected LIDAR data to yield good correspondence. The predicted AOD can enhance measured short- and long-term AOD and provide supplementary information for climatological studies and monitoring aerosol variation.

1 Introduction

Air quality issues in Asia can be attributed to unavoidable climate change impacts and the negative impact of anthropogenic activities arising from rapid population growth, industrialization and urbanization (IPCC, 2007, 2013). Aerosol optical depth (AOD) derived from remote sensing has potential for assessing air quality. In general, spatial and temporal variations in AOD data are large since they depend on production sources, transport and removal processes that are modulated by local and synoptic meteorological conditions. Many

1 small-scale studies on the optical properties of aerosols have been conducted by Chew et al.
2 (2013); Mishra et al. (2013); Salinas et al. (2013) using sun and sky scanning radiometers of
3 AErosol ROBotic NETwork (AERONET) (Holben et al., 1998). These methods are limited
4 spatially relative to satellite imagery, and therefore are complementary for comprehensive
5 studies on atmospheric aerosols. Continuous measurements of AOD data are difficult because
6 the atmosphere is frequently cloudy. To better monitor and understand aerosol variation,
7 sufficient measurements and a practical observation paradigm of aerosols are necessary
8 (Hansen et al., 1997; Tripathi et al., 2005; Kaskaoutis et al., 2007; Kaskaoutis and Kambezidis,
9 2008; Russell et al., 2010).

10 Southeast Asia (SEA) stands out globally as it hosts one of the most complex meteorological
11 and environmental conditions, making remote sensing difficult both for AERONET and
12 satellites (Reid et al., 2013). Cloud cleared data leave gaps in our remote sensing data record
13 and conversely residual cloud contamination of remotely sensed data cause challenging tasks
14 to scientists studying aerosols (Chew et al., 2011; Campbell et al., 2013). Moreover,
15 anthropogenic biomass burning activities have increased dramatically in recent decades for
16 land preparation and forest clearance (Field et al., 2009). These fire activities result in trans-
17 boundary and long-range transport of aerosols that often affect air quality in both source and
18 surrounding regions (Hyer and Chew, 2010; Reid et al., 2013; Salinas et al., 2013; Lin et al.,
19 2014b). Those aerosols mix with locally generated aerosols (Engling et al., 2014). Therefore,
20 it is potentially valuable to develop a regional/local model to estimate and monitor AOD.

21 Development of an empirical model to produce reliable AOD estimates for aerosol monitoring
22 at local scales is novel and necessary for SEA, with potential global applications (Chen et al.,
23 2013; Fan et al., 2013). Several researchers have used models as alternative tools to predict
24 AOD values by using various ground based meteorology measurements (Wang et al., 2009;
25 Qin et al., 2010; Lin et al., 2014a). However, this approach has not yet been applied over the
26 Malay Peninsula region of SEA.

27 Previous studies indicate that AOD is proportional to air quality parameters such as particulate
28 matter (PM) with diameters less than 10 or 2.5 μm (PM_{10} or $\text{PM}_{2.5}$) (Wang and Christopher,
29 2003; Cordero et al., 2012; Mielonen et al., 2012; Mogo et al., 2012; Müller et al., 2012) but
30 inversely proportional to visibility (Vis) (Horvath, 1995; Li and Lu, 1997; Peppler et al., 2000;
31 Bäumer et al., 2008; Singh and Dey, 2012) assuming most of the aerosol is at the surface.
32 However, there are studies stating that AOD is not always highly correlated to surface or
33 horizontal measurements especially with the occurrence of an elevated layer of AOD from
34 transported dust or biomass burning (Mahowald et al., 2007; Barladeanu et al., 2012; Chen et
35 al., 2013; Toth et al., 2014).

36 In this paper, our goal is to build on previous experience to develop an AOD prediction model
37 based on three types of measured data, namely (i) RH, (ii) Vis and (iii) air pollution index
38 (API). These parameters are measured routinely at many ground-based stations. The AOD
39 prediction model based on these routine measurements is necessary to establish a long-term
40 database for i) climatological studies, ii) providing continuous atmospheric columnar AOD
41 data, and iii) monitoring aerosol variation such as diurnal cycles of AOD. Meanwhile, it is
42 important to understand the source of and dominant type of aerosol in this study. There is an
43 absence of understanding these factors on a local scale.

44 AOD measurements were obtained through the AERONET site located in Universiti Sains

1 Malaysia (USM) with geo-coordinates 5.36° N and 100.30° E. All AERONET direct sun data
2 used were Level 2 quality assured (Smirnov et al., 2000). The Vis and API data were taken
3 from the meteorological stations at the Penang International Airport and USM. All data were
4 taken between 2012 and 2013. The aerosol characteristics in Penang are comprehensively
5 analyzed for variation based on changes in seasonal monsoons. A near real-time AOD model
6 is established based on multiple regression analysis of Vis and API. The accuracy and
7 efficiency of the model are evaluated to assess air quality at Penang.

8 **2 Methodology and statistical model**

9 The present work was based on previous studies of Tan et al. (2014a, b). They predicted AOD
10 using multiple regression analysis based on meteorological and air quality data. The AOD
11 prediction model has been validated and successfully proven for the southwest monsoon
12 period (June–September, 2012) in Penang Island, Malaysia. However, the following issues
13 require reconciliation: (i) under- and overprediction of AOD were not **assessed** because of the
14 lack of available LIDAR data to monitor the variations in the vertical profile of the aerosol
15 distribution, (ii) the algorithm was insufficiently robust because only a four month dataset **was**
16 considered; and (iii) seasonal changes other than southwest monsoon were not included in
17 their study. The present study uses a two-year dataset (2012, 2013) at Penang to validate the
18 algorithms proposed by Tan et al. (2014a, b).

19 Penang is an island located in the northwestern region of Peninsular Malaysia and lies within
20 latitudes 5.20° N to 5.50° N and longitudes 100.15° E to 100.43° E (Fig. 4). The weather is
21 warm and humid year-round. However, two main monsoon seasons exist, northeast and
22 southwest monsoons. Considering previous analyses of aerosol and air quality (Awang et al.,
23 2000; Krishna Moorthy et al., 2007; Suresh Babu et al., 2007; Kumar and Devara, 2012;
24 Chew et al., 2013; Xian et al., 2013), the monsoon periods in this study were classified as
25 follows: (i) northeast monsoon (December–March), (ii) transition period of northeast to
26 southwest monsoon or pre-monsoon (April–May), (iii) southwest monsoon (June–September),
27 and (iv) transition period of southwest to northeast monsoon or post-monsoon (October–
28 November).

29 The AOD and Angstrom exponent are analyzed to identify the aerosol and broadly
30 characterize properties in Penang during each period. Precipitable water (PW) was used to
31 indicate the amount of the total water content in the atmosphere. The seasonal variations in
32 AOD, Angstrom exponent, and PW based on frequency distribution patterns are identified.
33 The aerosol types are seasonally discriminated from scatter plots of Angstrom exponent
34 against the AOD. Threshold values in the scatter plot for aerosol classification have been
35 previously reported by Smirnov (2002b, 2003), Pace et al. (2006), Kaskaotis (2007), Toledano
36 et al. (2007), Salinas et al. (2009), and Jalal et al. (2012). The data selection criteria proposed
37 by Tan et al. (2014a) are used in this study. Seasonal back-trajectory frequency plots from the
38 Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT_4) model **are** used to
39 identify the frequency occurrence of origin sources for aerosol and transported pathways
40 (Draxler and Hess, 1998; Wai et al., 2014).

41 AOD, API, and Vis data were selected according to the procedure of Tan et al. (2014a) to
42 generate predicted AOD data. AOD is computed from the solar transmission measured at 340,
43 380, 440, 500, 675, 1020, and 1640 nm using the automatic tracking sun and sky scanning
44 radiometers (Holben et al., 1998). These AOD data can be obtained from the AERONET web

1 page (<http://aeronet.gsfc.nasa.gov>). AERONET data has three different levels. Level 1.0 is
2 cloud-unscreened data, and Level 1.5 is cloud-screened data. Only Level 2.0 was employed in
3 this study because they are cloud screened and quality assured (Smirnov et al. 2000). Vis data
4 were retrieved online from Weather Underground (<http://www.wunderground.com>) or from
5 NOAA satellite (<http://www7.ncdc.noaa.gov/CDO/cdo>). Hourly data free from rainfall,
6 thunderstorms, or fog during the calculations were utilized to predict AOD. Air quality in
7 Malaysia is reported in terms of API, which can be obtained from the Department of
8 Environment in Malaysia (<http://apims.doe.gov.my/apims/>). API is calculated from carbon
9 monoxide, ozone, nitrogen dioxide, sulfur dioxide and PM₁₀. Malaysia is mainly polluted by
10 PM₁₀ (DOE, 2010). Therefore only API that are predominantly due to PM₁₀ are used in this
11 study. API is computed using the technique developed by US-EPA. The Malaysian
12 Department of Environment provides a standardized procedure on how to calculate API
13 values (DOE, 1997). The conversion between API and PM₁₀ has been shown in the guideline
14 provided in DOE (1997).

15 A total of 790 data points from 2012 to 2013 were used. Initially, the datasets were separated
16 into five sets as follows: (i) northeast monsoon, (ii) pre-monsoon, (iii) southwest monsoon, (iv)
17 post-monsoon and (v) annual dataset (overall). The number of data points for northeast
18 monsoon, pre-monsoon, southwest monsoon and post-monsoon were 257, 132, 235, and 166
19 respectively. In a particular seasonal monsoon period there are n data, {D₁, D₂, D₃, D₄,
20 D₅, ...D_n}, which are arranged sequentially in time. The data for each seasonal monsoon were
21 further divided into two subsets, in the form of {D₁, D₃, D₅, ...} and {D₂, D₄, D₆, ...}. The first
22 data subset was used to calibrate (Eq. 1) for AOD at 500 nm, given below:

$$\text{AOD} = a_0 + a_1(\text{RH}) + a_2(\text{RH})^2 + a_3(\text{RH})^3 + a_4(\text{Vis}) + a_5(\text{Vis})^2 + a_6(\text{Vis})^3 + a_7(\text{API}) + a_8(\text{API})^2 + a_9(\text{API})^3, (1)$$

23 where RH is the surface relative humidity (Tan et al., 2014a).

