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Overview of atmospheric aerosol studies in Malaysia: known and unknown

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Abstract

Atmospheric aerosols particularly those originated from anthropogenic sources can affect human health, air quality and the regional climate system of Southeast Asia (SEA). Population growth, and rapid urbanization associated with economic development in the SEA countries including Malaysia have resulted in high aerosol concentrations. Moreover, transboundary smoke plumes add more aerosols to the atmosphere in Malaysia. Nevertheless, the aerosol monitoring networks and/or field studies and research campaigns investigating the various aerosol properties are not so widespread over Malaysia. In the present work, we summarize and discuss the results of previous studies that investigated the aerosol properties over Malaysia by means of various instrumentation and techniques, focusing on the use of remote sensing data to examine atmospheric aerosols. Furthermore, we identify gaps in this research field and recommend further studies to bridge these knowledge gaps. More specifically gaps are identified in (i) monitoring aerosol loading and composition over urban areas, (ii) examining the influence of dust, (iii) assessing radiative effects of aerosols, (iv) measuring and modelling fine particles and (v) quantifying the contribution of long range transport of aerosols. Such studies are crucial for understanding the optical, physical and chemical properties of aerosols and their spatio-temporal characteristics over the region, which are useful for modelling and prediction of aerosols' effects on air quality and climate system.

Keywords: Aerosols, Malaysia, remote sensing, particulate matters, climate change, atmosphere

1. Introduction

Increased concentrations of atmospheric aerosols, especially those originated from biomass burning, fossil-fuel combustion, industrial emissions, etc. have become the main climate issue over Southeast Asia (Carmichael et al., 2009; Reid et al., 2013; Lin et al., 2013; Lin et al., 2014). Depending on their type and composition, aerosols scatter and/or absorb the solar light (direct radiative effect) hence affecting the amount and spectral distribution of the incoming solar radiation and the Earth's radiation-energy budget (e.g., Haywood and Boucher, 2000; Ramanathan et al., 2001; Kanniah et al., 2010). Indirectly, aerosols affect the

radiation budget and climate through their impact on cloud microphysical processes and cloud condensation nuclei (e.g., Lohman and Feister, 2005; Rosenfeld et al., 2008). Aerosols also render harmful effects on human health, mostly lung and eye diseases, as a major component of the air pollution (PM_{10} and $PM_{2.5}$) and degradation of visibility (Poschl, 2005; Li et al., 2010; Reid et al., 2013). The aerosol effects on climate, solar radiation, clouds and human health are strongly dependent on their optical, physical and chemical characteristics including the columnar optical depth, particles concentration, size distribution, single scattering albedo, refractive index, and chemical composition, which are defined from aerosol source, type and mixing processes in the atmosphere (e.g., NASA Facts, 2005; Kahn et al., 2009; Rosenfeld et al., 2014). Due to their various sources, high spatio-temporal variability and complex optical properties, aerosols remain as one of the largest uncertainties in the Earth's climate system (IPCC, 2007, 2013). Therefore, aerosol properties are crucial aspects to be investigated in order to understand their influence on radiation budget, climate, air quality, ecosystems and human health.

Southeast Asia (SEA) has been recognized as one of the most vulnerable regions worldwide in respect to climate change (IPCC, 2007), due to the complex aerosol field as a result of the diverse land surface and intricate meteorology and hydrology, rendering atmospheric observations, analysis and future predictions over this region a challenging task (Reid et al., 2013). Population growth, rapid urbanization and development in SEA countries (including Malaysia) have resulted in high aerosol concentrations (Afroz et al., 2003; Abdul Rahman et al., 2011). Malaysia receives considerable amount of pollutants from trans-boundary sources (mostly from biomass burning in Indonesia and Indochina) as it lies in the main pathway of the Southeast Asian pollution outflow (Lawrence and Lelieveld, 2010; Reid et al., 2013; Wang et al., 2013), which contributes significantly to the local aerosol and pollutant emissions. Transboundary aerosols constitute an important research topic 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 (Kanniah et al., 2012). Consequently, Malaysia is ranked as the 117th worst country among 180 nations worldwide in terms of air quality (EPI, 2016).

Nevertheless, the aerosol monitoring networks and/or field studies and research campaigns investigating the various aerosol properties are not so widespread over Malaysia (Kanniah et al., 2015). Therefore, in the present work, we summarize and discuss the results of previous studies that examined the aerosol properties over Malaysia by means of various

instrumentation and techniques, focusing on the use of remote sensing data for atmospheric aerosols monitoring. Furthermore, we identify gaps in this research field and recommend further studies to bridge these knowledge gaps which are crucial for understanding the optical, physical and chemical properties of aerosols and their spatio-temporal characteristics over the region. Such studies are also useful for atmospheric aerosols modelling and prediction of aerosols' effects on air quality and climate system. The following section (Section 2) describes the methods (ground based and remote sensing) used for aerosol monitoring over Malaysia, while Section 3 reviews results from previous studies conducted over Malaysia using ground-based and remote-sensing monitoring techniques. Section 4 highlights the research gaps in atmospheric aerosol studies and the last section summarizes and concludes the paper.

2. Monitoring of aerosols over Malaysia

2.1 Ground-based aerosol monitoring

The near-surface atmospheric aerosols are regularly monitored in Malaysia using continuous particulate matter (PM) monitoring instrument such as Met-One Beta Attenuation Method (BAM) (Azmi et al. 2010; Dominick et al. 2012) and manual 24-h monitoring system such as High Volume Air Samplers (HVAS) (MetMalaysia, 2015). Two main air-pollution monitoring networks have been established by the Department of Environment, Malaysia (DOE) and Malaysian Meteorology Services (MetMalaysia). DOE has awarded a 20-years concession to a private company, known as Alam Sekitar Malaysia Sdn Bhd (ASMA), for the establishment of environmental data collection, processing, interpretation, analysis and distribution (DOE, 2015). 52 Continuous Air Quality Monitoring (CAQM) stations (Figure 1) and 19 Manual Air Quality Monitoring (MAQM) stations have been installed via this network. The monitoring stations have been strategically located at residential, industrial, busy-traffic and rural areas by taking into consideration past and current monitoring, representativeness, accessibility, availability of support services (power, telephone line etc.), security and effects of topography (DOE, 2012). These stations measure size segregated aerosol concentrations ($PM_{2.5}$, PM_{10}) and other pollution gases, such as Sulphur Dioxide (SO_2), Nitrogen Oxides (Nitric Oxide (NO) and Nitrogen Dioxide (NO_2)), Carbon Monoxide (CO), Ozone (O_3), Methane (CH_4), and Non-Methane Hydrocarbon (NMHC). Instruments from the Teledyne Technologies Inc., USA, such as the Teledyne API Model 100A/100E, the Teledyne API Model 300/300E, the Teledyne API Model 400/400E and the Teledyne API

