ANALYSES OF AEROSOL OPTICAL PROPERTIES AND DEVELOPMENT OF ITS ALGORITHM DURING SEASONAL MONSOON CIRCULATION

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by

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LIST OF ABBREVIATION

ADEOS-1	Advanced Earth Observing Satellite 1
AERONET	Aerosol Robotic Network
AMSL	Above Mean Sea Level
ARL	Air Resources Laboratory
ANOVA	Analysis of Variance
AOD	Aerosol Optical Depth
AOD_440	Aerosol Optical Depth at Wavelength 440 nm
AOD_500	Aerosol Optical Depth at Wavelength 500 nm
API	Air Pollution Index
ASD	Aerosol Size Distribution
ATCR	Annual Tropical Cyclone Report
ATSR-2	Second Along Track Scanning Radiometer
AVHRR	Advanced Very High Resolution Radiometer
BMA	Biomass Burning Aerosol
CCN	Cloud Condensation Nuclei
CMA	China Meteorological Administration
DA	Dust Aerosol
DDV	Dense Dark Vegetation
DOE	Department of Environment
ESRL	Earth System Research Laboratory
FLAMBE	Fire Locating and Modeling of Burning Emissions
GUI	Graphical User Interface
GDAS	Global Data Analysis System
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory model
IPCC	Intergovernmental Panel for Climate Change
ITCZ	Inter Tropical Convergence Zone

JTWC	Joint Typhoon Warning Center
LIDAR	Light Detection And Ranging
MA	Maritime Aerosol
MC	Maritime Continent
MISR	Multi-angle Imaging SpectroRadiometer
MIXA	Mixed-Aerosol
MODIS	Moderate Resolution Imaging Spectroradiometer
NASA	National Aeronautics and Space Administration
NCAR	National Center for Atmospheric Research
NCEP	National Center for Environment Prediction
NOAA	National Oceanic and Atmospheric Administration
NSMC	National Satellite and Meteorological Center
OLR	Outgoing Longwave Radiation
PC	Photon Counting
PM	Particulate Matter
PMT	Photomultiplier Tube
POES	Polar Orbiting Environmental Satellites
POLDER	Polarization and Directionality of Earth’s Reflectances
POR	Potential Outliers Removed
PW	Precipitable Water
RCS	Range Corrected Signal
RMSE	Root Mean Square Error
RH	Relative Humidity
SEA	Southeast Asia
SLP	Sea Level Pressure
TC	Tropical Cyclone
TOD	Total Optical Depth
TOMS	Total Ozone Mapping Spectrometer

TRMM	Tropical Rainfall Measuring Mission
UIA	Urban and Industrial Aerosol
UTC	Coordinated Universal Time
UV	Ultra-violet
wMAPE	Weighted Mean Absolute Percentage Error

LIST OF SYMBOLS

$a_0, a_1, ..., a_n$	Coefficients of First Predictor with n, Number of Order
α	Ångström Exponent
$Ångström_{440-870}$	Ångström Exponent at Wavelength between 440 and 870 nm
$b_0, b_1, ..., b_n$	Coefficients of Second Predictor with n, Number of Order
β	Ångström Turbidity Coefficient
$D_1, D_2, ..., D_N$	Data with N, Number of Data Points
i	Individual Measurements
λ	Wavelength
m	Measured Values
%	Percentage
p	Predicted Values
R	Correlation Coefficient
R^2	Coefficient of Determination
S_1	Data Set 1
S_2	Data Set 2
τ_a	Aerosol Optical Depth
Temp	Temperature
Vis	Visibility