24 The root mean square error (RMSE), coefficient of determination (R^2), and weighted mean
25 absolute percentage error (wMAPE) between the measured and predicted AOD for each
26 seasonal model were calculated at 95 % confidence level. The wMAPE parameter was used to
27 quantify the systematic differences between the concentration levels. This parameter is given
28 as follows: $\text{wMAPE} = \left(\sum |((\text{AOD}_{p,i} - \text{AOD}_{m,i}) / \text{AOD}_{m,i})| * \text{AOD}_{m,i} / \sum \text{AOD}_{m,i} \right) * 100$, where
29 the subscript p refers to predicted, m to measured and i to individual measurements. The
30 ability of the proposed model to produce reliable AOD estimates for temporal air quality
31 monitoring can be quantitatively justified or falsified based on the value of the resultant
32 wMAPE.

33 Aerosols can be hydrophilic or hydrophobic, and these properties can have a non-trivial
34 impact on the magnitude of the retrieval AOD (Tang, 1996; Song et al., 2007; de Meij et al.,
35 2012; Singh and Dey, 2012; Ramachandran and Srivastava, 2013; Wang et al., 2013; van
36 Beelen et al., 2014). However, to discriminate between hydrophilic and hydrophobic aerosols
37 requires additional resources beyond the reach of the present study. Most fine mode aerosols
38 such as sulfates (that likely dominate urban industrial aerosol composition) are hydrophilic
39 such that one would expect RH to exert a significant influence on the measured AOD. Given
40 that Penang is dominated by urban industrial aerosols, one would expect RH to be an
41 important variable in the model. However, our pre-analysis showed that RH does not
42 contribute significantly to AOD prediction. We suggest that the RH, which is very high year
43 around in Penang, exerts less influence on AOD than we would see in drier climates. If RH
44 were considered as a predictor, its related factors (e.g., aerosol stratification (dust or smoke

aloft), convection, and hysteresis in particles) should otherwise be taken into account. The contribution of RH to the aerosol properties was integrated in the aerosol model of Srivastava et al., (2012), because the net effect of RH on aerosol and related factors were difficult to quantify. In similar spirit, the RH contribution is disregarded in the present model, yielding Eq. (2), given as follows:

$$\text{AOD} = a_0 + a_1(\text{Vis}) + a_2(\text{Vis})^2 + a_3(\text{Vis})^3 + a_4(\text{API}) + a_5(\text{API})^2 + a_6(\text{API})^3 \quad . \quad (2)$$

RMSE, R^2 , wMAPE were calculated for Eq. (2) in each monsoon season. The data for subset 1 (i.e., {D₁, D₃, D₅, ...}) was used for calibration. The data for subset 2 ({D₂, D₄, D₆, ...}) was used for cross-validation.

Lee et al. (2012) exclude days when the deviation between the measured and predicted values was greater than RMSE, or when the estimated AOD slope was negative because of measurement errors and cloud-contaminated AOD. The potential outliers in our model were removed following the approaches of Lee et al. (2012). The aforementioned procedures were used to calibrate the AOD prediction model, Eq. (2), using the resulting data from subset 1 after the elimination of outliers. The resultant coefficients of the calibration were then applied to data for subset 2 for cross validation, in which the predicted AOD values were compared with the measured data from AERONET.

Equation (2) was then applied to retrieve the AOD when no AOD values (from AERONET) were available. The under- and overpredicted AOD were examined using a RAYMETRICS LIDAR system if the data were available. Our LIDAR system is co-located with the CIMEL sunphotometer at the rooftop of School of Physics, USM (longitude 100.30° E, latitude 5.36° N). The detailed description of this LIDAR system can be found in articles written by Tan et al. (2013, 2014c). The LIDAR signals were pre-analyzed based on the protocol mentioned in Tan et al. (2013, 2014c). We shall briefly illustrate the protocol here. First, background solar radiation in the LIDAR signal has to be deducted. Then the analog and photon signals in the LIDAR signal are combined to enhance the near and far field signals. The range corrected signal (RCS) is obtained by multiplying the combined signal with a range square. To increase the signal-to-noise ratio, every 10 data files (each file contains data taken for one minute) are averaged over to give a single 10-minute averaged data. Then, the spatial resolution is determined by averaging over 10 bins (each bin is separated at a distance of 7.5m) spatially to obtain a 75m resolution profile. The RCS is then normalized by calibrating it against theoretical molecular backscatter according to the USSA976 standard atmosphere.

Often, the raw data are contaminated by presence of clouds during data-taking. Such contamination has to be removed so that the data is clean for the purpose of abstracting the values of AOD. LIDAR scattering ratio (defined as LIDAR signal divided by molecular backscatter signal) (Wang and Sassen, 2001; Lo et al., 2006) is used as a means to remove the cloud contaminated data. The referred LIDAR signal here is RCS, whereas molecular backscatter signal is referred to the attenuated molecular backscattering. The backscatter coefficients of the aerosol from LIDAR data are then determined using the method of Fernald (1984).

To strengthen our AOD prediction model, the variability in the retrieved AOD is compared to AOD retrieved from the LIDAR signal. Our LIDAR uses a laser pulse of wavelength 355 nm,

1 whereas the AERONET data are taken at a different wavelength. A conversion is performed
 2 to obtain AOD data from AERONET at 355 nm as described in Eq. (3) using the Angstrom
 3 power law (Ångström, 1929). It is used for Angstrom exponent estimation (α) in terms of
 4 AOD (τ_a) measured at wavelength $\lambda_1 = 340$ nm and $\lambda_2 = 380$ nm. In principle, if AOD and
 5 Angstrom exponent at one wavelength are known, AOD at a different wavelength can be
 6 computed, within the range of validity of Eq. (3), as

$$7 \quad \alpha = -\left[\left(\ln \frac{\tau_{a_2}}{\tau_{a_1}}\right) \div \left(\ln \frac{\lambda_2}{\lambda_1}\right)\right] \quad . \quad (3)$$

8 Therefore, AOD at wavelength 355 nm can be calculated as

$$9 \quad \tau_{a_{355}} = \tau_{a_{340}} \times \left(\frac{\lambda_{355}}{\lambda_{340}}\right)^{-\alpha} \quad . \quad (4)$$

10 After the conversions, we repeat the procedure as described above to obtain a new set of
 11 coefficients at 355 nm for the AOD predicting model.

12 Next, an AOD value is obtained from the LIDAR signal. A LIDAR ratio (L) is a constant,
 13 defined as the ratio of aerosol extinction coefficient (α_a) and backscatter coefficient (β_a) [see
 14 Eq. (5)]. The value of L depends on the particle size distribution, shape and composition. R in
 15 Eq. (5) is the range or altitude. α_a can be obtained once β_a and L are known. The value of L
 16 has to be assumed for an elastic LIDAR system (Fernald, 1984; He et al., 2006; Lopes et al.,
 17 2012). Normally, L values can range from 20-40 sr for clean and polluted marine aerosol
 18 particles or dust, urban aerosols (40-60 sr), and biomass burning aerosols (60-80 sr) as
 19 suggested by Chew et al. (2013).

$$20 \quad L(R) = \frac{\alpha_a(R)}{\beta_a(R)} \quad (5)$$

21 The value of L to be adopted for calculating α_a depends on which dominant aerosol type is in
 22 the atmosphere. To arrive at a specific value for L is somewhat arbitrary. Different authors
 23 adopt different strategies to fix the value of L. In this study, the following strategy is adopted:
 24 the aerosol type is first identified by using a scatter plot of the Angstrom exponent against the
 25 AOD (from AERONET data). Once the dominant aerosol type is determined the
 26 corresponding L value is set to be the mean value of the range suggested by Chew et al. (2013)
 27 for that particular aerosol type. Specifically, for clean and polluted marine aerosol particles or
 28 dust, L=30 sr; for urban aerosols, L=50 sr; for biomass burning aerosols, L=70 sr.

29 AOD values (τ_a) can be obtained using Eq. (6), where R_{max} is the maximum height of the
 30 aerosol distribution, and R_0 is the height where the overlap function, $O(R) = 1$, in our system
 31 R_0 is around 200 m. Inaccurate assumption of L can lead to large errors in the retrieval of α_a
 32 and τ_a (He et al., 2006), especially under inhomogeneous atmospheric conditions. Therefore,
 33 10 % uncertainty of L and typical values of 7 % uncertainty for β_a are set to estimate
 34 potentially erroneous values of α_a at any given R in an atmospheric profile. Finally, all
 35 uncertainties in the profile are summed to obtain the uncertainty of the estimated columnar
 36 AOD.

$$37 \quad \tau_a = \int_{R_0}^{R_{max}} \alpha_a(R) dr \quad (6)$$

1 The LIDAR estimated AOD values obtained were then compared against those predicted by
2 our developed AOD prediction model.

3 **3 Results and discussion**

4 **3.1 Climatology of Penang, Malaysia**

5 The climatological results derived from AERONET
6 (http://aeronet.gsfc.nasa.gov/new_web/V2/climo_new/USM_Penang_500.html), based on the
7 work of Holben et al., (2001), for USM Penang are tabulated in Table 1. The monthly AOD
8 (referred to as AOD₅₀₀, second column) shows that the two lowest values are 0.18 and 0.19
9 during the inter-monsoon period (October–November and May). During the southwest
10 monsoon period (June–September), smoke emitted locally and large-scale open burning
11 activities in Sumatra, Indonesia is transported to Malaysia, yielding the highest AOD at
12 approximately 0.31–0.73. However, AOD is 0.21–0.24 during the northeast monsoon period
13 (December–February). Small aerosol particles contribute primarily to the air pollution in
14 Penang, as the average **Angstrom exponent for wavelength between 440 nm and 870 nm**
15 (referred to as Angstrom_{440–870}) is higher than 1.1. On the other hand, the precipitable water
16 values (referred to as PW) were greater than 4.1, which indicate that Penang has humid
17 atmospheres (Okulov et al., 2002).

18 **3.2 Seasonal variations of AOD, Angstrom exponent, and PW based on** 19 **frequency distribution patterns**

20 AERONET parameters are plotted (Fig. 1) to reveal the relative frequency distribution at
21 Penang for each seasonal monsoon. Frequency histograms of AOD₅₀₀ and Angstrom_{440–870}
22 (Fig. 1a–b, respectively) indicate changes in the optical properties of aerosols, whereas Fig. 1c
23 shows the amount of water content **in the atmospheric column** for each season. These
24 histograms here help distinguish aerosol types (Pace et al., 2006; Salinas et al., 2009; Smirnov
25 et al., 2002a, 2011). Our results show that the distributed AOD mainly ranges from 0.2 to 0.4,
26 contributing to approximately 71 % of the total occurrence (Fig. 1a). Fig. 1b shows that the
27 Angstrom exponent is typically between 1.3 and 1.7, translating to ~ 72 % of the total. About
28 67 % of the total occurrence of PW ranged from 4.5 cm to 5.0 cm (Fig. 1c).