Model 200A/200E are used to monitor SO₂, CO, O₃ and Nitrogen Oxides respectively (ASMA, 2007). The Teledyne API Model 200A/200E is also employed to monitor NO and NO₂ parameters. Meanwhile, the gases (SO₂, NO_x and O₃) are determined using the UV fluorescence method, chemiluminescence detection method and UV absorption (Beer Lambert) method respectively (ASMA, 2007). The CAQM stations are also equipped with instruments from Met One Instrument, Inc., USA, namely Met One 010C sensor, Met One 020C sensor, Met One 062 and Met One 083D sensors to measure wind speed, wind direction, ambient temperature and relative humidity respectively (ASMA, 2007). Measurement of UV radiation is important due to its capability to affect the human health and its influence towards the formation of surface O₃. Therefore, UV radiation is measured at CAQM stations and it is determined by UV radiometer (Kipp & Zonen) (ASMA, 2007). Especially in the CAQM stations, HVAS are also used to measure the Total Suspended Particulates (TSP). Several heavy metals such as Lead, Mercury, Iron, Nickel, Copper etc. on filter papers equipped with HVAS are analysed in the laboratory. These parameters are measured once in every six days, manually collected and delivered for analysis. The data from the CAQM stations are provided by ASMA on a monthly basis (DOE, 2012).

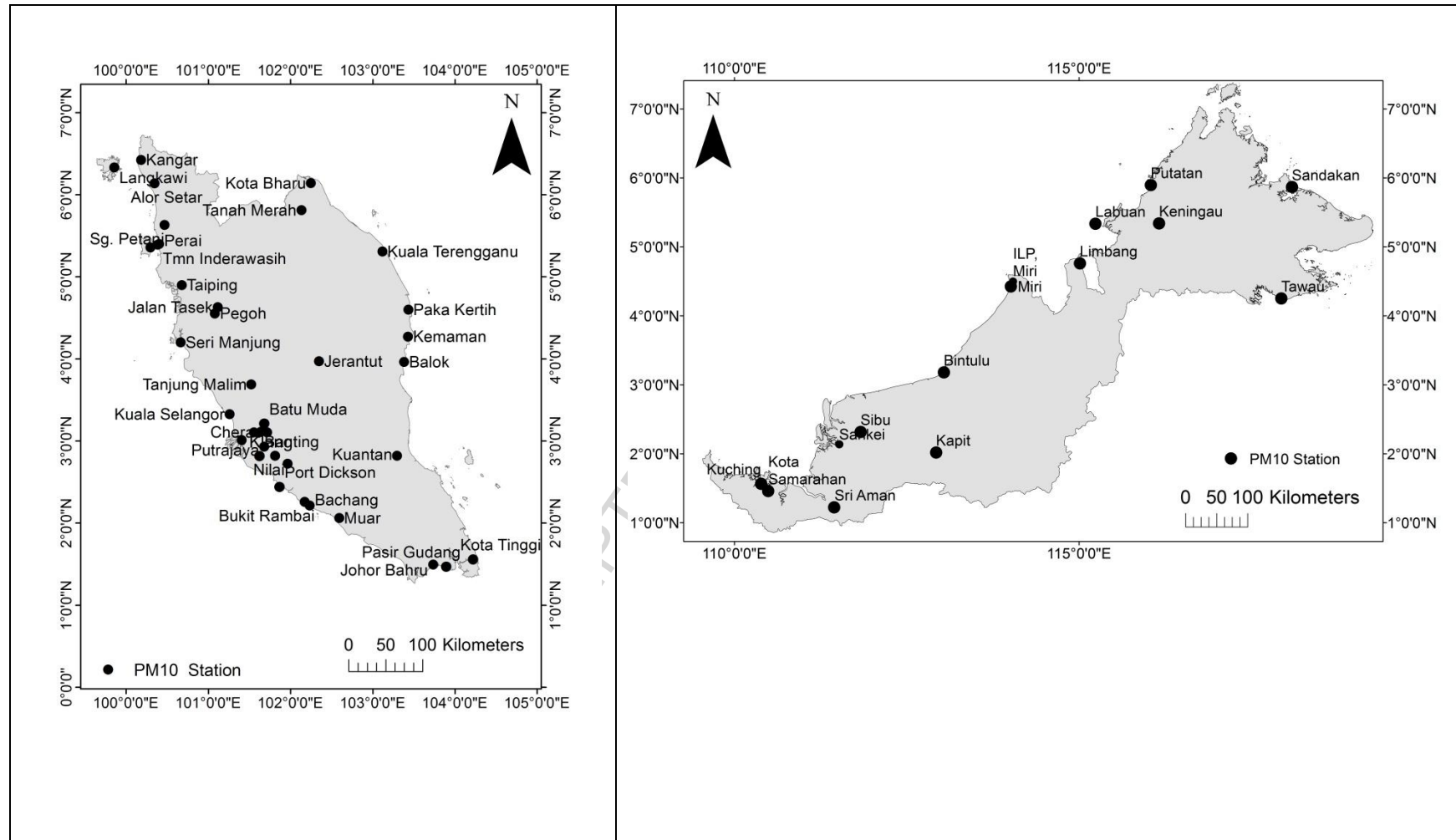


Figure 1: Locations of Continuous Air Quality Monitoring (CAQM) stations in Peninsular Malaysia (left) and in East Malaysia (right). [Source: Environmental Quality Report, 2010 by DOE]

On the other hand air-pollution monitoring network established by the Malaysian Meteorological Services (MetMalaysia) comprises a total of 22 stations (Figure 2) that are distributed over Peninsular and East Malaysia. Most of the stations are located at some distance away from the urban centres, so that only ambient conditions are monitored. They are co-located with meteorological/climatological stations for simultaneous and continuous recordings of both air pollution and meteorological data (MetMalaysia, 2015). Parameters such as TSP, PM₁₀, tropospheric ozone and reactive gases (i.e. surface O₃, CO, Volatile Organic Compounds (VOCs) , total oxidised nitrogen compounds (NO_y), and SO₂ are collected from these stations. It should be noted here that the VOCs are measured as non-methane hydrocarbons. TSP is measured at 14 stations by HVAS, while PM₁₀ is measured by High Volume PM₁₀ Sampler (HVPM10S) at 9 stations (MetMalaysia, 2015).