ANALISIS SIFAT-SIFAT OPTIK AEROSOL DAN PEMBANGUNAN ALGORITMA SEMASA PEREDARAN MONSUN BERMUSIM

ABSTRAK

Kesan pembakaran dan pencemaran ke atas Asia Tenggara (SEA) adalah kebimbangan besar kepada penyelidik perubahan iklim global. Lokasi geografi, topografi dan faktor-faktor meteorologi yang mempengaruhi cuaca SEA semuanya menyumbang kepada kerumitan sistem aerosol di seluruh dunia. Walau bagaimanapun, litupan awan yang kerap dalam rantau ini menyebabkan kehilangan data semasa pencerapan melalui teknik penderiaan jauh. Oleh itu untuk mendapatkan kedalaman optikal aerosol (AOD) secara berterusan adalah tugas yang sukar. Untuk menangani isu-isu tersebut, kajian ini mula-mula mengkaji korelasi antara aktiviti kebakaran, pelepasan asap, peristiwa hujan, aktiviti taufan dan sinaran gelombang panjang keluar di kedua-dua kawasan utara dan selatan SEA antara 2011 dan 2013. Analisis korelasi antara parameter-parameter di utara dan selatan SEA telah menunjukkan bahawa selatan SEA sering dilindungi oleh awan. Dalam kajian ini satu algoritma empirikal dibangunkan untuk menggantikan data yang terhapuskan disebabkan liputan awan yang kerap di kawasan kajian terpilih di Asia Tenggara selatan, iaitu Pulau Pinang. Ukuran darat seperti penglihatan dan indeks pencemaran udara digunakan dalam model sebagai data peramal untuk menjana data AOD yang hilang dari AERONET. Pekali model empirik ditentukan melalui analisis regresi bukan linear berbilang; pekali model ditentukur mempunyai pekali penentuan $R^2 = 0.72$. Nilai AOD ramalan daripada model tersebut dihasilkan dengan menggunakan pekali yang ditentukur dan dibandingkan dengan data ukuran melalui ujian statistik piawai, menghasilkan $R^2 = 0.68$ sebagai nilai ketepatan pengesahan. Keralatan dalam ralat peratus mutlak min berpemberat yang ditentukan adalah kurang daripada 0.50% berbanding dengan data sebenar. Keputusan menunjukkan bahawa model yang dicadangkan cekap meramalkan data AOD. Selain itu, keputusan daripada model tersebut dibandingkan dengan data daripada satu sumber bebas, iaitu Sistem Pengesanan Cahaya dan Penjejakan (LIDAR) untuk menghasilkan hubungan yang baik, dengan $R^2 = 0.86$. Sifat-sifat optik aerosol di Pulau Pinang, Malaysia, dianalisis untuk empat musim monsun termasuk monsun timur laut, pra-monsun, monsoon barat daya, dan pasca monsun berdasarkan data dari *Aerosol Robotic Network* (AERONET) direkodkan dari Februari 2012 hingga November 2013. Jenis-jenis aerosol di Pulau Pinang bagi setiap tempoh monsun secara kuantitatifnya dikenal pasti berdasarkan plot penyebaran eksponen Angstrom terhadap AOD. Kedua-dua sifat optik aerosol dan jenis aerosol digunakan untuk mengkaji model ramalan AOD yang dicadangkan. Model ramalan AOD bermusim yang dihasilkan telah menunjukkan hubungan yang jelas dalam jenis aerosol yang dominan dengan sifat-sifat optik yang berbeza dan corak trajektori kebelakang. Model yang dicadangkan juga boleh digunakan untuk meramalkan AOD pada setiap panjang gelombang antara 340 nm hingga 1020 nm. Sebagai ilustrasi penggunaan model, saiz aerosol yang ditentukan menggunakan data AOD untuk Pulau Pinang telah dibandingkan dengan yang ditentukan dengan menggunakan model. Ini dilakukan dengan memeriksa kelengkungan dalam plot-plot ln[AOD] lawan ln[gelombang] yang menggunakan data AOD daripada ukuran dan juga daripada ramalan pada tahun 2012 dan 2013. Berdasarkan konsistensi dalam kelengkungan plot-plot log lawan log tersebut, disimpulkan bahawa Pulau Pinang adalah dikuasai oleh mod aerosol halus pada tahun 2012 dan 2013. Keputusan ini menunjukkan bahawa model ramalan AOD yang dicadangkan menggunakan ukuran rutin sebagai input adalah alat yang berguna untuk pemantauan berkala variasi aerosol pada masa ketiadaan data AERONET. Model ramalan AOD tersebut boleh berfungsi sebagai alat alternatif untuk mengukur AOD secara jangka pendek dan jangka panjang dan boleh memberikan maklumat tambahan bagi kajian iklim dan pemantauan variasi aerosol.

ANALYSES OF AEROSOL OPTICAL PROPERTIES AND DEVELOPMENT OF ITS ALGORITHM DURING SEASONAL MONSOON CIRCULATION

ABSTRACT

The impact of biomass burning and pollution on Southeast Asia (SEA) is of considerable concern to global climate change researchers. Geographical location, topography and meteorological factors affecting SEA all contribute to the complexity of the aerosols system worldwide. However, frequent cloud cover in the region results in missing data during observations by remote sensing techniques. Therefore obtaining continuous aerosol optical depth (AOD) measurements is a difficult task. As a way to address such issues, this study first investigates the inter-relationship among fire activity, ground smoke emission, rainfall events, typhoon activity and outgoing longwave radiation in both the northern and southern regions of SEA during seasonal monsoon between 2011 and 2013. The analysis of the correlation among these parameters in northern and southern SEA shows that southern SEA is frequently covered by cloud. In this study an empirical algorithm is developed to compensate for the eliminated data due to frequent cloud coverage in a selected study area in southern SEA, namely Penang. Ground-based measurements such as visibility and air pollutant index are used in the model as predictor data to retrieve the missing AOD data from Aerosol Robotic Network (AERONET). The empirical model coefficients are determined through multiple non-linear regression analysis; the calibrated model coefficients have a coefficient of determination of $R^2 = 0.72$. The predicted AOD of the model is generated on the basis of these calibrated coefficients and is compared with data measured through standard statistical tests, yielding $R^2 = 0.68$ as validation accuracy. The error in weighted mean absolute percentage error is determined to be less than 0.50% than that of the actual data. The results reveal that the proposed model efficiently predicts the AOD data. Additionally, the results of the model are compared with other independent source, i.e., light detection and ranging system (LIDAR) data to yield good correspondence, with $R^2 = 0.86$. The optical properties of aerosols in Penang, Malaysia, are analyzed for four monsoonal seasons including northeast monsoon, pre-monsoon, southwest monsoon, and post-monsoon based on data from the AERONET recorded from February 2012 to November 2013. The aerosol types in Penang for each monsoonal period are quantitatively identified according to the scattering plots of the Ångström exponent against the AOD. Both aerosol optical properties and aerosol types are used to examine the proposed AOD-predicting model. The established seasonal AOD prediction models are observed to have a clear relationship in the dominant aerosol type with different optical properties and back-trajectories patterns. The proposed model is also applicable for predicting the AOD at each studied wavelength within 340 nm and 1,020 nm. As an illustration to apply the model, the aerosol size determined using measured AOD data for Penang was compared with that from the model. This was done by examining the curvature in the ln [AOD] versus ln [wavelength] plots using both measured and predicted AOD data in 2012 and 2013. Based on the consistency in the curvature of the log-log plots, it was concluded that Penang was dominated by fine mode aerosol in 2012 and 2013. These results indicate that the proposed AOD prediction model using routine measurements as input is a promising tool for regular monitoring of aerosol variation during non-retrieval times. The AOD prediction model can serve as an alternative tool for measuring short- and long-term AOD and can provide supplementary information for climatological studies and monitoring of aerosol variation.