29 The maximum AOD frequency was centered near 0.2 for all seasons. The clearest season was
30 the post-monsoon (Fig. 1a). Penang was most polluted in the southwest monsoon, most likely
31 due to active open burning activities in Sumatra. The AOD peak was approximately 1.4, with
32 three peaks distributed from AOD₅₀₀ = 0.1 to AOD₅₀₀ = 1.4 (Fig. 1a). The multiple peaks
33 imply the presence of various aerosol populations, because AOD histograms follow log-
34 normal distribution patterns (Salinas et al., 2009). By contrast, a single peak was observed for
35 the clearest season (post-monsoon).

36 The frequency distributions as a function of Angstrom exponent display a trend (Fig. 1b) in
37 which approximately 95% of the total occurrence **falls within** the range of 1 Å to 2 Å. This
38 result implies that the effect of coarse particles (e.g., dust) on the study site was minimal. This
39 statement is supported by Campbell et al. (2013), who showed that dust particles are
40 uncommon in Southeast Asia. However, **sometimes dust particle concentrations can be**
41 **enhanced above the boundary layer**. Two noticeable peaks were observed for the Angstrom
42 exponent during the northeast monsoon period (blue curve, Fig. 1b). These aerosols originated

1 from the northern part of Southeast Asia, particularly Indochina, transported by the monsoon
2 wind and mixed with locally emitted aerosols. Lin et al. (2013) analyzed aerosols in the
3 northern region of Southeast Asia. They found that biomass burning aerosols from Indochina
4 were transported in high- and low-level pathways to the west, and then later shifted to the
5 southwest by northeast monsoons.

6 Biomass burning aerosols were continuously transported to our study site, as the wind
7 circulation flows toward the southwest direction according to the monthly mean streamline
8 charts of Lin et al. (2013) from 1979 to 2010. During and before the southwest monsoon,
9 Angstrom exponents in Penang ranged between 1.4 and 1.8, indicating the likely presence of
10 biomass burning aerosols (Holben et al., 2001; Gerasopoulos et al., 2003; Toledano et al.,
11 2007). They are likely to originate from local and neighboring countries. Indonesia is known
12 to be very active in open burning during this season. Furthermore, southwest monsoonal
13 winds are likely to have transported these biomass burning aerosols to Penang.

14 The southwest monsoon period is the **driest season in Malaysia. PW frequency was**
15 **approximately 20 % lower than that of the northeast monsoon period with $PW < 4.0$** (Fig. 1c).
16 Marked variations in the PW frequency were observed during the northeast monsoon period.
17 Almost no data were obtained for $PW < 3.5$, except the northeast monsoon period with about
18 14 % less than this value. The most humid period took place in the post-monsoon, with PW
19 ranging from 5.0 to 5.5 (approximately 74 % of the total occurrence).

20 **3.3 Seasonal discrimination of aerosol types based on the relationship between AOD and** 21 **Angstrom exponent**

22 Aerosol clusters have been developed using relatively simple scatter plots of AOD and
23 Angstrom exponent. **Similar studies have performed this analysis** using AERONET data.
24 These datasets have been applied at different locations, such as the Persian Gulf (Smirnov
25 et al., 2002a); several oceanic regions (Smirnov et al., 2002b); Brazil, Italy, Nauru, and Saudi
26 Arabia (Kaskaoutis et al., 2007); Spain (Toledano et al., 2007); Singapore (Salinas et al.,
27 2009); Kuching (Jalal et al., 2012); and the Multi-filter Rotating Shadowband Radiometer in
28 Central Mediterranean (Pace et al., 2006). The scatter plot of $Angstrom_{440-870}$ against
29 AOD_{500} or AOD_{440} was used to characterize the aerosol type. The wavelength range of
30 $Angstrom_{440-870}$ was used because of its nearness to the typical size range of aerosol based on
31 spectral AOD (Eck et al., 1999). The relation between AOD values at 500 nm and Angstrom
32 440–870 is commonly used for aerosol classification in scatter plot **diagrams** (Kaskaoutis et
33 al., 2007). Optically, 500 nm is an effective visible wavelength suitable for aerosol study
34 (Stone, 2002).

35 Aerosols are classified into five types, including dust, maritime, continental/urban/industrial,
36 biomass burning, and mixed aerosols (Ichoku et al., 2004). Mixed aerosols in practice
37 represent an indistinguishable type that cannot be categorized into any of the previous types.
38 To effectively identify the aerosol distribution types in our study sites, the results are
39 compared using different threshold criteria (Table 2), as presented in Fig. 2.

40 The thresholds proposed by Pace et al. (2006) and Kaskaoutis et al. (2007) **failed** to
41 distinguish the maritime aerosol (MA) and dust aerosol (DA). Instead, they show that mixed-
42 type aerosols (MIXA) **are dominant** at Penang (50–72 %). Urban and industrial (UIA) and
43 biomass burning (BMA) aerosols are grouped into a single class (28–50 % of the total

1 occurrence). Meanwhile, the threshold suggested by Smirnov et al. (2002b, 2003) failed to
2 identify DA, UIA, and BMA, but efficiently identified MA. As a result, a large amount of
3 MIXA was obtained (> 80 % of the total occurrence). These results reveal the extent of
4 regional uncertainty. Indistinguishable aerosol types in the study sites are significant.

5 Salinas et al. (2009) suggested that the determination of DA and BMA does not correspond
6 entirely to the range of threshold used in our study, in which the amount of MIXA
7 (approximately 43 % of the total occurrence) was large. Jalal et al. (2012) efficiently
8 identified aerosol types using an alternative threshold criterion. Using their threshold, we find
9 a low amount of MIXA of approximately 21 %. However, the determination of DA was
10 unsatisfactory. The threshold criteria of Toledano et al. (2007) provided the least MIXA (<
11 5 %; Fig. 2). All thresholds consistently increased from June to September (Fig. 2c),
12 coinciding with the occurrence of haze. UIA was constantly and highly distributed over
13 Penang. Overall, the thresholds provided by Toledano et al. (2007) were selected for our study.

14 Based on the criteria suggested by Toledano et al. (2007), the UIA class was determined as
15 the highest frequency of occurrence in the overall study period (Fig. 3). This could be the
16 result of Penang being an urban area. The next highest was the MA class, likely due to its
17 geolocation (i.e., surrounded by the sea). BMA is also one of the major pollutants in Penang,
18 which was produced by active burning in local and neighboring countries. These results are
19 consistent with records from our Department of Meteorological, DOE (2010). The study site
20 was minimally affected by DA, which were less than 5 % in each seasonal monsoon. These
21 results are supported by Campbell et al. (2013) who suggest UIA, MA, and BMA are likely
22 the most common aerosol types in Southeast Asia and the maritime continent.

23 BMA, UIA, and MA obtained in our study during the southwest monsoon were about 45, 24,
24 and 19 %, respectively. During the northeast monsoon period, UIA (approximately 38 %) was
25 the major aerosol in Penang, followed by MA (30 %), BMA (20 %), dust (4 %), and
26 unidentified substances (8 %). However, MIXA reached 17 % in the pre-monsoon, which was
27 the highest among the seasonal monsoons. MA and UIA were 38 %; the MA level was
28 significant during the post-monsoon period (51 %), followed by UIA (40 %) and BMA (<
29 1 %). The aerosol distribution in Penang is highly seasonally dependent.

30 **3.4 Seasonal flow patterns of air parcels from the HYSPLIT_4 model for** 31 **identification of aerosol origins**

32 From seven-day seasonal plots of the back-trajectory frequency sourced from the HYSPLIT_4
33 model, flow patterns reaching the Penang site were determined (Fig. 4) for each monsoon
34 season, averaged between the ground surface up to 5000 m. Residence time analysis was
35 performed to generate the frequency plot and determine the time percentage of a specific air
36 parcel in a horizontal grid cell across the domain.

37 During the northeast monsoon period, air parcels flow southwestward from the northern part
38 of Southeast Asia (Fig. 4a), including Indochina, through the South China Sea to Penang.
39 Aerosols observed during the northeast monsoon period are also locally produced, whereas
40 those observed during the southwest monsoon period are predominantly from the Andaman
41 Sea, Malacca Strait, and Sumatra (site of open active burning).

42 Fig. 1b indicates differences in patterns (bimodal distribution pattern) of the seasonal relative

1 frequency of occurrence for $\text{Angstrom}_{440-870}$ during the northeast monsoon compared with
2 the other monsoon periods. These differences are likely attributable to the mixing of various
3 aerosol sources from the northern (e.g., Indochina, Philippines, Taiwan, and eastern China)
4 and southern (e.g., Malaysia and Indonesia) parts of Southeast Asia (refer Fig. 4a). Biomass
5 burning aerosols are likely different for northern and southern SEA because of different types
6 of burning processes (Wardoyo, 2007; Lopes et al., 2012; Bougiatioti et al., 2014; Kaskaoutis
7 et al., 2014). As a result, a bimodal pattern was observed only for the northeast monsoon
8 period (Fig. 1b).

9 Figure 1b reveals that the distribution patterns of Angstrom exponent between the post-
10 monsoon and northeast monsoon are similar. Figure 4a and d also indicate similarities
11 between the air flow patterns for these monsoon seasons. Hence, a clear correspondence was
12 observed between Fig. 1b with Fig. 4a and d. The similarity in the patterns of Angstrom
13 exponents for the post-monsoon and northeast monsoon may be attributed to the mixture of
14 aerosols from the northern and southern parts of Southeast Asia. Given the classification
15 results (Fig. 3), the occurrence frequency of MA was higher during the post-monsoon and
16 northeast monsoon compared with the southwest and pre-monsoon period. The large amount
17 of MA originates from the South China Sea and Andaman Sea.

18 For the pre-monsoon period, aerosols observed at Penang originate from the Malacca Strait,
19 Andaman Sea, the northern and some eastern areas of Sumatra, and the western part of
20 peninsular Malaysia, especially the local regions marked in yellow (Fig. 4b). During this
21 season, air flow patterns are similar to those during the southwest monsoon (Fig. 4c).
22 However, a small percentage of aerosols are transported from the northern part of Southeast
23 Asia to Penang during the pre-monsoon period. Indonesia is known to be very active in open
24 burning activities during the southwest monsoon. Therefore the BMA observed in Penang
25 during this season is mainly due to local and transboundary aerosol from Indonesia. This
26 phenomenon is reflected in the narrower and sharper curves on larger values of the Angstrom
27 exponent in Fig. 1b (detailed explanation in Sect. 3.5). A clear correlation is observed
28 between Fig. 1b and Fig. 4b and c during pre-monsoon and southwest monsoon.