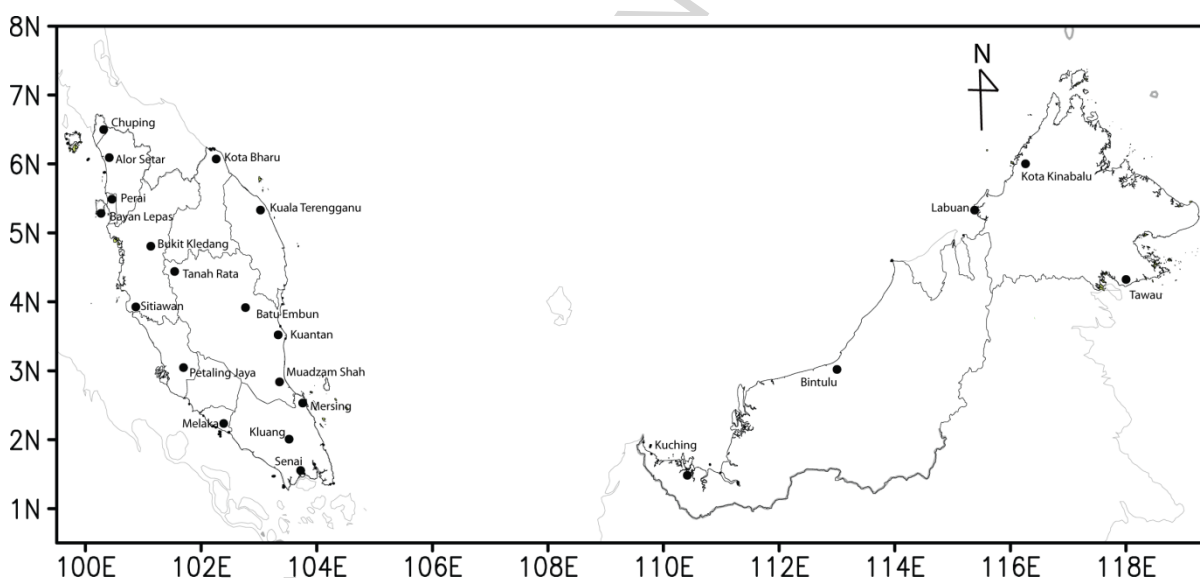


Figure 2: Distribution of the air-pollution monitoring stations of Malaysian Meteorological Services (MetMalaysia). [Source: MetMalaysia, 2012]

It should be noted that HVAS measures TSP with diameter less than 100 μm , while the HVPM10S measures only particulates that are below 10 μm in diameter. The total ozone column, as well as concentrations of tropospheric ozone, and the vertical ozone profile are also monitored at several locations in Malaysia. The columnar ozone is automatically monitored at Petaling Jaya (industrial area - Figure 2) using Brewer spectrophotometer, while the vertical ozone profile is measured at Kuala Lumpur International Airport (KLIA) by means of ozonesondes released at the beginning and middle of each month (Boynard et al., 2009). The reactive gases, SO₂ and NO₂ are monitored at Petaling Jaya and Tanah Rata (hilly site) using passive samplers, an instrument that collects samples without using any pump (WMO- <ftp://ftp.wmo.int/Documents/PublicWeb/arep/gaw/gaw122.pdf>). **The air-pollution**

monitoring networks operated by DOE and MetMalaysia are crucial for air-quality assessment over major cities in Malaysia and have been used in several studies as described in Section 3.

2.2 WMO Global Atmospheric Watch (GAW) Network of stations

The Global Atmospheric Watch (GAW) consists of a worldwide network of aerosol monitoring stations (30 global, 400 regional and many national stations) coordinated by the World Meteorological Organisation (WMO). The data collected are particularly essential in understanding the relationship between changing atmospheric composition and human-induced changes in regional and global climate.

In Malaysia, there are one global (Danum Valley, Sabah) and two regional (Tanah Rata in Cameron Highlands and Petaling Jaya) GAW stations established and operated by MetMalaysia. The global station at Danum valley, Sabah (Figure 3) is aimed at monitoring the background concentrations of atmospheric parameters, studying the transboundary transport of pollutants and the ability of tropical forests to remove atmospheric pollutants primarily by uptake via leaf stomata, though some gases are removed by the plant surface; furthermore, trees may also remove pollution by intercepting airborne particles (Nowak et al., 2006). Meanwhile, the GAW station at Petaling Jaya monitors the urban air quality and meteorology for forecasting urban air-pollution (Mohamad, 2011; Jahaya, 2013), in addition to TSP measurements using the Sierra Andersen High Volume Air Sampler. Furthermore, PM₁₀ are collected on Teflon filters using the Ecotech SolarVol Particulate Sampler, which operates at a much lower flow rate of 4 litres per minute. The concentration of PM₁₀ is determined in the same manner as with the HVAS. The samples are then sent to the Chemistry department in Malaysia for analysis to determine its chemical compositions. The GAW station at the Cameron Highlands hilly site (1545 m) is characterised as background aerosol site and measures the amount and chemical compositions of aerosols, rainfall, surface ozone and other reactive gases.