CHAPTER 1

INTRODUCTION

1.0 Overview

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

Southeast Asia (SEA) stands out globally because it hosts some of the most complex meteorological and environmental conditions, making remote sensing difficult both for the Aerosol Robotic Network (AERONET) and satellites (Reid *et al.*, 2013). Cloud cleared data leave gaps in the remote sensing data record, and residual cloud contamination of remotely sensed data creates challenges in the study of aerosols (Chew *et al.*, 2011; Campbell *et al.*, 2013). Moreover, anthropogenic biomass burning activities have increased dramatically in recent decades for land preparation and forest clearance (Field *et al.*, 2009). These fire activities result in trans-boundary and long-range transport of aerosols, which often affects air quality in both the source and surrounding regions (Hyer and Chew, 2010; Reid *et al.*, 2013; Salinas *et al.*, 2013; Lin *et al.*, 2014c) because these aerosols mix with locally generated aerosols (Engling *et al.*, 2014). Therefore, it is potentially valuable to develop a regional/local model for estimating and monitoring AOD.

Asian sources are known to differ from those in Europe and North America (Salinas *et al.*, 2009). Additional amounts of absorbing soot and organic components are included in the Asia–Pacific atmosphere because of the substantially greater amounts of coal and biomass burning emissions and long-range transport by wind (Lelieveld *et al.*, 2001; Huebert *et al.*, 2003; Seinfeld *et al.*, 2004). However, aerosols in SEA remain poorly characterized, which makes the global impact of aerosols on the Earth's climate difficult to quantify. Therefore, the origins of aerosol sources and their distribution in the atmosphere should be determined to understand why different locations have different aerosol types and are affected by environmental development, monsoons, and southern oscillation variation, and seasonal change.

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

Development of an empirical model for producing reliable AOD estimates for aerosol monitoring at local scales is novel and necessary for SEA and has potential global applications (Chen *et al.*, 2013; Fan *et al.*, 2013). Several researchers have used models as alternative tools for predicting AOD values by using various ground-based meteorology measurements (Wang *et al.*, 2009; Qin *et al.*, 2010; Lin *et al.*, 2014a). However, this approach has not been applied thus far over the Malaysia Peninsula region of SEA. In addition, aerosol transported pathways and dominant aerosol type in these areas are also unclear.

1.1 Atmospheric Aerosols

Aerosols, defined as systems of solid or liquid particles suspended in the atmosphere, play vital roles in air pollution, local weather, and climate change issues. These particles are larger than a few molecules but are smaller than cloud droplets. The diameters of aerosols range from 1×10^{-3} to 100 μm. They can be directly emitted into the atmosphere as primary aerosols or can be formed in the atmosphere as secondary aerosols by gas-to-particle conversion. They can originate from both natural sources such as sea spray, dust storms, and volcano eruption; and from anthropogenic sources such as open burning for plantation, and, urban and industrial emissions. These two sources (natural and anthropogenic) are generally difficult to distinguish.

Larger aerosol particles such as dust and sea salt may fall out of the atmosphere near the source. Fine aerosol particles such as those from urban and biomass burning may remain suspended for many days, enabling travel at transboundary distances. Compared with that of greenhouse gasses, the lifetime of sub-micron aerosols in the atmosphere is smaller, at a scale of days to weeks. In addition, numerous different sources of aerosol emission may be mixed in the atmosphere. Thus, aerosols are highly variable in both space and time.

Aerosols have direct and indirect effects on the climate system. The indirect effect is reflected in their ability to act as condensation nuclei that lead to cloud formation with small droplets. The direct effect includes the umbrella effect of scattering of solar radiation, resulting in atmospheric cooling, and absorption of solar radiation by aerosols, resulting in atmospheric heating. In the scattering effect of solar radiation by aerosol, a fraction of solar irradiance is scattered back into space, and another fraction of the incoming radiation is scattered in the forward direction to reach the Earth's surface. In the absorption effect of solar radiation by aerosol, mineral dust and carbonaceous aerosols absorb short- and long-wave radiation.

To study and understand aerosol characteristics, AOD is an important parameter used to summarize the optical activity of aerosols in the atmospheric column. AOD can be retrieved from either satellite data or ground-based data. In this study, AOD retrieval only from the ground-based data is discussed. The monitoring system for ground-based data is simple, highly accurate, and easily maintained. Ground-based AOD can be retrieved from various instruments such as a sun-photometer; hand-held sun-photometer, known as the Microtops II; light detection and ranging system (LIDAR); spectroradiometer; Brewer spectrophotometer; and pyrheliometer. Holben *et al.* (2001) comprehensively discussed and summarized the existing AOD measurement devices, citing numerous references from 1969 to 2000. The sun-photometer, a highly accurate instrument with high temporal resolution, has been employed worldwide in recent decades under the AERONET program.

1.2 **Problem Statement**

The availability of AOD data is limited owing to the difficulty to maintain year-long measurements. In addition, cloud contamination has also rendered removal of many AOD data. The problem of limitation in the availability of AOD data occurs in all instruments, including AERONET, causing many gaps in the database. This issue has to be resolved for effective monitoring and the understanding of the variations in aerosols in a specific location. This is especially so for the case in SEA due to the complexity in the meteorological and environmental conditions that cause some difficulties in applying remote sensing techniques to study the aerosol.

The differences in the effects of environmental and meteorological factors in between northern and southern SEA on fires activity, ground smoke emission, rainfall, typhoons, and outgoing longwave radiation (OLR) are unclear, and need to be investigated. For example, the inter-relationship of these factors in term of fire activity, smoke emission, precipitation, typhoon activity and outgoing longwave radiation in both regions might directly or indirectly influence the aerosol variation in local and regional areas of a country. Global models which have been used for aerosol studies including AOD retrieving were merely reasonable for regional study. They may not be so for local study. Moreover, the inevitable cloud contamination in SEA increases the level of difficulty of aerosol monitoring and research.