29 The dominant aerosol types are UIA and MA (Fig. 3). The yellow portions in Fig. 4e indicate
30 that for Penang, the second largest city in Malaysia and one of the most industrially-
31 concentrated cities, UIA is a major aerosol type. MA contribution to the overall aerosol
32 distribution is likely influenced by proximity of the surrounding sea.

33 3.5 Examination of predicted AOD values

34 The optical properties of aerosol for each monsoonal season are obtained by analyzing the
35 relative frequency occurrence of AOD_{500} and $\text{Angstrom}_{440-870}$ as shown in Fig. 1a and 1b.
36 We hypothesize that the proposed AOD prediction model should exhibit different accuracies
37 seasonally because the sensitivity for AOD prediction depends on the distribution patterns of
38 the measured AOD; these values were used as inputs to derive the correlation parameters of
39 the model. The sensitivity of AOD prediction is low when the major occurrence frequency is
40 clustered around small AOD values. The insensitivity of the aerosol models to clear
41 atmospheric conditions (e.g., when AOD is low) was also previously observed (Zhong et al.,
42 2007).

43 Model performance for each monsoonal season was tested (Table 3). The pre-monsoon and

1 southwest periods exhibited R^2 of 0.65 (RMSE = 0.11) and 0.77 (RMSE = 0.17). However,
2 for the transition period between post-monsoon to northeast monsoon, R^2 values were smaller
3 than 0.45 and RMSE ranged from 0.06 to 0.11. The accuracy of AOD prediction is improved
4 for cases with higher aerosol concentrations. This result is in agreement with the hypothesis
5 mentioned in the previous paragraph. The analysis of twenty two months of data (the so called
6 “overall” model) is satisfactory, with $R^2 = 0.72$ and RMSE = 0.13. The low value of wMAPE
7 (< 1 %) indicates that the model yielded relatively accurate results for all seasons. Given the
8 criteria that a low wMAPE corresponds with a good prediction, the “overall” dataset yields
9 the least biased prediction. Therefore it is deemed that the "overall" model (which is obtained
10 by training the model using twenty two months of data) can be interpreted as an effective and
11 representative model which can predict AOD in every period.

12 High correlation was observed between the measured and predicted AOD for the pre-
13 monsoon and southwest monsoon, in which similar air flow patterns occurred (Fig. 4b and c).
14 On the other hand, the prediction accuracy of the AOD model in the post-monsoon and
15 northeast monsoon seasons was moderate. The air flow patterns in Fig. 4a and d, which are
16 associated with northeast and post-monsoons respectively, also show similarity. This
17 observation is consistent with Figure 1b which displays the relative frequencies of occurrence
18 of Angstrom_{440–870}. When scrutinizing the seasonal curves pair by pair, the post-monsoon
19 and northeast monsoon pair (purple and blue curves) appears to be broader and flatter whereas
20 the other two seasons (red and green curves) are sharper and narrower. To be more
21 quantitative, the slopes for the purple and blue pair begin to pick up at a relatively fast pace
22 (compared to the red-green pair) at around Angstrom exponent of 1.1, dropping at around
23 Angstrom exponent of 1.7 from peak values, maintaining a relatively flat profile between
24 these two limits. Whereas the slopes for the red and green pair begin to pick up at a relatively
25 gentle pace (compared to the purple-blue pair) at around Angstrom exponent of 1.1 (for red
26 curve) and 1.3 (for green curve), dropping at around Angstrom exponent of 1.6 (for red curve)
27 and 1.7 (for green curve) from peak values. The profile for the red-green pair is relatively
28 narrower and sharper between their pick-up and dropping limits. As a result, a clear
29 correlation between aerosol optical properties in Fig. 1b and seasonal wind flow patterns in
30 Fig. 4 is observed.

31 The broader and flatter curves in the post-monsoon and northeast monsoon indicate that
32 coarser aerosols are more frequently loaded in the atmosphere. This observation is proved in
33 Fig. 3, in which both monsoons are showing higher occurrence of MA and lesser BMA in
34 Penang. In fact, we realized that AOD predominantly falls on smaller values if MA is the
35 dominant aerosol type, because clean atmosphere is dominated by MA. When the atmosphere
36 is dominated by UIA then the AOD values are larger than MA. Normally, when BMA is the
37 dominant aerosol type, AOD values are large. In other words, if BMA is absent or small,
38 AOD will have a narrower range of distribution. As a result, only a moderate accuracy in the
39 AOD prediction is obtained for the post-monsoon and northeast monsoon (refer to Table 3).

40 By comparing the types of dominant aerosol observed during each monsoon, we observe that
41 the results, as obtained in Table 3, correlate well with the information from Fig. 3. Table 3
42 shows higher coefficients of determination of the proposed AOD prediction model, which can
43 be associated with higher amounts of BMA during the pre-monsoon and southwest monsoon
44 periods. Such observation implies that the aerosol types are possibly indirectly correlated with
45 the AOD prediction model. This result was also noticed by Chen et al. (2013). However, the
46 relationship between the predicted AOD and aerosol type as observed in our model is

1 qualitative and preliminary. Further study is needed. In addition, as mentioned in Lee et al.,
2 (2012) and Gupta et al., (2013), the relationship between AOD and particulate matter at the
3 surface depends also on extent of atmospheric mixing, relative humidity, chemical
4 composition, aerosol size distribution, etc.

5 **3.6 Validation of the predicted AOD**

6 In this subsection the procedure to validate the proposed AOD prediction model is presented.
7 To validate the model accuracy, $\{a_i\}$ was used to generate a set of 'predicted AOD' values
8 that are to be directly compared with those AOD values in data for subset 2. In this case, $\{a_i\}$
9 are optimized coefficients in Eq. (2); they are obtained from the data for subset 1 of the
10 overall dataset. This set of a_i shall be denoted as 'overall-calibrated $\{a_i\}$ '. The comparison is
11 shown in Fig. 5. The predicted AOD exhibits a high correlation with the measured AOD ($R^2 =$
12 0.68). The temporal characteristics of predictions between 2012 and 2013 are similar to those
13 of the measured AOD. Table 4 shows the performance of the predicted AOD as compared to
14 the measured ones in terms of RMSE and wMAPE. It is found that the RMSE for the
15 predicted AOD is nearly the same as that for the calibration data (as shown in Table 3).
16 Additionally, the error of the validation data is less than 1.0 % in terms of wMAPE (similar
17 accuracy was obtained for the calibration data).

18 To examine potential bias, the approach proposed by Lee et al. (2012) was performed to
19 remove outliers when the deviation of the predicted AOD was larger than the overall RMSE
20 (0.13). Approximately 21 % of the total data were removed using this method. After filtering,
21 the remaining data were used in the calibration of a_i (this set of a_i shall be denoted as
22 'overall_{POR}-calibrated $\{a_i\}$ ') in Eq. (2). Note that the values of a_i so obtained are different than
23 that using the original dataset. These two sets of a_i are optimized based on different data sets.
24 R^2 of this fitting increased to 0.92, with RMSE = 0.06 and wMAPE = 0.13 %. The values of
25 R^2 , RMSE and wMAPE for the cases with and without outliers removed are shown in Table 3.
26 Thus, by filtering the outliers, R^2 and RMSE were enhanced, but wMAPE only slightly
27 increased from 0.04 % to 0.13 %, although the error value remained less than 1 %.
28 Subsequently, these a_i coefficients (based on the outliers-removed data set) were used to
29 predict AOD, which were then compared against the measured values in data for subset 2 for
30 validation.

31 In the process of validation, the accuracies of the two sets of AOD values (one set is predicted
32 using $\{a_i\}$ with potential outliers removed, while the other without) are compared, see Table 4.
33 It is found that, in terms of R^2 , the AOD predicted using overall_{POR}-calibrated $\{a_i\}$ fails to
34 improve when compared to the AOD predicted using overall-calibrated $\{a_i\}$. The wMAPE of
35 AOD prediction before and after filtering the potential outliers are nearly the same. The two
36 sets of AOD predicted can be visually compared in the time series plot in Fig. 5. Such
37 observation implies that the removed data might not be the genuine outliers. In fact the errors
38 were attributed to non uniformly-loaded atmospheric aerosols at different altitudes. We
39 believe that the non-uniform atmospheric mixing caused the high deviations in our predicted
40 results, according to previous studies (Qiu and Yang, 2000; Toth et al., 2014). The proposed
41 model uses ground-based sources as input. It assumes (1) the aerosols are well-mixed, and (2)
42 the air above the planetary boundary layer (PBL) is aerosol free. Any aerosol, if present,
43 above the PBL is not taken into account by the model. If these assumptions are true, the model

1 can then be correctly compared to the columnar measurement of the sun photometer. However,
2 in reality, aerosol could be present above the PBL, or not always well-mixed, giving rise to
3 some uncertainties in the AOD predicted by the model. These uncertainties are quantified in
4 terms of RMSE.

5 Figure 5 indicates that most of the predicted AOD values are lower than the measured
6 counterparts. Tan et al. (2014c) analyze the underprediction in these values. They used
7 a LIDAR system to determine the vertical profile of aerosols in Penang and found that the
8 aerosol concentration decreased with height up to the planetary boundary layer (PBL). This
9 layer was less than 2 km during the study period. The large amount of transported aerosols
10 above the boundary layer yielded residual layers (Toth et al., 2014). Significant
11 underestimation of AOD occurred for thick residual layers. By comparing the measured and
12 predicted data in Fig. 5, it is found that only a few small time segments are significantly
13 underpredicted, possibly due to the presence of aerosol residual layers above the PBL. Studies
14 in Cyprus (Retalis et al., 2010) suggest that the extent of atmospheric mixing was relatively
15 homogeneous on scales of a few meters to tens of kilometers. Hence, the predicted results
16 were representative of the large samples. The predicted AOD was underestimated because all
17 measured data were taken from the ground. However, overprediction would be significant if
18 local burning were to occur near the measurement station.