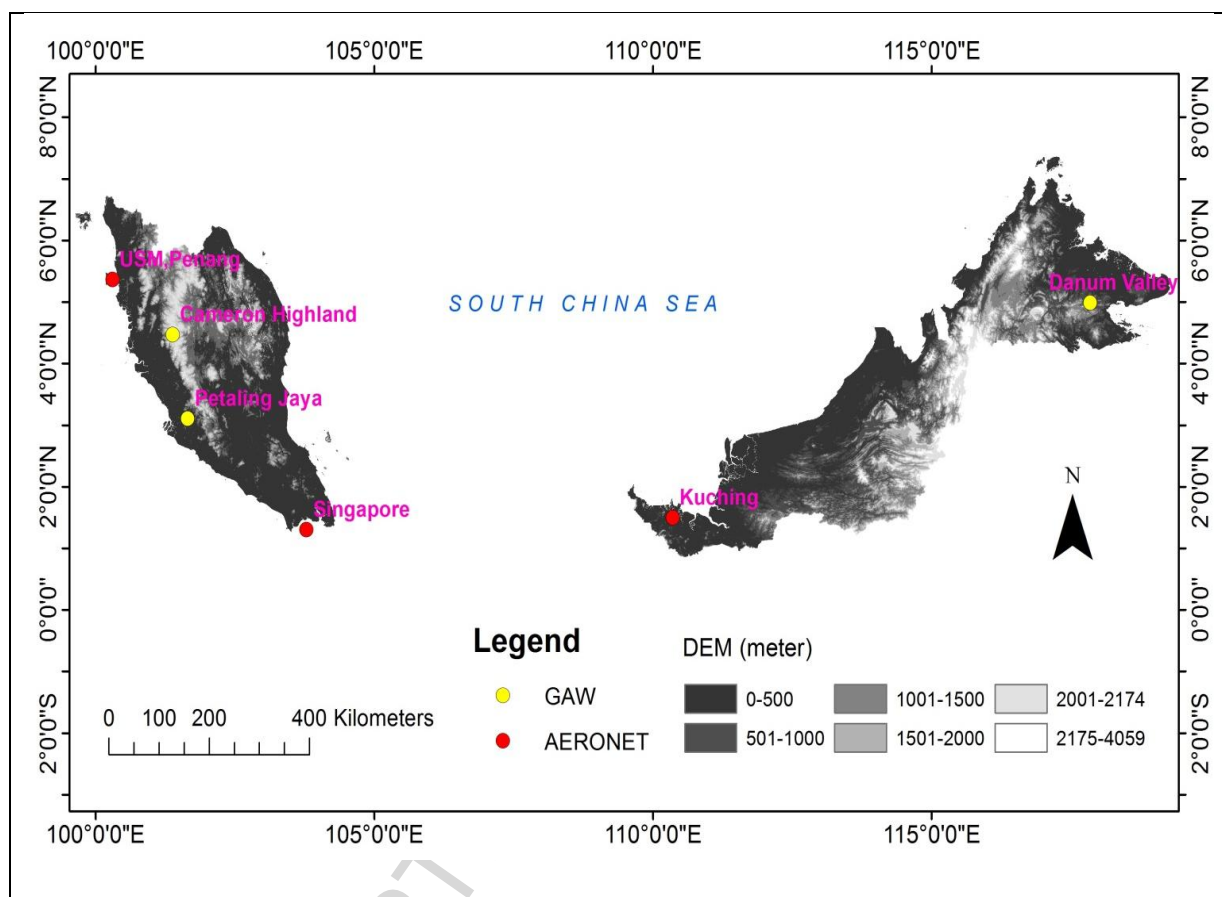


Figure 3: Locations of World Meteorological Organization Global Atmospheric Watch (GAW) and AERONET stations over Malaysia. The AERONET site in Singapore is also shown. The stations are overlaid onto a Digital Elevation Model generated from Shuttle Radar Topography Mission (SRTM).

2.3 AERONET stations

Columnar aerosol properties i.e. total amount and characteristics are critical for examining their impact on solar radiation, cloud condensation processes and climate change (e.g. Guan et al., 2010; Dumka et al., 2015) because the flow of aerosols can occur from the higher troposphere to near surface (Gogoi et al., 2011; Srivastava et al., 2012). In this context, Aerosol Robotic Network (AERONET) stations and satellite remote sensing can be alternative approaches to study the temporal and spatial distribution of aerosol properties from local to global scales. **The columnar aerosol properties** via the AERONET program **are available** over Malaysia since 2011 (AERONET, 2012). AERONET

(<http://aeronet.gsfc.nasa.gov/>), is a ground-based remote sensing aerosol network program established by the *National Aeronautics and Space Administration* (NASA) (Holben et al., 1998), and provides long-term and continuous measurements of aerosol optical, microphysical and radiative properties covering parameters such as **aerosol optical depth (AOD)**, **Angstrom exponent**, solar flux and aerosol inversion products (i.e. single-scattering albedo, size distribution, fine-mode fraction, phase function, refractive index, etc.).

In Malaysia, currently there are only two AERONET stations in operation (Figure 3). Kuching and Universiti Sains Malaysia (USM), Penang stations were established in August and November 2011, respectively. Meanwhile, the Tahir station (located near to USM) operated only for a short period between June and October 2012. This site was mainly established for a field campaign (Distributed Regional Aerosol Gridded Observation Networks-DRAGON Southeast Asia) and the station ceased after the campaign. The AERONET stations use the Cimel sun photometer to measure aerosol and radiation parameters. The Kuching AERONET station is installed at Kuching International airport, Sarawak (on top of Kuching Meteorological Station). AOD and aerosol inversion products at levels 1.0, 1.5 and 2.0 from Kuching AERONET station are available at http://aeronet.gsfc.nasa.gov/new_web/photo_db/Kuching.html. In USM Penang station, the sun photometer was installed in the main campus of Universiti Sains Malaysia (USM), which is located in Penang Island in the northwest coast of Peninsular Malaysia (Figure 3). Data from the AERONET stations in Malaysia have been recently used to study the aerosol properties as discussed in Section 3. Nevertheless, these stations are spatially limited and incapable to monitor regional air quality and climate system. Alternatively, satellite remote sensing data provide synoptic view to understand the aerosol characteristics and their impact as described in the following sub-sections.

2.4 Space-borne remote sensing of aerosols

Due to their large spatio-temporal variability and the great effects of aerosols on regional and global climate, many satellite sensors provide observations and retrievals that can be used for studying the aerosol properties. Details on the main satellite sensors, aerosol products, their spatial resolutions and accuracy are summarized in Table 1. The first operated sensor that was used for aerosol studies was the Advanced Very High Resolution Radiometer (AVHRR), which employs visible and shortwave-infrared channels of the electro-magnetic spectrum to retrieve AOD and Ångström exponent, respectively but only over the oceanic surfaces

(Stowe et al., 1997; Mishchenko et al., 1999). Other sensors, such as Total Ozone Mapping Spectrometer (TOMS) on-board Meteor-3, Nimbus-7 and Earth Probe satellites, were used for monitoring biomass-burning smoke and dust plumes due to their specific sensitivity on UV channels, where dust and smoke exhibit significant absorption of the solar light (Kirchstetter et al., 2004). Further discrimination between mineral and carbonaceous aerosols was made possible by Ozone Monitoring Instrument (OMI), which provides higher spatial resolution and more spectral channels (740 wavelengths) compared to TOMS (Curier et al., 2007). Aerosol retrievals over both ocean and land (including bright desert and sun-glint regions) have been made possible by Multi-angle Imaging Spectro-Radiometer (MISR) instrument on board Terra satellite, which has a wide range of along-track view angles (Kahn et al., 2009). MISR provides specific information on particle size and shape of aerosols and these properties can be used to assess the aerosol types (Kahn et al., 2009). Polarization and Directionality of the Earth's Reflectance (POLDER) sensor with multidirectional and polarization measurements at eight spectral bands in the visible and NIR spectrum, enables the derivation of the aerosol size, scattering phase function and AOD that can be used for better separation of the contribution of the fine-mode anthropogenic aerosols (di Girolamo et al., 2004; Kokhavosky et al., 2007). For aerosols over land, Moderate Resolution Imaging Spectroradiometer (MODIS) is one of the most widely used satellite sensors in atmospheric aerosol studies on board Terra and Aqua satellites. The "Deep Blue" algorithm applied to MODIS data has the capability to retrieve aerosol properties over bright land surfaces such as arid, semiarid and urban areas which are very bright in the red to the near infrared spectra region, but relatively darker in the blue wavelength region (Hsu et al., 2013). The long-term aerosol data availability (since February 2000), high accuracy of the AOD retrievals over land (i.e. $\pm 0.05 \cdot \text{AOD}$ to $\pm 0.15 \cdot \text{AOD}$ under clear and moderately cloudy conditions respectively) and the twice (morning and afternoon) daily coverage of the Earth (Remer et al., 2008) made this sensor very popular among the atmospheric scientists.