The impact of monsoons in SEA on aerosol transportation and distribution has not been reported thus far, nor has the dominant aerosol type in different seasonal monsoons. Determination of the dominant aerosol type for each region is important for modeling purposes because this information helps to characterize the effects of the aerosols on the atmosphere (Kaskaoutis *et al.*, 2007; Omar *et al.*, 2005; Lee *et al.*, 2010). Such information is highly useful in the radiative transfer and climate model.

Moreover, the aerosol system is dynamic and highly variable in time and space. Therefore, aerosol monitoring techniques at local scales need to be developed for potential application in global modeling analysis. An effective hourly AOD retrieval model to fill in the data gaps owing to cloud contamination has yet to be improved. Such a model is necessary for providing continuous AOD readings to monitor aerosol variation. It is also a tool to determine the influence to aerosol type distribution at different period of the year due to different seasonal monsoons.

Malaysia is a developing country in SEA in which anthropogenic activities arising from rapid population growth, industrialization, and urbanization occur. Rapid development of the country has increased air pollutants as a result of the increases in fossil-fuel combustion and biomass burning. However, continuous AOD readings in Malaysia is very limited due to lack of measuring station and inevitable cloud. Therefore, a model to retrieve continuous AOD is deemed needed in order to better monitor and understand the variation of aerosol in Malaysia.

1.3 Objectives

This research is designed to achieve the following objectives:

1) To investigate the seasonal change on various parameters such as fires activity, ground smoke emission, rainfall, typhoons, and outgoing longwave radiation (OLR) in northern and southern SEA between 2011 and 2013 and to establish a new algorithm for retrieving AOD in a specific local region.

2) To examine the established AOD prediction model relative to aerosol optical properties, the dominant aerosol type, and back-trajectory analysis.

3) To validate the established AOD prediction model and compare the predicted results with those of other AOD models and an independent source such as LIDAR.

4) To evaluate the applicability of the proposed algorithm at other wavelengths to predict AOD and to apply this model in the absence of measured AOD data.

1.4 Scope of the Study

In order to have better understanding the effect of environmental and meteorological factors in between northern and southern SEA, this study has qualitatively and semi-quantitatively investigated and identified the inter-relationships among fire activities, ground smoke emission, rainfall events, typhoon activities, and outgoing longwave radiation variation in SEA during seasonal monsoon changes between 2011 and 2013. In addition, the area where the cloud cover is more frequent in SEA will be determined for retrieving AOD in a specific local region based on outgoing longwave radiation.

This study mainly focuses on the development of new algorithms for retrieving continuous hourly AOD in Penang, Malaysia. The empirical model is focused only on multiple regression approaches and statistical analysis based on data collected from ground-based meteorological, air quality and AERONET level 2.0 data.

The database for ground-based meteorological data was obtained from Weather Underground (http://www.wunderground.com) for the site at Penang International Airport (5.30°N, 100.26°E). The air quality data was obtained from the Department of Environment, Malaysia (http://www.doe.gov.my/apims/index.php), for the USM site (source code: API CA0038).

Penang International Airport was selected for ground-based meteorological data because no visibility data could be obtained from the station nearby. In addition, the air pollution index (API) was used in this study rather than that of particulate matter (PM) because the former has free access in Malaysia.

The dominant temporal aerosol type in Penang was investigated based on the impacts caused by seasonal monsoon circulation. The method of scattering plots of

AOD against the Ångström exponent from AERONET data was used for aerosol classification. Seasonal dominant aerosol type and back-trajectories analysis are used to examine the performance of the established AOD prediction model. Finally, an independent source, i.e., light detection and ranging (LIDAR) data is used to check the validity of the AOD prediction model.

1.5 Novelty and Significance of the Study

The novel comparison of the inter-relationships among environmental and meteorological parameters between northern and southern SEA reveals that aerosol stations in southern SEA are lacking of continuous monitoring of aerosol variation. A new algorithm is established in this study for an hourly AOD prediction model to overcome the cloud contamination problem. The new algorithm efficiently retrieves hourly AOD data to create a supplemental dataset to compensate for the missing data in AERONET and satellite records owing mainly to the inevitable cloud cover. The diurnal AOD variation in Penang is thus obtained. Comparisons of various threshold criteria for aerosol classification have not been reported thus far, nor have comparisons of dominant aerosol type in Penang during different monsoon seasons. Additionally, the classification results provide helpful information to identify the LIDAR ratio. Such information reduces the uncertainty in the guessed value of the LIDAR ratio in the elastic LIDAR measurement.

1.6 Outline of the Thesis

This dissertation consists of six chapters, which are described in brief as follows:

Chapter 1 provides an overview of this study. Additionally, this chapter presents a brief background of atmospheric aerosols. A statement of the problem and the scope of the study are presented, as are the study's objectives, novelty and significance.

Chapter 2 provides an overview of the literature and related works on aerosol issues, and tasks, and explains the relevance of these studies to the present study. The computation of AOD is described in this chapter, and previous studies on AOD estimation methods, and related parameters of the AOD estimation model are being discussed. In addition, the aerosol classification method and several threshold criteria suggested in the literature are discussed.

Chapter 3 describes the study area, instrumentation data, and data pre-processing in detail. The research procedures and all techniques and methods used to achieve the research objectives are clearly described and explained.

Chapter 4 includes the analysis of all obtained results. First, the results of the impacts of seasonal monsoon circulation in SEA on fires, ground smoke emission, rainfall, typhoons, and OLR are discussed. The inter-relationships among these parameters are also discussed, and an interesting finding that influences aerosol research is identified. Next, the methods used to address this problem and the model performance results are mentioned. Finally, validation and examination of the proposed model are presented in this chapter.

Chapter 5 provides the application and comparison results of the proposed model. This chapter first discusses the applications of the proposed model in the absence of measured AOD data and subsequently compares the results with other methods such as LIDAR. The results from the proposed model are also compared with those of an existing model. Furthermore, the applicability of this proposed model is further examined.