19 To properly validate the prediction, these data coincide in time with those measured from API,
20 Vis, and AOD level 2. In our case, the LIDAR data coincided only once on 12 July 2013
21 (Fig. 6). Figure 6a shows the vertical profile of the aerosol backscatter coefficient as
22 a function of time (morning to evening). The brown vertical line represents the instance when
23 both the measured and predicted AOD could be compared with the LIDAR data.

24 Figure 6b displays the profiles of aerosol backscatter coefficient obtained at 10:00 and 11:00
25 a.m. local time, respectively. Aerosols had accumulated near the ground at 10:00 a.m. and our
26 model indicated that the predicted AOD was overestimated by 0.039. By contrast, most
27 aerosols at 11.00 a.m. were at a higher level; therefore the model predicted value was
28 underestimated by 0.044. Therefore, the predicted AOD values were acceptable because they
29 exhibited small deviations against the measured AOD. This result was thus valid as long as
30 the aerosols did not considerably differ at altitude levels beneath the planetary boundary layer.
31 The LIDAR data should be therefore considered as an independent validation method for
32 ground-based prediction models.

33 Aerosols are not always well-mixed in the atmosphere over Penang. Particles transported
34 within the free troposphere are a factor (Toth et al., 2014). If a significant number of elevated
35 aerosol plumes (equivalent to aerosol residual layer) occur over the region, then a large
36 deviation from the predicted value will be produced. Therefore, it can be inferred that a small
37 group of highly underpredicted results (Fig. 5) may be attributed to a significant layer of high-
38 level transported aerosol.

39 40 **3.7 Applications of the proposed model in the absence of measured AOD data**

41 In this section, we shall apply our AOD-predicting model in the absence of measured AOD
42 data. For the purpose of AOD prediction, the overall-calibrated coefficient $\{a_i\}$, will be used.

1 The overall_{POR}-calibrated coefficient $\{a_i\}$, obtained with **potential outliers removed**, are not
2 used as they may not be genuine outliers, as discussed in Sect. 3.6.

3 Our proposed model generates AOD data when those from AERONET are unavailable. We
4 described the procedure to predict AOD data. Only the API data for 7.00 a.m., 11.00 a.m., and
5 5.00 p.m. (local time) were available (<http://apims.doe.gov.my>) before 24 June 2013. The API
6 data were provided hourly beyond this date. In this study, approximately 5 % of the data were
7 discarded due to fog, rain, or thunderstorms, and only 4493 data points were retained. Figure 7
8 shows the predicted results from 2012 to 2013, which overlapped with the measured AOD
9 data to simplify the comparison. It is observed that the variation in the predicted AOD
10 matches with that of the measured AOD from AERONET. Hence, as an application of the
11 AOD predicting model, information missed out by sun photometer (i.e., AERONET) could be
12 reasonably well reproduced. These ‘retrieved’ AOD can be used in other aerosol studies. For
13 example, the diurnal variability of AOD can be significant, depending on location and
14 dominant aerosol type (Arola et al., 2013). They observed that the measurement-based
15 estimates of aerosol direct radiative forcing (also known as aerosol direct radiative effect) at
16 regional or individual sites are substantially influenced by the diurnal variability of AOD. In
17 Pandithurai et al. (2007), they found that the diurnal AOD variation depends on
18 meteorological factors such as relative humidity, winds, temperature and convection activities.
19 Our model provides a helpful means to investigate the uniqueness of diurnal variability of
20 AOD in different seasons of a specific region.

21 The boxes marked in Fig. 7 are the time windows in which AOD measurements are
22 unavailable. An independent method, i.e., LIDAR is used to estimate AOD at that particular
23 time window (refer to Fig. 8a). In our case, we set $L = 70$ sr, because this window period is
24 commonly associated with biomass burning aerosol (refer to the relative frequency of
25 dominant of aerosol types in the southwest monsoon, in Fig. 3). Additionally, other studies
26 conducted by Tesche et al. (2011) and Lopes et al. (2012) also suggested $L = 70$ sr for
27 biomass burning aerosols. Via the procedures as mentioned in Sect. 2, and using the obtained
28 aerosol backscatter coefficient and an assumed L , aerosol extinction coefficients were
29 calculated (based on Eq. 5). Integrating over these aerosol extinction coefficients, AOD values
30 were then estimated using Eq. 6.

31 If the LIDAR signals were affected by cloud, the AOD data calculated from the LIDAR signal
32 are removed. Then, the predicted AOD from our model and that calculated from LIDAR
33 signal are compared. **The result of the comparison** between the predicted AOD (by our model)
34 and that derived from LIDAR, as shown in Fig. 8b and c. Fig. 8b, shows that the correlation
35 between these two sets of data is high, as R^2 obtained is 0.86 with $RMSE = 0.20$. Fig. 8c also
36 indicates that the predicted AOD values from our model are within the error bars of estimated
37 AOD from the LIDAR signal. However, the AOD prediction model is less sensitive during
38 clear atmospheric conditions on 13 Aug (as shown in Fig. 8c).

39

40 **3.8 Comparison with other linear regression models**

41 The proposed model is compared against other AOD-predicting models from the literature.
42 This is done by comparing the predicted AOD values by our model against the measured
43 AOD in data for subset 1. Table 5 shows the R^2 values of selected AOD-predicting models
44 calculated using the data for subset 1 by our model (Sect. 2). Retalis et al. (2010) suggest

1 a simple linear regression analysis to predict AOD from the Vis data. Mahowald et al. (2007)
2 suggest a similar linear regression model for the AOD prediction model, in which the Vis data
3 were converted to surface extinction coefficients b_{ext} using the Koschmieder equation $\text{Vis} =$
4 K/b_{ext} , where $K (= 3.912)$ is the Koschmieder constant (Koschmieder, 1924). Two other AOD-
5 predicting models were also compared (Gao and Zha, 2010; Chen et al., 2013).

6 In these models, linear regression analysis for AOD and PM_{10} was carried out to predict the
7 surface air quality. The approaches can also be used to retrieve AOD after appropriate
8 conversion procedures. Initially, we converted the API data into PM_{10} via the guidance on air
9 pollutant index from DOE (1997). The obtained PM_{10} values were then inputted into the
10 linear regression formula to predict AOD. The linear regressions in these models yielded $R^2 \leq$
11 0.6 with RMSE approximately 0.16 and above, which was lower than that of our model ($R^2 \leq$
12 0.72 with RMSE = 0.13). wMAPE of these models (0.05 % ~ 0.08 %) were found to be
13 similar but slightly higher than the present model (0.05 %). These figures are reported in
14 Table 5.

15 **4 Conclusions**

16 Seasonal variation in primary aerosol types and their physical characteristics at Penang,
17 Malaysia are analyzed from February 2012 to November 2013. The aerosol types for
18 a specific monsoonal period were determined by applying threshold criteria to scatter plots, in
19 which Angstrom exponent is plotted against aerosol optical depth (AOD). The threshold
20 criteria from Smirnov et al. (2002b, 2003), Pace et al. (2006), Kaskaotis et al. (2007),
21 Toledano et al. (2007), Salinas et al. (2009), and Jalal et al. (2012) were used to distinguish
22 the aerosol types. The testing results indicate that the threshold criteria by Toledano
23 et al. (2007) were the most reliable, because of the minimal occurrence value of the
24 indistinguishable aerosols (referred as mixed-type aerosols, MIXA).

25 For the study period, biomass burning aerosols (BMA) abruptly increased during the
26 southwest monsoon period, because of active open burning activities in local areas and
27 neighboring countries. During the northeast monsoon period, the optical properties (e.g., size
28 distribution patterns) of the aerosols were unique. Two noticeable peaks were observed in the
29 occurrence frequency of the Angstrom exponents compared with the single peaks for other
30 monsoon seasons. These results were attributed to the mixing of aerosols from local sources
31 with those from the northern part of Southeast Asia, caused by the northeast monsoon winds.
32 Urban and industrial aerosols (UIA) and marine aerosol (MA) were the major aerosols in
33 Penang throughout the year. Dust aerosols (DA) negligibly contributed to the emissions in
34 Penang. The variation in aerosol types for different monsoon seasons clearly yields distinct
35 optical properties.

36 Previous models have used simple regression analysis between AOD and meteorological
37 parameters to predict the corresponding AOD data. In this study, multiple regression analysis
38 was used **in analyzing the proposed model**. Two predictors (API and Vis) were introduced to
39 increase statistical reliability. To verify the robustness of multiple regression analysis, in
40 contrast to the simple regression approach, AOD data based on previous simple models were
41 retrieved (Mahowald et al., 2007; Gao and Zha, 2010; Retalis et al., 2010; Chen et al., 2013).
42 R^2 , RMSE and wMAPE values in our calibration model are ≤ 0.72 , 0.13, 0.05 % respectively.
43 The accuracies are obtained by comparing the predicted AOD values **in the current study**

1 against measured AOD in data for subset 1. These figures are compared with the results of
2 other relevant work, which obtained $R^2 \leq 0.60$ and RMSE approximately 0.16 and above. The
3 comparison indicates that the quality of our AOD prediction is statistically better than those
4 simple models, **which makes sense** given its tuning to local condition.

5 Predicted AOD from our model are compared with the data derived from a LIDAR system.
6 The values of R^2 and RMSE (0.86 and 0.20) indicate favorable agreement between our model
7 and LIDAR-derived data at wavelength 355 nm. This has added additional weight to the
8 robustness of the developed AOD prediction model.

9 Our algorithm predicts AOD data during non-retrieval days caused by the frequent occurrence
10 of clouds in the equatorial region. The proposed model yields reliable near real-time AOD
11 data despite the availability of the measured data for limited time points. The predicted AOD
12 data are beneficial for monitoring aerosols in short- and long-term scenarios, their behavior,
13 and provides supplementary information for climatological studies and monitoring aerosol
14 variation.

15 The technique proposed in this work nevertheless ought to be further **stress-tested** for the
16 extent of its feasibility by applying it in more cases **using a higher** volume of data. This
17 technique is pragmatic and cost effective for such environmental study.

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1

2 **Tables**

3

4 **Table 1.** Average values of model-related parameters from the database collected from
 5 November 2011 to November 2013 in USM Penang (latitude, 05°21' N; longitude, 100°18' E;
 6 elevation, 51 m).