MODIS aerosol products have been used in many studies covering radiation and climate (e.g., Yu et al., 2006; Yang et al., 2012; Feng and Christopher, 2013), air quality (e.g. Chu et al., 2003; Al-Saadi et al., 2005; van Donkelaar, 2010; Chudnovsky et al., 2014) and the spatio-temporal patterns of aerosols over various geographical regions (e.g., Prasad and Singh, 2007; Kosmopoulos et al., 2008; Song et al., 2008; Alam et al., 2011; Marey et al., 2011; Kittaka et al., 2011; Xu et al., 2013; Kanniah et al., 2014a). Furthermore, aerosol size-related products derived by MODIS, such as fine-mode fraction (FMF) and Ångström

Exponent (AE) have also been used to discriminate different types of aerosols over the globe (Barnaba and Gobbi, 2004; Kaskaoutis et al., 2007, 2012; Kim et al., 2007; Santese et al., 2007; Deng et al., 2012). Recently MODIS offers AOD product at 3-km spatial resolution worldwide (Remer et al., 2013) which is critical to study the urban atmosphere. This product has been used for estimating PM_{2.5} in Beijing (Xie et al., 2015) although some studies have concluded that this product has more noise (urban area) and, therefore, it should be used cautiously (Munchak et al., 2013; Remer et al., 2013).

The Along Track Scanner Radiometer (ATSR) and its successor Advanced Along Track Scanner Radiometer (AATSR) flown on Envisat platform have dual view and can provide more accurate aerosol properties because they eliminate the influence of land reflectance on top of the radiation. Since AATSR has dual view and its applications have excellent accuracy after comparison with AERONET it is used in various land surfaces including the desert environments (North et al., 2009). Although the AATSR mission ended in 2012, its successor Sea and Land Surface Temperature Radiometer on board of Sentinel 3 mission is a conical scanning geometry that provides two views of the Earth at different view angles to enable accurate atmospheric correction (North et al., 2009).

MERIS is another sensor on board Envisat-1 satellite that provides aerosol data over land and it showed satisfactory accuracy with ground-based data (Vidot et al., 2008; Kaskaoutis et al., 2010). The capability of MERIS to provide better estimation of particulate matter is tested by modification of its algorithm (Kaskaoutis et al., 2010; Rohen et al., 2011) and by synergising MERIS data with AATSR (North et al., 2009). The synergised MERIS/AATSR AOD data has been used to estimate PM₁₀ over the Athens urban area by Benas et al. (2013). Similar to AATSR even though MERIS's mission ended in 2012, its capability will be continued by Ocean and Land Colour Instrument (OLCI) on board of Sentinel-3 (North et al., 2009). The latest instrument, which can provide aerosol and cloud properties, is the Visible Infrared Imager Radiometer Suite (VIIRS) on board of Suomi National Polar-orbiting Partnership (NPP) satellite. However, currently most of VIIRS data are still in the evaluation stage (Hillger et al., 2013).

Despite the availability of various satellite sensors providing aerosol products, rather few aerosol studies have been conducted over Malaysia using remotely sensed data/techniques due to frequent cloud coverage. These studies were conducted using mostly AVHRR,

MODIS, Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY), Atmospheric Infrared Sounder (AIRS) and MERIS data. The following sections highlight the aerosol studies over Malaysia using satellite remote sensing techniques, their limitations, our current knowledge and understanding on aerosol characteristics, future perspectives and studies needed.

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Table 1: Characteristics of different satellite sensors used for aerosol retrievals.

Satellite sensor	Operational period	Aerosol products	Resolutions	Wavelengths used	Accuracy	Remarks	Reference
AVHRR	Since 1979	Aerosol Optical Depth (AOD), Angstrom exponent	Daily, weekly, monthly, 1 degree	Visible Shortwave infrared	AOD (10%) for single channel AOD (3.6%) for two channel	Providing aerosol products over worlds oceanic surfaces only since 1988	Stowe et al. (1997); Mishchenko et al. (1999) Ignatov et al. (2004)
TOMS	1979-1993 and 1996-2005	Ultra Violet (UV)-absorbing aerosol index, UV Aerosol Optical Depth (AOD)	Daily, Monthly, 1 degree	Ultraviolet channels (331 and 360 nm)	AOD (20 ~ 30%)	Could differentiate biomass-burning smoke and dust	Torres et al. (2002)
OMI	Since 2004	UV Aerosol Index, Aerosol Absorption Optical Depth, Aerosol Extinction Optical Depth (AOD) and Single Scattering Albedo	Daily, monthly, 13 × 12 km ² , 13 × 24 km ² , 13 × 48 km ²	Includes 330 to 380 nm channels	AOD (30%)	Can detect minerals and carbonaceous aerosols	Torres et al. (2007)
SeaWIFS	1997 – 2010	aerosol optical thickness and fine mode fractional volume, Angstrom Exponent	Daily and monthly 13.5x13.5 km at the center of the swath	510 nm, 670 nm, and 865 nm	AOD (~5 -10%)	Uses Deep Blue algorithm to retrieve AOD and size parameters over ocean, land and dessert	Sayer et al. (2012) Hsu et al. (2013)

MODIS	Since 2000 (Terra) Since 2002 (Aqua)	AOD, Angstrom exponent, Fine mode fraction	Daily, 8days and monthly 10 km, 1 degree	550 nm	AOD (~5 -15%)	Widely used to study air pollution, climate and health impacts of aerosols	Remer et al. (2005)
MISR	Since 2000	AOD, Angstrom exponent, single scattering albedo, non-spherical fraction	1.1km (non-red band) 275m (red band)	555 nm	AOD (10 ~20%)	Wide range of along track view angles (one at nadir and eight symmetrical views at 26.1, 45.6, 60.6 and 70.5 degrees forward and aft of the nadir). Enables aerosol retrieval over ocean, land, bright desert and sun-glint regions Yields information on particle size and shapes which can be used to identify aerosol types	Kahn et al. (2005)
POLDER	1996 -1997 2002 – 2003 – 2004 – present	AOD, Angstrom exponent, non-spherical fraction, scattering phase function	Daily 18.5 km	0.443, 0.490, 0.565, 0.665, 0.765, 0.865 and 0.910 μ m	AOD (20 ~30%) Angstrom exponent correlated well with AERONET, but with underestimation of 30%	Provides multi-directional and polarization measurements and can separate atmospheric contribution from surface contribution	Herman et al. (1997)