Finally, the conclusions and suggestions for future research are presented in Chapter 6.

CHAPTER 2

LITERATURE REVIEW

2.0 Introduction

This chapter discusses previous studies conducted on aerosol issues and tasks in SEA including those on AOD estimation methods and the parameters related to the AOD estimation model. In addition, an aerosol classification method with several suggested threshold criteria is discussed.

2.1 Aerosol Study in SEA

The Intergovernmental Panel for Climate Change (IPCC, 2007, 2013) has reported that high uncertainty remains for aerosols under climate change owing to the complexity of the effects of direct and indirect radiation. This uncertainty is even more difficult to understand and to quantify over the Maritime Continent (MC) regions of SEA. The MC was first considered by Ramage (1968), who reported that the interaction of maritime (humid) and continental (dry) conditions is highly complicated owing to the geographical location and topography of SEA. MC regions are also known to host one of the most convoluted meteorological scales, which contributes to the complexity of aerosol systems throughout the world (Reid *et al.*, 2013). The complexity of the aerosol system in SEA is increased by human factors (Vayda, 2006), the unpredictability of which has been known to challenge scientists in many sectors, particularly in climate change. In addition, it remains unclear how anthropogenic activities vary with meteorological factors such as El Niño Southern Oscillation, seasonal migration of the Inter-tropical Convergence Zone and monsoonal season, Madden–Julian Oscillation, tropical cyclones and regional convection activities (Reid *et al.*, 2012).

According to the comprehensive overview of the SEA aerosol system by Reid *et al.* (2013), satellite data for aerosols, clouds, and precipitation are more divergent in SEA, than those in other regions of the world. This might be attributed to the complex relationship among geography and topography, socio-economics, the microphysical properties and atmospheric distribution of aerosols, and meteorological changes. In recent decade, most regions in SEA and the MC such as Indonesia, Malaysia, Singapore, and Indochina (Thailand, Myanmar, Vietnam, Laos, and Cambodia) have seen significant economic and population growth (Yuen and Kong, 2009). Consequently, the pollutants from biomass burning, industry, automobiles, and domestic cooking have degraded the air quality in SEA. Moreover, high demands for agricultural land for food productions such as palm oil, sugar cane, and rice can lead to intense deforestation, and the subsequent increase in open burning activities results in further deterioration of the air quality in SEA (Kim Oanh *et al.*, 2011). The annually occurring fire activity in SEA, one of the largest sources of anthropogenic aerosols and trace gases in the global atmosphere, attracts the attention of researchers because large amounts of biomass burning aerosols are injected into the atmosphere and contribute to changes in atmospheric composition and the climate (Reid *et al.*, 2013; Wang *et al.*, 2013a; Lin *et al.*, 2014c).

Smoke transportation and distribution in SEA depends on sea breezes, trade winds, typhoons, and the topography of the continental regions (Reid *et al.*, 2013; Wang *et al.*, 2013a). Transboundary aerosols are important issues because they can lead to regional visibility impairment and human health problems; in addition, they can affect the biosphere and direct and indirect climate forcing. Wang *et al.* (2013a) reported that although smoke is frequently transported to Peninsular Malaysia from Kalimantan, this activity occurs much less often when the southeastern trade wind shifts to the northeast. The primary contributor of transboundary aerosols in Peninsular Malaysia is smoke from local and Sumatra during the burning season.

2.2 Aerosol Issues Needed to be Addressed

Characterization of aerosol properties is necessary because of rapid growth in population and economic activities, both of which cause the anthropogenic aerosol emission rate to increase as a result of the increases in fossil-fuel combustion and biomass burning. These pollutants directly affect the climate and increase haze, fog, and clouds (Mukai *et al.*, 2006), which decreases visibility, particularly under high turbidity. According to Salinas *et al.* (2009), Asian sources are known to differ from those in Europe and North America. More absorbing soot and organic components are added to the Asia–Pacific atmosphere because of a substantially greater amount of coal and biomass burning emissions and long-range transport by wind (Lelieveld *et al.*, 2001; Huebert *et al.*, 2003; Seinfeld *et al.*, 2004).

Although studies on aerosols have reduced the uncertainties associated with climate change and have improved the understanding of aerosol characteristics, further improvement is still needed, particularly at the regional scale (Srivastava *et al.*, 2012). Large uncertainty in emission factors and insufficient long-term measurements of aerosol properties persist (Ganguly *et al.*, 2009). The aerosol system is dynamic and is influenced by a combination of various factors (Sherwood *et al.*, 2013; Tesfaye *et al.*, 2013); complex meteorological and environmental conditions for specific locations have increased the difficulty of research in this field (Reid *et al.*, 2013).

The trans-boundary long-range transport of aerosols may interact with local aerosols, particularly with cloud droplets, thus further modifying their microphysical properties. That is, precipitation processes and the radiative properties of aerosols are influenced by different aerosols which comprised of local and transported sources (Ichoku *et al.*, 2004; Lin *et al.*, 2013; Rosenfeld, 2007; Andreae and Rosenfeld, 2008). The origins of aerosol sources and their distribution in the atmosphere should be determined to understand why aerosol types differ among locations and why they are affected by environmental development, monsoons, southern oscillation variation, and seasonal change.

2.3 Aerosol Optical Depth (AOD)

The optical activity of aerosols in the atmospheric column can be summarized by AOD. AOD is a wavelength-dependent measure of the total extinction of incoming solar irradiance due to scattering and absorption by aerosols. That is, AOD is a measurement of the transparency of the atmosphere.