Month	AOD₅₀₀	sigma AOD₅₀₀	Angstrom 440–870	sigma Angstrom 440–870	PW	sigma PW	N	Number of Month
JAN	0.24	0.09	1.33	0.18	4.19	0.47	21	1
FEB	0.21	0.09	1.39	0.23	4.44	0.58	18	2
MAR	0.36	0.16	1.41	0.19	4.15	0.58	31	2
APR	0.32	0.19	1.42	0.16	4.78	0.53	29	2
MAY	0.19	0.07	1.10	0.33	4.48	0.43	11	2
JUN	0.48	0.35	1.30	0.33	4.56	0.37	14	2
JUL	0.31	0.18	1.39	0.21	4.50	0.49	14	2
AUG	0.73	0.39	1.50	0.19	4.58	0.25	13	1
SEP	0.35	0.23	1.40	0.17	4.78	0.45	14	2
OCT	0.19	0.08	1.31	0.19	4.48	0.32	16	2
NOV	0.18	0.07	1.31	0.20	4.72	0.41	24	3
DEC	0.21	0.04	1.41	0.20	4.67	0.27	8	1
YEAR	0.31	0.16	1.36	0.10	4.53	0.20	213	22

7

1 **Table 2.** Threshold values of AOD and Angstrom₄₄₀₋₈₇₀ for aerosol classification. Abbreviations: MA = maritime, DA = dust, UIA = urban
 2 and industrial, BMA = biomass burning, MIXA = mixed-type aerosols. MIXA represents indistinguishable aerosol type that lies beyond the
 3 threshold ranges.

Aerosol Type	Jalal et al. (2012)		Toledano et al. (2007)		Salinas et al. (2009)		Pace et al. (2006) and D. Kaskaotis (2007)		Smirnov (2002b, 2003)	
	Angstrom ₄₄₀₋₈₇₀	AOD	Angstrom ₄₄₀₋₈₇₀	AOD	Angstrom ₄₄₀₋₈₇₀	AOD	Angstrom ₄₄₀₋₈₇₀	AOD	Angstrom ₄₄₀₋₈₇₀	AOD
MA	0.5–1.7	≤ 0.3	0–2	≤ 0.2	0.5–1.7	≤ 0.15	≤ 1.3	≤ 0.06	≤ 1.0	≤ 0.15
DA	≤ 1.0	≥ 0.4	≤ 1.05	≥ 0.11 (only this value is for AOD ₈₇₀)	≤ 1.0	≥ 0.4	≤ 0.5	≥ 0.15	≤ 0.7	≥ 0.2
UIA	≥ 1.0	0.2–0.4	≥ 1.05	0.2–0.35	≥ 1.0	0.2–0.4	≥ 1.5	≥ 0.1	≥ 1.5	≥ 0.4
BMA	≥ 1.0	≥ 0.7	≥ 1.4	≥ 0.35	≥ 1.0	≥ 0.8				

4

1 **Table 3.** Calibration results (data for subset 1) for the AOD prediction model [Eq. (2)] from 2012 and 2013
 2 data.

Monsoon	R^2	RMSE	wMAPE (%)	N
Northeast monsoon	0.41	0.11	0.40	129
Pre-Monsoon	0.64	0.11	0.37	65
Southwest monsoon	0.77	0.17	0.08	117
Post-monsoon	0.42	0.06	0.21	83
Overall	0.72	0.13	0.04	394
Overall _{POR}	0.92	0.06	0.13	307

3 Note: POR = potential outliers removed, N = number of data

4

5 **Table 4.** Validation results (data for subset 2) for the AOD prediction model [Eq. (2)] from 2012 and 2013
 6 data.

	R^2	RMSE	wMAPE (%)	N
Overall (using overall-calibrated $\{a_i\}$)	0.68	0.14	0.05	395
Overall (using overall _{POR} -calibrated $\{a_i\}$)	0.68	0.14	0.03	395

7 Note: POR = potential outliers removed, N = number of data

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9

1 **Table 5.** R^2 values of the AOD predicted by selected linear regression models from the literature. The values
 2 of R^2 , RMSE and wMAPE shown in this table are obtained by comparing the predicted AOD values against
 3 measured AOD data from subset 1.

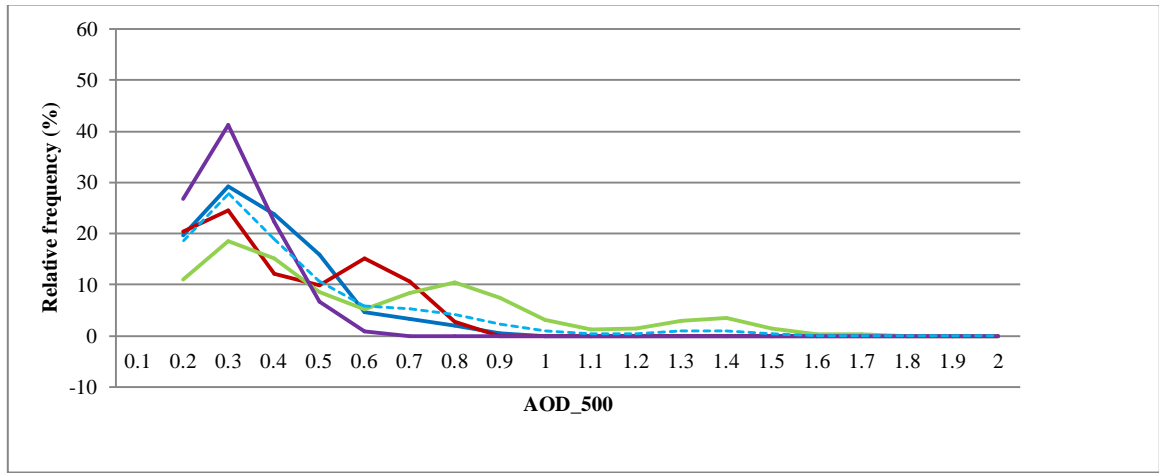
Model	Author(s)	R^2	RMSE	wMAPE (%)
$AOD = a_0 + a_1(Vis)$	(Retalis et al., 2010)	0.56	0.17	0.08
$AOD = a_0 + a_1(b_{ext})$	(Mahowald et al., 2007)	0.58	0.17	0.07
$AOD = a_0 + a_1(PM10)$	(Gao and Zha, 2010;Chen et al., 2013)	0.60	0.16	0.05
$AOD = a_0 + a_1(Vis) + a_2(Vis)^2 + a_3(Vis)^3 + a_4(API) + a_5(API)^2 + a_6(API)^3$	Current Study	0.72	0.13	0.05

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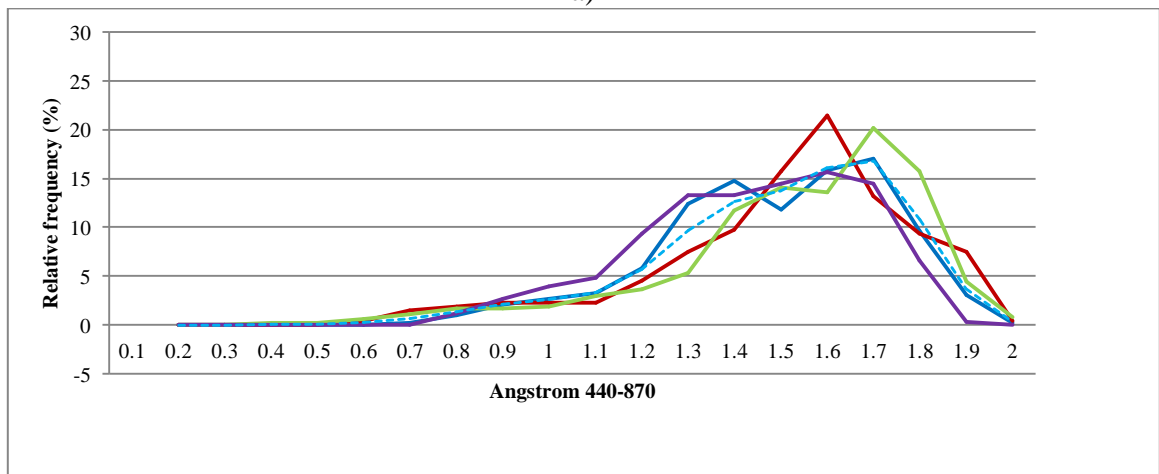
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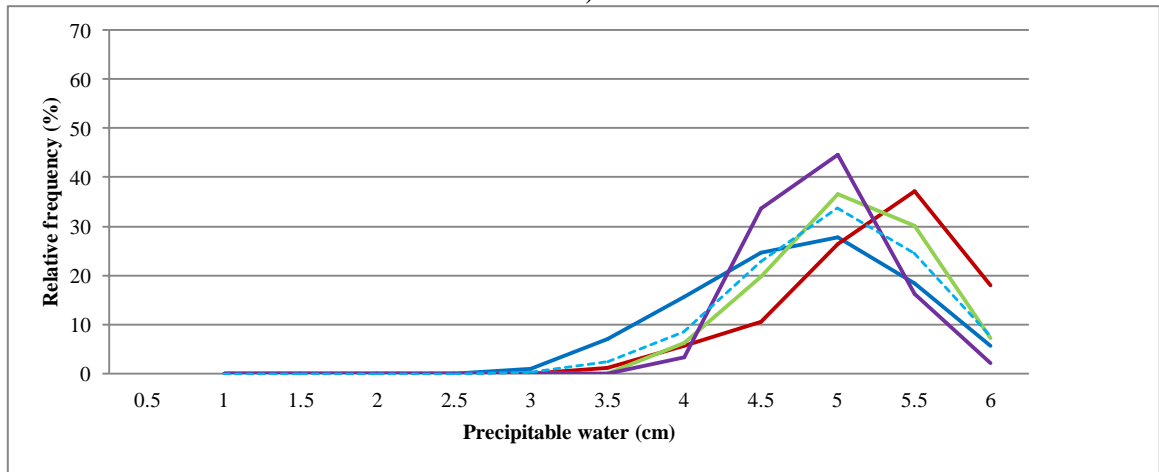
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2 **Figures**



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4 a)