SCIAMACHY, GOME 1, GOME 2	2002-2012 1995-2003 2010-present	UV-absorbing Aerosol Index (AI)	Daily, monthly 60 x 30 km 320 x 40 km 80 x 40 km	340 and 380 nm	AI (~40%)	Able to detect and discriminate desert dust and biomass burning aerosols	Graaf and Stammes (2005)
MERIS	2002-2012	AOD, Angström exponent	Daily, 1.2km	15 channels between 0.39-1.04 μm	AOD (~20%) AOD retrieved at both blue and red bands show an overestimation of AOD compared to AERONET AOD	Not suitable for high reflectance area or retrieval of non-spherical particles likes desert dust aerosols (Kokhanovsky et al., 2007)	Vidot et al (2008)
MERIS/AATSR	2002-2012	Synergistic Aerosol Optical Depth (AOD)	Daily, 7km (default based on BEAM software toolbox)	21 TOA radiance (8 from AATSR and 13 from MERIS)	Good correlation with AERONET (R^2 between 0.60 and 0.90) mainly depending on land cover types	Combining the spectral and angular information from the AATSR and MERIS instruments shows potential for improved characterization of aerosol properties and surface reflectance compared with single-instrument retrievals	North et al. (2009) Benas et al. (2013)
SUOMI NPP	Available since 2013	Visible Infrared Imaging Radiometer Suite	AOT and Angstrom exponent at 6 km and 10 km at nadir	22 channels covering from 0.412 to 12.05 μm	VIIRS AOD is biased high compared to MODIS on	New generation of operational satellite sensors that are able to provide aerosol products with a similar quality to	Jackson et al. (2013) Xiao et al. (2016)

		(VIIRS) provides AOD, Angstrom exponent, suspended matter, Dust single scattering albedo	and edge respectively Suspended matter at 0.75 km and 1.2 km at nadir and edge respectively.		Aqua (by 50% or more over land, and 10% over ocean). Comparing against	MODIS	
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3. Aerosols studies in Malaysia (the Known)

The in-situ data provided by MetMalaysia, DOE and WMO GAW stations and remotely sensed data provided by the AERONET stations and aircraft/space-borne sensors on daily, monthly and annual basis have been used in Malaysia for studying:

- (i) Air quality trends and spatio-temporal variation of PM_{10} concentrations
- (ii) Aerosol optical depth, types and size distributions
- (iii) Trajectory pathway of aerosols/major air pollutants
- (iv) Particulate Matter modelling
- (v) Relationship between major air pollutants i.e. PM_{10} , NO_2 , CO , SO_2 , O_3 , and meteorological variables
- (vi) Health impact of fine-mode particles

3.1 Air quality studies

At the present, the scientific knowledge on the spatial and temporal patterns of aerosols over Malaysia is well established. Air quality assessment conducted by Awang et al. (2000) and Abas (1996, 2004) demonstrated that severe air pollution existed only in the highly-urbanized areas such as Kuala Lumpur, Johor Bahru and George Town. The main cause of air pollution during that period was the motor-vehicle exhausts with high emissions of suspended particulate matter (SPM) mainly during the morning rush hours, while the late evening peaks were mostly influenced by the meteorological conditions (low mixing height and decreased wind speed) favouring the accumulation of aerosol and pollutants near the surface. More recent studies conducted over the Klang Valley area (one of the most developed industrial and economic regions in the country) also concluded that the heavy traffic flow was the main reason for the elevated atmospheric pollutants in this region (Azmi et al., 2010).

Spatio-temporal analysis of particulate pollution (measured by the DOE) covering entire Malaysia was also conducted by Juneng et al. (2009) revealing that the PM_{10} concentrations exhibited a remarkable seasonal variation with maximum values from May to September (dry season) associated with the prevalence of south-westerly winds. The minimum PM_{10} concentrations were found during the north-easterly monsoon (rainy season) from December to March (Abas, 2004). More specifically, the highest concentrations were recorded during the month of August over the western part of Borneo, while the PM_{10} concentrations over Northern Malaysia maximized during the late winter - early spring and summer seasons. Over

the northern part of Borneo, the PM_{10} concentrations exhibited less seasonality with higher concentrations during spring and lower during early winter monsoon. The washout effects of heavy rainfall plus absence of strong ground-based inversion are the main reasons for the low concentrations of atmospheric particles in the rainy season (Inouye and Azman, 1986, Chow and Lim, 1983). Apart from the seasonal variations, another interesting finding of Juneng et al. (2009) work is that the surface PM_{10} concentration had also a remarkable intra-seasonal variation at quasi-bi weekly frequencies of 10-20 days as well as at lower frequencies of 30-60 days. They postulated that the variations in surface aerosol concentrations and properties are modulated by the large-scale tropical climate variations.

The concentration of $PM_{2.5}$ has been recorded extensively for the first time by the United States Environment Protection Agency, (US EPA) at Petaling Jaya, Malaysia during the haze episode in November, 1997 (Pinto et al. 1998). The results revealed that the $PM_{2.5}$ concentrations far exceeded the concentration limits suggested for 24- hour averages by US EPA ($35 \mu\text{gm}^{-3}$) and World Health organization ($25 \mu\text{gm}^{-3}$). Further study on the chemical composition of $PM_{2.5}$ revealed the dominance of organic substances in $PM_{2.5}$ concentration. Two major inorganic trace elements i.e., Potassium (K^+ : 280ngm^{-3}) and Sulfur (S^2 : 2400ngm^{-3}) were also found to dominate the $PM_{2.5}$ concentrations during the haze episode (Pinto et al. 1998).

Further investigations on $PM_{2.5}$ mass concentration and composition have been conducted by Tahir et al. (2013) in the east coast of Peninsular Malaysia in Kuala Terengganu, by Ee-Ling et al.(2015) and Khan et al. (2016) in Bangi (west coast of Peninsular Malaysia) and by Amil et al. (2016) in Petaling Jaya, Malaysia. These results indicated that the $PM_{2.5}$ concentrations exceeded the limit of air quality standards ($25 \mu\text{gm}^{-3}$) suggested by the World Health organization most of the time, while during haze episodes the $PM_{2.5}$ concentrations maximized and exceeded the limit suggested by US EPA ($35 \mu\text{gm}^{-3}$). Source apportionment studies using principal component analysis (PCA) and positive matrix factorisation (PMF) indicate that the major sources of these $PM_{2.5}$ are mostly biomass burning, motor vehicles, road dust, industrial and sea salt (Amil et al. 2016; Khan et al. 2016).

Besides PM, variations in O_3 concentrations were also investigated using data from DOE and MetMalaysia. Five years (2005-2009) of O_3 data taken from three stations representing the urban, suburban and rural environments Banan et al. (2013) found the highest O_3

concentration was recorded in the suburban area with an average daily maximum value of 60 ± 20 ppbv. Furthermore, the O_3 concentration was significantly affected by the amount of Nitrogen Oxides due to the photochemical processes and effects of oxidation (Banan et al., 2013). In another study Latif et al. (2012) analysed the seasonal pattern of tropospheric O_3 and its sources in one of the most industrialised regions in Malaysia (Klang valley- Figure 1), revealing that almost half of the high- O_3 days were caused by localized circulation patterns and local emissions while, the rest of the high- O_3 days were due to the long-range and mid-range transport from northeast across the South China Sea (Latif et al., 2012).