The atmosphere is composed of aerosols in addition to molecules of other gasses. Therefore, to obtain AOD, other light-scattering atmospheric constituents must be subtracted from the total optical depth (TOD). The TOD can be obtained using **Eq. (2.1)** according to the Beer–Lambert–Bouguer law:

$$V(\lambda) = V_0(\lambda) d^2 \, e^{-\tau(\lambda)_{TOD} * m}, \tag{2.1}$$

where V is the digital voltage measured at wavelength λ, V_0 is the extraterrestrial voltage, d is the ratio of the average to actual Earth–Sun distance, $\tau(\lambda)_{TOD}$ is the total optical depth, and m is the optical air mass (Holben *et al.*, 1998). The voltage (V)

measured by a sunphotometer is proportional to the spectral irradiance (*I*) reaching the instrument at the surface. The estimated top of the atmosphere spectral irradiance (I_o) in terms of voltage (V_o) is recorded by sun photometer measurements at Mauna Loa Observatory in Hawaii.

Specifically, computing the TOD accounts for the extinction of many other constituents such as water vapor, ozone, and mixed gasses. Rayleigh scattering of atmospheric molecules is an additional type of extinction. Finally, the remainder is the aerosol extinction, or AOD. The calculation is shown in **Eq. (2.2)**:

$$\tau(\lambda)_{Aerosol} = \tau(\lambda)_{TOD} - \tau(\lambda)_{water} - \tau(\lambda)_{Rayleigh} - \tau(\lambda)_{o_3} - \tau(\lambda)_{NO_2} - \tau(\lambda)_{CO_2} - \tau(\lambda)_{CH_4}. \tag{2.2}$$

Aerosol content data are important for improving the accuracies of weather forecast models and global environmental assessments. Therefore, AOD is the most comprehensive variable for assessing the aerosol load in the atmosphere and is widely utilized to estimate atmospheric air quality and to monitor air pollution conditions. AOD can also be used for atmospheric correction in space-borne remote sensing.

2.3.1 Estimation of AOD

Estimation of AOD is commonly performed by using satellite remote sensing images. Essentially, satellite images retrieve the attenuated radiation by aerosols from the reflections of the atmosphere and the ground surface. However, this task is challenging because the surface reflectance on the ground is always complex and undistinguishable. One of the most commonly used instruments for AOD estimation is the National Aeronautics and Space Administration (NASA) Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Terra and Aqua satellites launched respectively on December 17, 1999, and May 4, 2002 (Kaufman *et al.*, 2003; Remer *et al.*, 2005). MODIS has global coverage every two days, with a one-day or more frequent repeat at latitudes higher than 30° due to orbital convergence. This instrument uses the dense dark vegetation (DDV) method for retrieving AOD. The DDV algorithm has limited use on area in which the surface reflectance is very low such as that when more than 60% of the area is covered by vegetation. Therefore, this method might lead to bias when applied to areas with bright surface such as deserts and urban areas. Later works have attempted to improve the accuracy and limitations by considering band correlation based on the vegetation index and the scattering angle to increase the AOD retrieval accuracy (Levy *et al.*, 2007; Li *et al.*, 2007); the deep blue algorithm was developed specifically to overcome the bright surface problem (Hsu *et al.*, 2004; Hsu *et al.*, 2006).

In addition to the MODIS satellite, other satellites are used for AOD retrieval. For example, multiangular information is obtained from version 2 of the Along Track Scanning Radiometer (ATSR-2) (North, 2002) and the Multi-angle Imaging SpectroRadiometer (MISR) instrument on the Terra satellite (Diner *et al.*, 2005); polarization information is obtained from the Polarization and Directionality of Earth's Reflectances (POLDER) instrument onboard the Advanced Earth Observing Satellite 1 (ADEOS-1) (Deuzé *et al.*, 2001); and multitemporal information is obtained from the Total Ozone Mapping Spectrometer (TOMS) by the Nimbus-7 and Earth Probe Satellites (Torres *et al.*, 2002) and the Advanced Very High Resolution Radiometer (AVHRR) onboard National Oceanic and Atmospheric Administration's (NOAA's) Polar Orbiting Environmental Satellites (POES) (Li *et al.*, 2013).

With the large uncertainty in characterizing aerosol, a local study for each region is essential for verifying satellite imagery because the extraction of aerosol optical properties from remote sensing data has limited accuracy despite its capability for providing global-scale coverage of aerosol properties (Levy *et al.*, 2005; Tripathi *et al.*, 2005; Yoram *et al.*, 2002; Gupta *et al.*, 2013). Local studies on aerosol optical properties can be conducted by using reliable equipment such as sun-photometers or sky radiometers (Salinas *et al.*, 2009; Holben *et al.*, 1998; Remer *et al.*, 2008).

The accuracy of satellite-derived daily AOD is frequently assessed by comparing satellite-based AOD with data of AERONET, a network of ground-based sun photometers. AERONET is widely used for monitoring, investigating, and characterizing aerosol optical properties (Holben *et al.*, 1998). This network likewise provides a database for atmospheric correction and validation of satellite-based aerosol retrievals. Presently, AERONET includes more than 400 sites worldwide.

Satellites are commonly used for estimating AOD because this method is low in cost and has high spatial coverage to easily and conveniently study areas that are difficult to reach. However, satellites operating in sun-synchronous orbit are low in temporal resolution; therefore, only limited data are obtained from a specific location. To compensate, other models have been developed to provide a better understanding of quantifying aerosol characteristics. However, these methods are usually limited to space coverage. Therefore, ground- and space-based measurements are both necessary for performing reliable and comprehensive studies on atmospheric aerosols.

In fact, improvement is still needed in methods for obtaining AOD values in high temporal resolution. Although many ground-based instruments such as sun-photometers, radiometers, and LIDAR are able to provide very reasonable and accurate AOD values, the presence of clouds limits the ability of satellites or ground-based systems to measure and retrieve AOD. Consequently, current observational AOD datasets include gaps due to factors such as cloudiness. Because the existing knowledge of aerosol loading under cloudy skies is limited, it would be advantageous to use proxy datasets to fill these gaps. Thus, a robust predictor of AOD is needed.