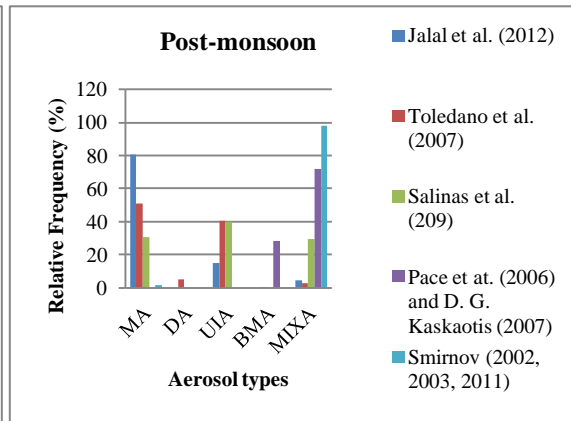
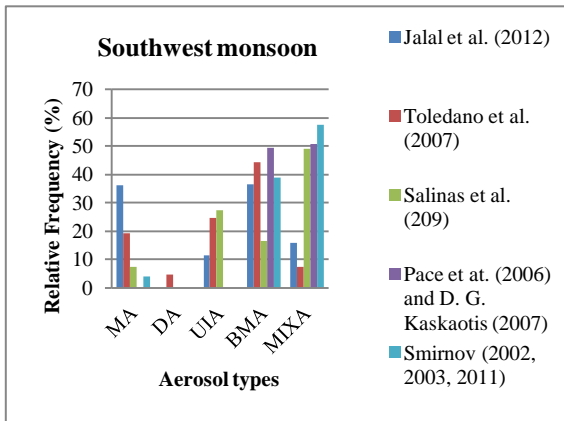
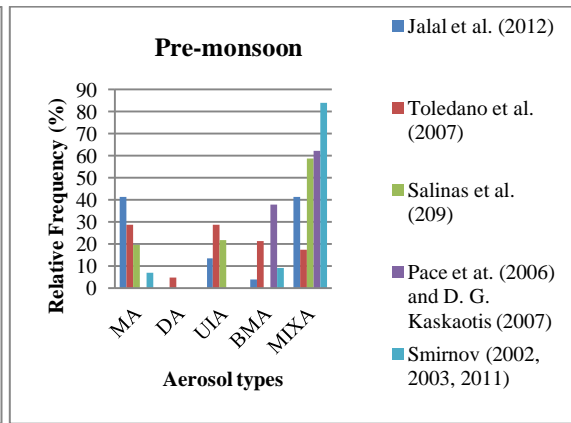
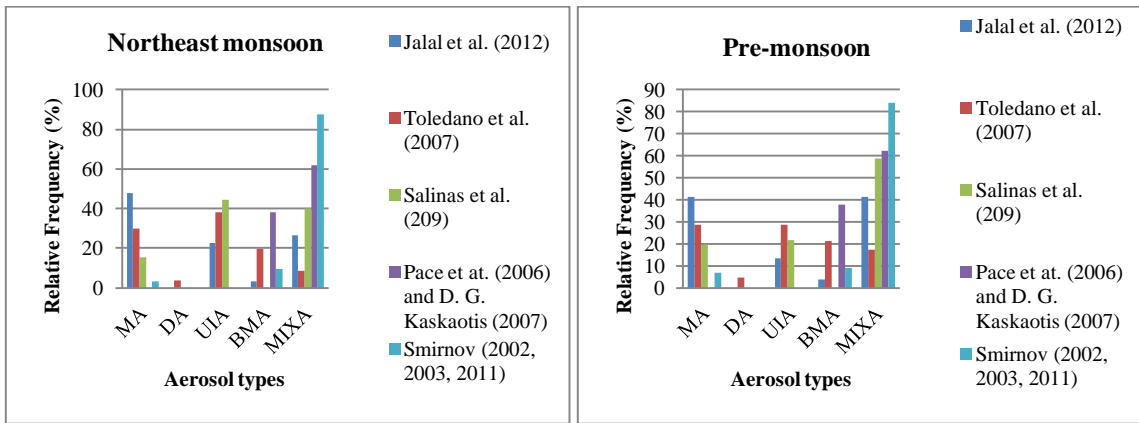
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6 b)



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8 c)

9 — Northeast monsoon — Pre-monsoon — Southwest monsoon — Post-monsoon — All

10 **Figure 1.** Seasonal relative frequencies of occurrences of (a) AOD_500, (b) Angstrom₄₄₀₋₈₇₀, and (c) PW in
11 Penang for February 2012 to November 2013. Each curve was smoothed by using a moving average
12 technique.



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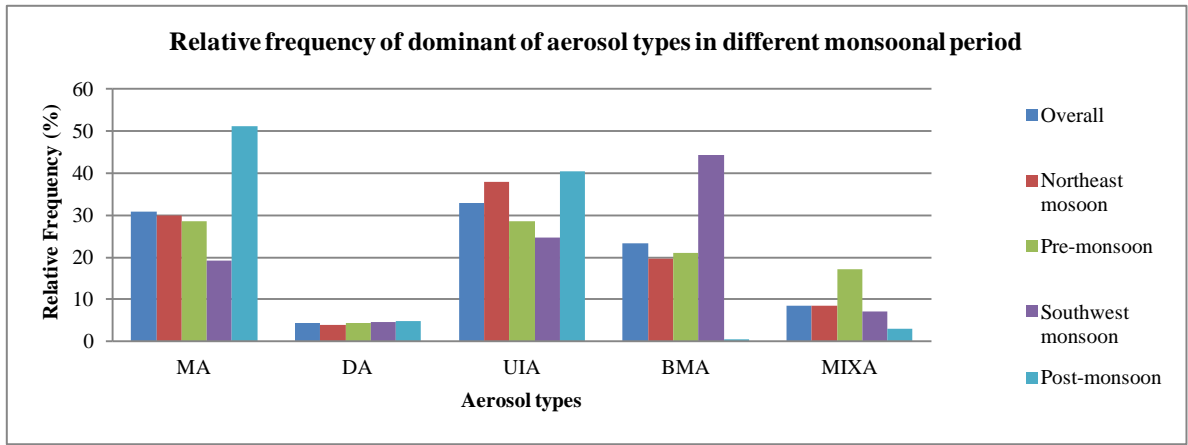
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6 **Figure 2.** Classification of aerosol types for a) December–March, b) April–May, c) June–September, and d)
 7 October–November based on AOD–Angstrom_{440–870} scatter plots by proposed thresholds.

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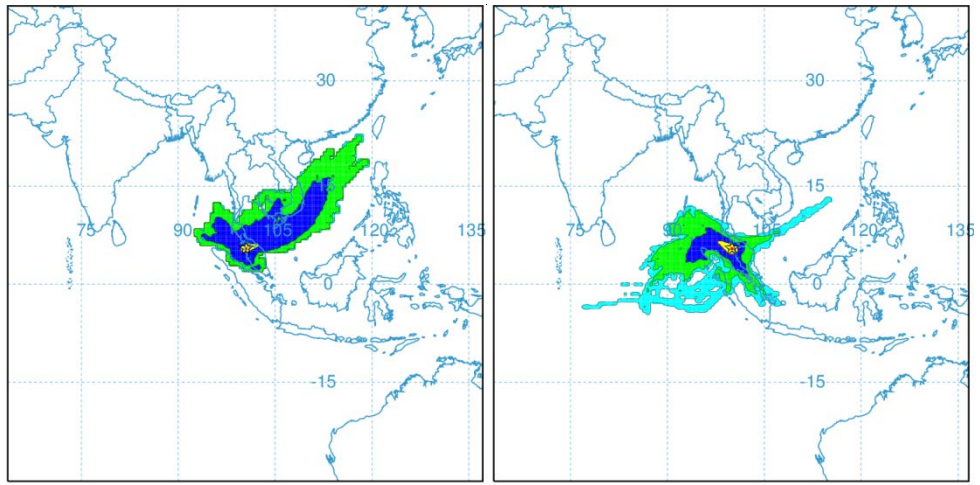


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3 **Figure 3.** Seasonal classification of aerosol types based on AOD–Angstrom_{440–870} scatter plots by the
4 threshold proposed by Toledano et al. (2007).

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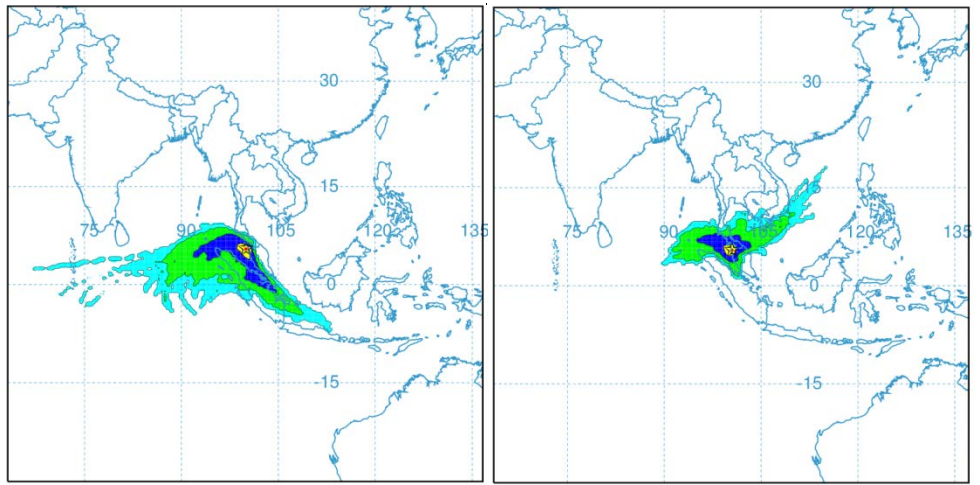
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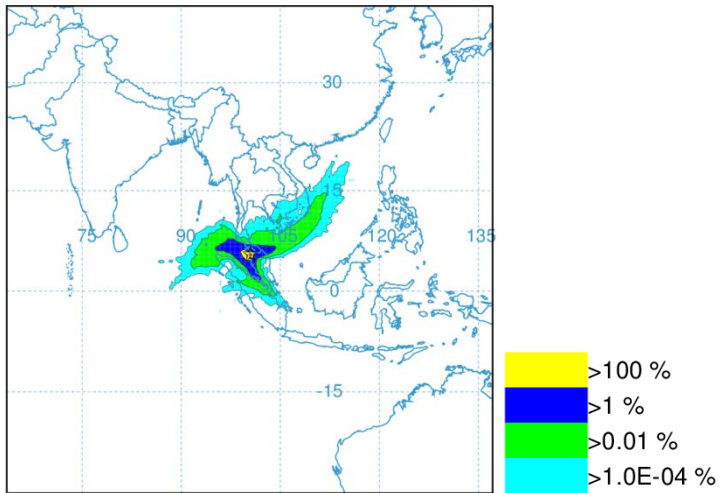
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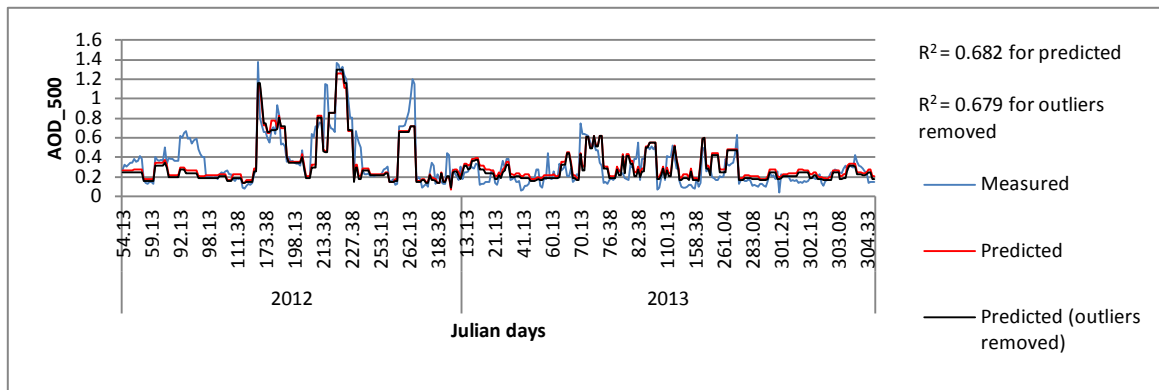
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e)