The surface O_3 concentrations at the Tanah Rata regional GAW station (Figure 3) from June 2006 to August 2008 have been analysed by Toh et al. (2013). The afternoon maximum and night-time minimum dominated in the diurnal variation of surface O_3 . A significant finding was the much lower average O_3 concentrations during the rainy compared to non-rainy days. During the spring inter-monsoon months (March-April) and the northeast monsoon (November-March), hourly-averaged O_3 concentrations and meteorological parameters (i.e. temperature and relative humidity) were significantly correlated, indicating the effect of regional transport of air pollutants from Indonesia, while during the burning season from May to late September the biomass-burning emissions could lead to enhanced photochemical production of O_3 . Five-day backward trajectories during two high- O_3 episodes showed that biomass burning in Sumatra and long-range transport of pollution from Indochina were the reasons for the high tropospheric O_3 concentrations on August 7, 2006 (40.0 ppb) and February 24, 2008 (45.7 ppb), respectively. That study revealed the major sources of transported air pollution over Malaysia, i.e. Indonesia tropical peat and forest fires and Indochina pollution plumes.

Variations of columnar CO_2 and tropospheric O_3 concentrations over entire Peninsular Malaysia were analysed by means of data retrieved from satellite remote sensing i.e. SCIAMACHY (Tan et al., 2013, 2014). Monthly-averaged (between 2003 and 2009) atmospheric CO_2 column dry air mole fractions (XCO_2) showed an increase of 15 ppm over Peninsular Malaysia, which was attributed to biomass burning, fossil-fuel combustion and land use changes. CO_2 showed a significant spatio-temporal variation with high values (399-414 ppm) during the end of summer (October) and early winter (November) period. The high CO_2 concentrations were associated with cold air outbreaks from Siberia that passed through the heavily polluted East Asia before reaching the SEA (Tan et al., 2013).

3.2 Aerosol optical depth

In addition to PM_{10} , the integrated columnar aerosol extinction corresponding to the aerosol optical depth (AOD) has been also examined to understand the spatial and temporal variations of aerosols and their impact on radiative transfer over Malaysia (IPCC, 2007). AOD measurements from the three AERONET stations in Malaysia were used by Kanniah et al. (2014a) to illustrate the diurnal pattern and seasonality in aerosol columnar loading. All three sites (Kuching, USM Penang and Tahir representing coastal, urban and non-urban locations respectively) exhibit either insignificant diurnal variations or slightly higher AODs during the afternoon, while their short-term availability does not allow for a trend analysis at the present. Using a 4-months (August–November, 2011) AERONET dataset (AOD and Angstrom exponents) from the Kuching station, Salinas et al. (2013) studied the aerosol physical and optical characteristics during the dry season, which is dominated by biomass-burning activities. Their findings reveal that Kuching exhibited relatively low aerosol loading (<0.2), which increases with regional episodes of biomass burning.

Level 2 quality assured spectral AOD and Angstrom exponent retrievals obtained through the AERONET sites at USM, Penang (north of Peninsular Malaysia) were used by Tan et al. (2015) to analyse the seasonality in optical depth, types of aerosols and their source apportionment. Using two years of data they found AOD values ~ 0.73 during the southwest monsoon (June –September) which is associated with forest fires in Indonesia. Meanwhile, reduced AOD levels (0.18–0.19) were found in the inter- monsoon period (October–November and May). In the same study, Tan et al. (2015) used visibility and air pollutant index (calculated from Carbon Monoxide, Ozone, Nitrogen Dioxide, Sulfur Dioxide and PM_{10} as well as AOD from AERONET) in order to develop an empirical model for estimating of the missing AOD values due to frequent cloudiness over Malaysia. The estimated AODs showed satisfactory accuracy with the AERONET values ($R^2 = 0.68$ and $RMSE = 0.14$) which allows for filling the missing data and obtaining a continuous AOD data series for climatological studies in Malaysia. Ground-based backscatter Lidar data (operated at 355 nm) from the same USM site was used by Hee et al. (2014) to monitor a short-term (8 days) haze event. Hee et al. (2014) examined the daytime variation of the AOD, its vertical distribution and the planetary boundary layer (PBL) height during the haze days concluding that most aerosols were contained below the PBL at around 1000 – 2000 m in height.

AERONET AOD data were also used to validate AOD retrievals from MODIS, MERIS and MERIS/AATSR sensors over Malaysia (Kanniah et al., 2014a, b). These studies found that MODIS AOD is well correlated with AERONET AOD and, therefore, MODIS retrievals can be used to study the columnar aerosols over Malaysia (Kanniah et al., 2014a). However, although MERIS sensor provides a plausible spatial and temporal pattern in AOD over Peninsular Malaysia, the absolute AOD values were biased (Kanniah et al., 2014b) similar to the findings reported by Vidot et al. (2008). Thus, the MODIS-derived AOD was used to describe the spatial, seasonal and inter-annual variability of aerosols over Peninsular Malaysia (Kanniah et al., 2014b), possessing a significant importance for examining the influence of aerosols on regional climate. The results by Kanniah et al (2014a) show that an average AOD₅₀₀ of ~0.40 - 0.47 was recorded in cities located in the west coast of Peninsular Malaysia, whereas the lowest AODs (0.12 ± 0.09) were found in mountainous locations with minimal local aerosol production in the middle of Peninsular Malaysia. The AOD variability over a period of 10 years (Terra satellite) shows a neutral-to-declining trend. In general, during the inter-monsoon season (April-May), the northernmost part has relatively higher AODs (0.4-0.7), while during the wet season the AOD is at its lowest levels, especially over the eastern coast (<0.2), which receives the largest amount of the northeast rainfall (Kanniah et al., 2014a). However, during the end of the dry season (September-October), the AODs tend to be higher over Peninsular Malaysia closely related to the accumulation of the biomass-burning aerosols coming mainly from Indonesia. The Terra-MODIS AOD values showed stronger AOD inter-annual variability over Peninsular Malaysia with considerably higher AOD values (0.37 ± 0.59) in 2004 and 2006. The source of the intense 2006 haze that covered the whole Southeast Asia (Hyer and Chew, 2010) was largely anthropogenic but its cross-boundary transportation and large extension are particularly influenced by the El-Niño (Tanggang et al., 2010).