Retrieving surface air quality in terms of PM from AOD estimated by satellite data is a popular topic because increased anthropogenic aerosol emission rate is a real air quality concern. Such studies have clearly proved that surface air quality is strongly correlated to columnar AOD. For a local region, continuous aerosol variation monitoring is important because the aerosol system is dynamic and may change significantly in a short time. However, this type of monitoring is not common practice.

2.3.2 Parameters Used in the AOD Algorithm Development

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

Previous studies indicate that AOD is proportional to air quality parameters such as PM with diameters less than 10 μm (PM_{10}) or 2.5 μm ($PM_{2.5}$) (Cordero *et al.*, 2012; Mielonen *et al.*, 2012; Mogo *et al.*, 2012; Müller *et al.*, 2012; Wang and Christopher, 2003a) but is inversely proportional to visibility (Vis) (Horvath, 1995; Li and Lu, 1997; Peppler *et al.*, 2000; Bäumer *et al.*, 2008; Singh and Dey, 2012; Lin *et al.*, 2014a), assuming most of the aerosol is at the surface. Surface visibility is a good indicator of columnar AOD (Hand *et al.*, 2004) because visibility normally decreases with an increase in AOD. This occurs likely because most aerosol particles are concentrated near the surface layer of the atmosphere or within the planetary boundary layer (PBL). However, both visibility and AOD could increase because of the varied distribution patterns of aerosol particles in the troposphere (Qiu and Yang, 2000).

A good relationship has been observed between AOD and $PM_{2.5}$ during summer (Cordero *et al.*, 2012) because the relationship between $PM_{2.5}$ and fine-mode AOD shows a high correlation, $R^2 = 0.6132$. A correlation coefficient of 0.70 was obtained between AOD from AERONET and PM_{10} from *in situ* measurements obtained during several periods between 2007 and 2009 (Müller *et al.*, 2012). Additionally, a correlation coefficient of 0.64 was calculated for AOD and PM_{10} by using the simple linear regression method (Mogo *et al.*, 2012). An additional study has likewise shown that a good agreement existed between AOD and ground-based measurements during the Russian wildfires in the summer of 2010 (Mielonen *et al.*, 2012).

The aforementioned studies have concluded that the spatial coverage of columnar AOD can be extended to the surface for air quality monitoring because of the high correlation between AOD and PM and visibility. However, other studies by Mahowald *et al.* (2007); Barladeanu *et al.* (2012); Chen *et al.* (2013); Toth *et al.* (2014) have reported that AOD is not always highly correlated to surface or horizontal measurements, particularly with the occurrence of an elevated layer of AOD from transported dust or biomass burning.

Relative humidity is an important factor that must be considered in global climate models (Ramachandran and Srivastava, 2013) and aerosol air-quality measurements (Ziemba *et al.*, 2013). Several studies have analyzed the association of air quality measurements and relative humidity. For example, aerosols have been shown to account for approximately 90% degraded visibility in non-foggy conditions, whereas relative humidity accounts for more than 80%; its relative contribution decreases rapidly because of the increase in fog droplets, thereby leading to a high rate of radiation attenuation (Singh and Dey, 2012). A reduction in the number of water-soluble particles in the atmosphere would lead to greater visibility than that with a high amount of water-soluble aerosols (de Meij *et al.*, 2012; Singh and Dey, 2012). However, visibility does not respond strongly to reduction in the mass concentration of insoluble particles. A subsequent study (Ramachandran and Srivastava, 2013) determined that AOD increases rapidly when the relative humidity exceeds 70% in continental clean and urban aerosol models; the value remains unchanged when the relative humidity exceeds 50% in maritime aerosols such as sea salt.

In addition, atmospheric aerosols that include hydrophilic particles can be influenced by atmospheric humidity, whereas those with hydrophobic particles cannot. Hydrophilic particles may adsorb water and consequently increase in size, which results in a higher scatter and absorption rate of light radiation. Thus, hydrophilic particles at a higher relative humidity environment may cause a higher extinction rate in the atmosphere and increase the AOD value (Tang, 1996; Song *et al.*, 2007; van Beelen *et al.*, 2013).

Other studies by Gao and Zha (2010); Gunaseelan *et al.* (2014); Liu *et al.* (2007); Smirnov *et al.* (2012); Zha *et al.* (2011); You *et al.* (2015) have revealed that besides aforementioned parameters, other meteorological parameters such as temperature, pressure, and wind can influence the accuracy of AOD prediction. They have also suggested that meteorological parameters contribute to AOD prediction accuracy to varying degrees during different seasons.

The presently used empirical model for AOD estimation does not consider multiple non-linear regression analysis. In practical use, simple linear and multiple linear regression analysis alone might not suitable for comprehensive and optimal empirical model analysis because aerosol variation is a complex system in the atmosphere. Although some studies have considered the empirical model for seasonal change, they are not used to investigate the effect of aerosol dominant type in that particular season. Additionally, the bias in previous studies on AOD estimation models has not been quantitatively investigated due to the lack of the vertical profile, which is used for identifying the distribution of aerosols in the atmosphere.