8 **Figure 4.** Seasonal back-trajectory frequency plot by the HYSPLIT_4 model (Draxler and Hess, 1998) for a)
9 northeast monsoon, b) pre-monsoon, c) southwest monsoon, d) post-monsoon, and e) overall study period at

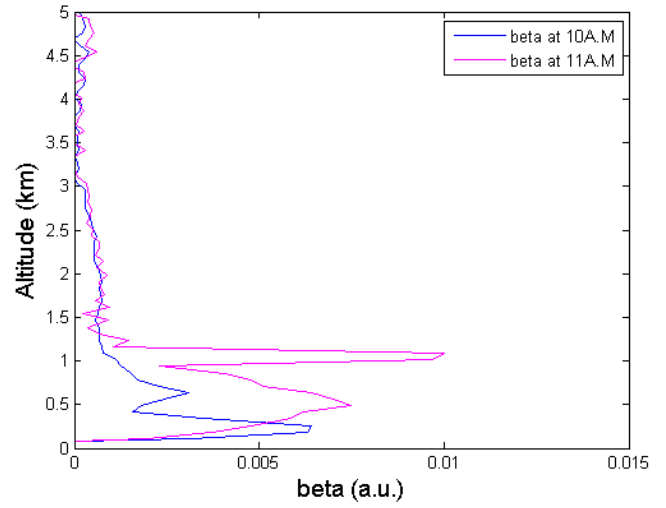
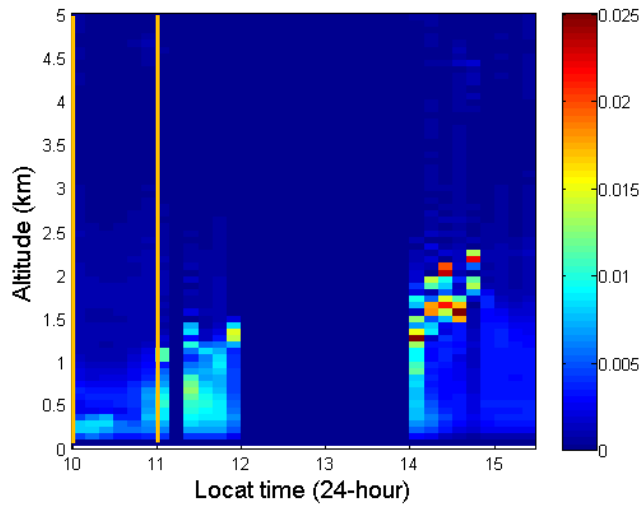
1 Penang, which was marked as a five-edged star (5.50° N and 100.50° E, from altitude 51 m).



2

3 **Figure 5.** Predicted and measured AOD at 500 nm for 2012 and 2013 for validation dataset (subset 2, with
4 395 data points). The plot in black are predicted AOD values using $\{a_i\}$ obtained dataset with potential
5 outliers removed, whereas that in red is with dataset without removing potential outliers.

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4 **Figure 6.** a) Profiles of the aerosol backscatter coefficients ($\text{km}^{-1}\text{sr}^{-1}$) recorded on 12 July 2013. No data were
 5 acquired from 12 PM to 2 PM. The brown lines represent the moment of acquisition of sun photometer; b)
 6 profiles of the aerosol backscatter coefficient (beta) obtained from 10 AM to 11 AM for the brown lines in a).

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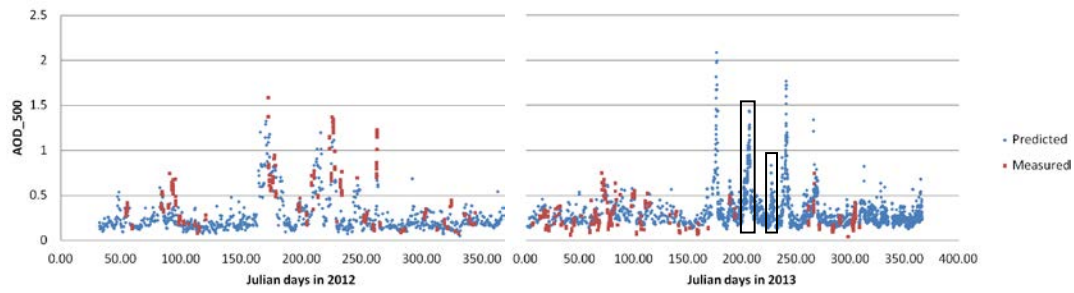
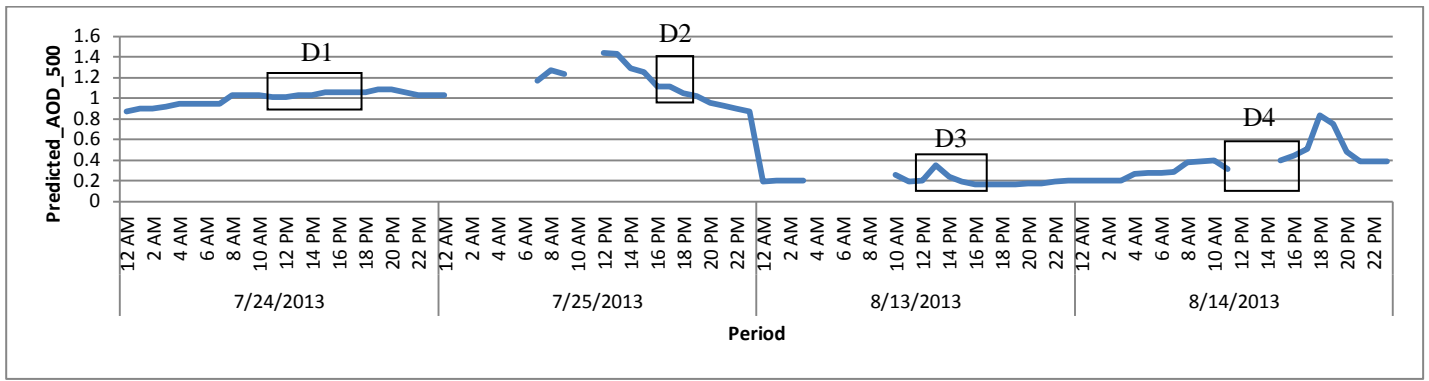
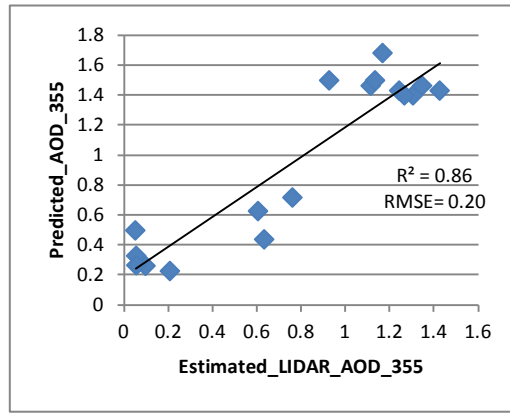


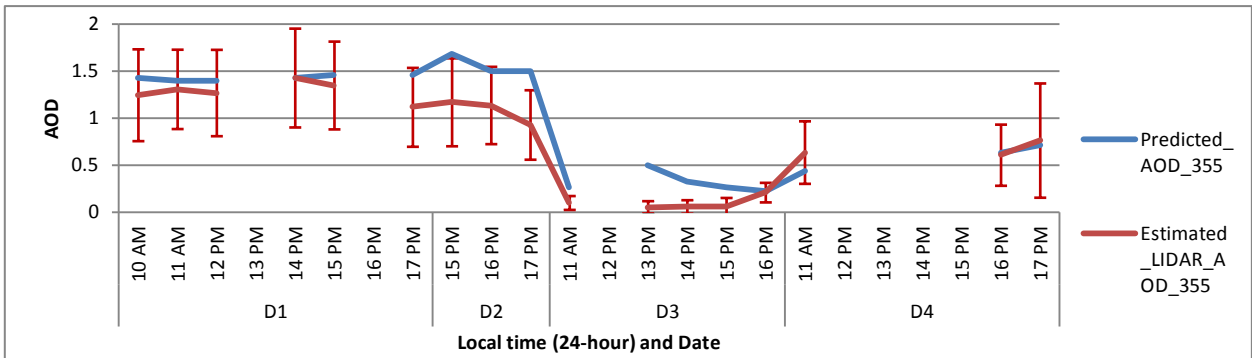
Figure 7. Predicted AOD₅₀₀ data plotted against the period from 2012 to 2013 (input all Vis and API available data into the established model to predict AOD, with 4493 data points). Rectangles 1 and 2 correspond to the data recorded on 24–25 July and 13–14 August 2013, respectively. These data were used for comparison with those obtained from LIDAR (**Fig. 8**).



a)



b)



c)

7 **Figure 8.** a) Hourly retrieved AOD recorded on a) 24–25 July and 13–14 August 2013 (the gaps are due to
 8 fog, rain, or when API value is predominantly caused by O_3 but not PM_{10}). b) A scatter plot for AOD₃₅₅
 9 predicted from our model versus the AOD calculated from Raymetrics LIDAR system. c) Predicted AOD
 10 from our model and estimated AOD from LIDAR plotted versus local time and date (the gaps indicate no
 11 data available at the particular time due to LIDAR system was switched off or cloud contamination above the
 12 LIDAR system). Error bars for estimated AOD from LIDAR are shown.