2.4 Aerosol Optical Properties Adopted for Aerosol Classification

Aerosol classification studies have been extensively analyzed by using AERONET datasets applied in different locations such as at the Persian Gulf (Smirnov *et al.*, 2002a), Brazil, Italy, Nauru, Saudi Arabia (Kaskaoutis *et al.*, 2007), Spain (Toledano *et al.*, 2007), Singapore (Salinas *et al.*, 2009), Kuching (Jalal *et al.*, 2012), and several ocean regions (Smirnov *et al.*, 2011a); the Multi Filter Rotating Shadowband Radiometer was applied in the central Mediterranean (Pace *et al.*, 2006). Generally, researchers draw scatter plots for AOD_500/AOD_440 with

Ångström$_{440–870}$ to identify the aerosol type. Ångström$_{440–870}$ is used because this wavelength is typical for determining aerosol size from spectral AOD (Eck *et al.*, 1999). Values of AOD at 500 nm are typically used to determine the relationship of Ångström$_{440–870}$ because they are indicative of turbidity conditions and aerosol type (Cachorro *et al.*, 2001; Smirnov *et al.*, 2002b; Kaskaoutis *et al.*, 2007; Pace *et al.*, 2006; Salinas *et al.*, 2009; Smirnov *et al.*, 2003). On the contrary, AOD_440 with Ångström$_{440–870}$ is also commonly used for the same purpose.

At different locations, different data sources generally have very different patterns in Ångström–AOD (α–τ) scatter plots in the literature, therefore causing difficulty in interpretation; this finding has also been reported by Holben *et al.* (2001). In current study will use the threshold criteria suggested by other researchers for their aerosol classification methods. Generally, aerosol classes are divided into the following classes: desert dust (DA), maritime (MA), continental/urban/industrial (UIA), biomass burning (BMA), and mixed aerosols (MIXA) that are difficult to clearly distinguish (Ichoku *et al.*, 2004). The following sub-section describes these criteria.

2.4.1 Threshold Values from α–τ Scatter Plots for Aerosol Classification

Kaskaoutis *et al.* (2007) used threshold values of AOD_500 and Ångström$_{440–870}$, as did Pace *et al.* (2006). According to their scheme suggestion, i) clean maritime background conditions, characteristic of remote oceanic areas, should have AOD <0.06 and Ångström <1.3; ii) AOD >0.1, Ångström >1.5 correspond to BMA and UIA; iii) AOD >0.15, Ångström <0.5 indicate DA; and iv) the threshold of AOD and Ångström that do not belong to any of the above groups are characterized as MIXA. Moreover, Smirnov *et al.* (2002b, 2003) suggested a threshold of AOD <0.15 and

Ångström <1.0 for MA; AOD >0.4, Ångström >1.5 for BMA; and AOD >0.2, Ångström <0.7 for DA.

According to Salinas *et al.* (2009), when Ångström <1, the size distributions are dominated by coarse-mode particles with radii >0.5 μm suspected as dust and sea salt, whereas values of Ångström >1 represent dominance by fine-mode particles with radii <0.5 μm, which are typically associated with urban or industrial pollutants and biomass burning (Kaufman, 1993; Gobbi *et al.*, 2007; Salinas *et al.*, 2009; Kaskaoutis *et al.*, 2007; Schuster *et al.*, 2006; Eck *et al.*, 1999). Their study determined that the threshold for dust particles should be AOD >0.2 associated with Ångström <1.0 because these particles tend to have larger sizes combined with large AOD (Salinas *et al.*, 2009). In addition, they also found that MA should have values of AOD <0.2 with Ångström <1.0, although AOD should be <0.15 (Smirnov *et al.*, 2003). Moreover, values of AOD between 0.2 and 0.4 with Ångström numbers >1.0 but <2.0 is indicative of urban or industrial pollutants from fossil fuels with free dust and sea-salt aerosols. Little evidence was obtained for aerosols resulting from trans-boundary BMA sources from Indonesia based on AOD >0.8 and Ångström >1.0. Although Salinas *et al.* (2009) used AOD_500 with Ångström at a wavelength of 500 nm, Ångström$_{440–870}$ is also acceptable for this threshold range because that at 500 nm is included in the interpolation results from Ångström$_{440–870}$.

A study by Jalal *et al.* (2012), who referred mainly to Salinas *et al.* (2009), proposed another threshold result for AOD_440 and Ångström$_{440–870}$. They suggested that values of AOD <0.3 and Ångström between 0.5 and 1.7 represent MA; values of AOD between 0.2 and 0.4 are associated with Ångström >1.0 and are indicative of UIA; BMA also has Ångström >1.0 with AOD >0.7; and the value of Ångström for coarse dust particles is <1.0 with AOD >0.4. Moreover, Toledano *et al.* (2007) also extensively studied the relationship of AOD with Ångström to determine aerosol type. They proposed a scatter plot of AOD at a wavelength of 440 nm and Ångström$_{440–870}$ to define MA at a value of AOD <0.2 and Ångström between 0 and 2.0. UIA can also be determined at AOD between 0.2 and 0.35 with Ångström >1.05, whereas the values of AOD and Ångström for BMA are >0.35 and >1.4, respectively. However, DA should use AOD at a wavelength of 870 nm with values >0.11 and Ångström <1.05.

The threshold criteria for aerosol classification determined by Smirnov *et al.* (2002b, 2003), Pace *et al.* (2006), Kaskaoutis *et al.* (2007), Toledano *et al.* (2007), Salinas *et al.* (2009), and Jalal *et al.* (2012) are summarized in **Table 2.1**.

2.5 Seasonal Monsoon Circulation in SEA

Monsoon circulation is a massive, seasonally changing of wind direction due to temperature different between land and ocean. Normally during summer period, the land is warmer than the ocean. This causes air to rise over the land and air to blow in from the ocean to fill the void left by the air that rose. Therefore, southwesterly winds flow is observed in SEA. Rising air leads to cloud formation and precipitation over land area. The land may stay warmer than the ocean for most of the time during summer and the ocean is a constant source of moisture into land region, causing rain events to occur more frequently.

On the other hand, air flow pattern and weather phenomena during winter is opposite to what happen in summer period because ocean is warmer than land during winter. A surface wind flows northeasterly to southwesterly in the SEA from winter to summer. During winter time, the atmosphere over SEA is drier. In addition, there are two transition periods between the southwest (summer) and northeast (winter)