3.3 Aerosol size distribution and types

Knowledge about the aerosol size distribution and the various types from local to global scales are necessary in radiative forcing and climate change studies (Kim and Ramanathan, 2008; Sinha et al., 2013). In general, the aerosol size distribution usually exhibits two distinct modes, i) for fine particles (aerosols originated from anthropogenic sources such as biomass burning, urban/industrial etc.) with diameter $<0.6 \mu\text{m}$ and ii) for coarse particles (natural sources of aerosols such as sea salt and desert dust) with diameters $>0.6 \mu\text{m}$ (Dubovik et al.,

2002), although in several cases, mostly over desert environments, the aerosol size distribution can be even tri-modal (Dubovik et al., 2002; Masmoudi et al., 2003). The various aerosol types have different optical, physical and chemical characteristics.

Data from ground-based (such as AERONET, Multifilter Rotating Shadowband Radiometer) and satellite-based (TOMS, MODIS) measurements have been used to discriminate the aerosol types over the globe (e.g. Barnaba and Gobbi, 2004; Pace et al., 2006; Kaskaoutis et al., 2007a; Reinart et al., 2008; Yang and Wenig, 2009). Several aerosol parameters such as AOD, Fine Mode Fraction (FMF)¹ and Angstrom Exponent (AE)² retrieved from satellite and ground based instruments have been used to study the aerosol sizes and types. The Ångström exponent is inversely related to the average size of aerosol particles; the smaller the particles, the larger the AE (Kahn et al., 2009), while its determination at shorter or longer wavelengths gives specific information about changes in fine-mode radius and in fine-mode fraction, respectively (Schuster et al., 2006; Kaskaoutis et al., 2007b). In a different classification, the aerosol particles are indicated as coarse mode (> 2.5 µm in diameter) when the AE is < 1 and as fine mode (0.5 – 2.5 µm) for AE values above 1 (Queface et al., 2003).

The definition of a range of values for AOD, AE and/or FMF that are assumed as representative for a particular aerosol type is not an easy task and varies from location-to-location based on several considerations such as the distribution of the AOD, AE and/or FMF values, the aerosol-mixing processes in the atmosphere such as coagulation, condensation, humidification and gas-to-particle conversion (Smirnov et al., 1995, 2002). One of the most common methods used for aerosol type classification is the correlation between AOD, AE and/or FMF (e.g. Barnaba and Gobbi, 2004; Pace et al., 2006; Kaskaoutis et al., 2007a, b; Kaskaoutis et al., 2009; Jalal et al., 2012, among many others). Jalal et al. (2012) had implemented a scatter plot between AOD and AE (obtained from AERONET) in order to classify the aerosol types for the first time over Kuching, Malaysia. The AOD and AE values that were used as thresholds for specific aerosol type classification by Jalal et al. (2012) are

$$^1 FMF = \frac{\text{Fine mode AOD at } 500(550) \text{ nm}}{\text{Total AOD at } 500(550) \text{ nm}}$$

AOD at 500 and 550 nm from AERONET and MODIS respectively

² The AE (or α) expresses the spectral dependence of the aerosol optical depth (τ) with respect to wavelength (λ) as inverse power law: $\tau \propto \lambda^{-\alpha}$.

summarized in Table 2. Based on this classification, four aerosol types were identified namely maritime, dust, continental/urban and biomass burning.

Table 2: Thresholds of AOD and Ångström exponent for classification of aerosols over Kuching AERONET site, Malaysia by Jalal et al. (2012).

Aerosol types	AOD (440nm)	Ångström exponent
Maritime	< 0.3	0.5 – 1.7
Dust	>0.4	<1.0
Continental/urban	0.2 – 0.4	>1.0
Biomass burning	>0.7	>1.0

More recently, AE data obtained from two AERONET stations (for a period of 3 months-September to November) in the north part of Peninsular Malaysia were used to characterise the aerosol size (Alias et al., 2014). Although the average AOD at the sites was relatively low (0.05 – 0.4), mostly mixed to fine-mode particles were found to dominate ($\alpha > 0.75$). Nevertheless, one site (USM campus), which is located near to the sea, exhibited dominance of coarse-mode particles. The limited number of data (only 3 months of measurements) as well as the large variability of the aerosol types may be the cause for a low correlation ($r^2 = 0.029$ at USM, northern part of Peninsular Malaysia) and $r^2 = 0.013$ at UiTM west coast of Peninsular Malaysia) between AOD and AE suggesting that the AE can either increase with AOD, indicating additional presence of fine-mode aerosols, or even decrease revealing enhanced presence of maritime aerosols or dust or even indicating aerosols growth due to gas-to-particle conversion and coagulation processes. Furthermore, the high levels of humidity over tropical Malaysia may contribute to the hygroscopic growth of aerosol particles and other mixing processes in the atmosphere, resulting in a mixture of aerosol (external or even internal) over the region. The spectral AOD and AE values from a 4-months Kuching-AERONET dataset during the dry season (August-November 2011) indicated the dominance of fine-mode aerosols from urban, aircraft and biomass-burning origin (Salinas et al., 2013). Similarly, in Penang (northern island of Peninsular Malaysia) biomass aerosols were the major pollutant, mostly originated from active burning in local and

neighbouring countries. Other aerosol types, i.e. dust, only minimally affected the area (Tan et al., 2015).

In another study Kanniah et al. (2014a) identified different aerosol types over peninsular Malaysia based on FMF data from MODIS sensor covering a period of a decade (2000-2009). As it was expected relatively higher monthly-mean FMF values (between 0.85 and 0.9) were observed in the dry season especially in the months of June, September and October and low values (0.26 - 0.32) were recorded during the rainy season (November to January). This indicates the dominance of fine-mode aerosols in the dry season and coarse-mode aerosols in the wet season that are mostly of marine origin or mixing of marine with continental aerosols. The results from the correlation between MODIS FMF and AOD showed that about 38% of the aerosols observed in Peninsular Malaysia between 2000-2009 are of continental/urban and biomass-burning types with $FMF > 0.8$ (small size), followed by dust type aerosols (27%) with $FMF > 0.6$ and $AOD > 0.4$, and maritime aerosols (12%) with $FMF < 0.7$ and $AOD < 0.3$. These findings can be reasonably explained due to the occurrence of forest fires in Indonesia during the dry season, since the smoke emissions composed mostly from fine particles were transported to Peninsular Malaysia by the southwest monsoon. Moreover, during the wet season, the precipitation is much higher and the wind blows from northeast directions crossing the South China Sea, thus increasing the amount of coarse-mode particles of maritime origin. Besides, coarse aerosols characterized as dust were found more abundant in the wet season although Peninsular Malaysia is not affected by transported dust plumes from southwest Asia (Gautam et al., 2013). These aerosols maybe a mixture of marine particles with continental emissions, which can grow up in size via the humidification process. More detailed investigation is needed in this issue by examining the spectral dependence of several aerosol parameters, like AOD, AE, single scattering albedo (SSA), asymmetry factor, etc (Bahadur et al., 2012).

3.4 Trajectory pathways of aerosols/major air pollutants

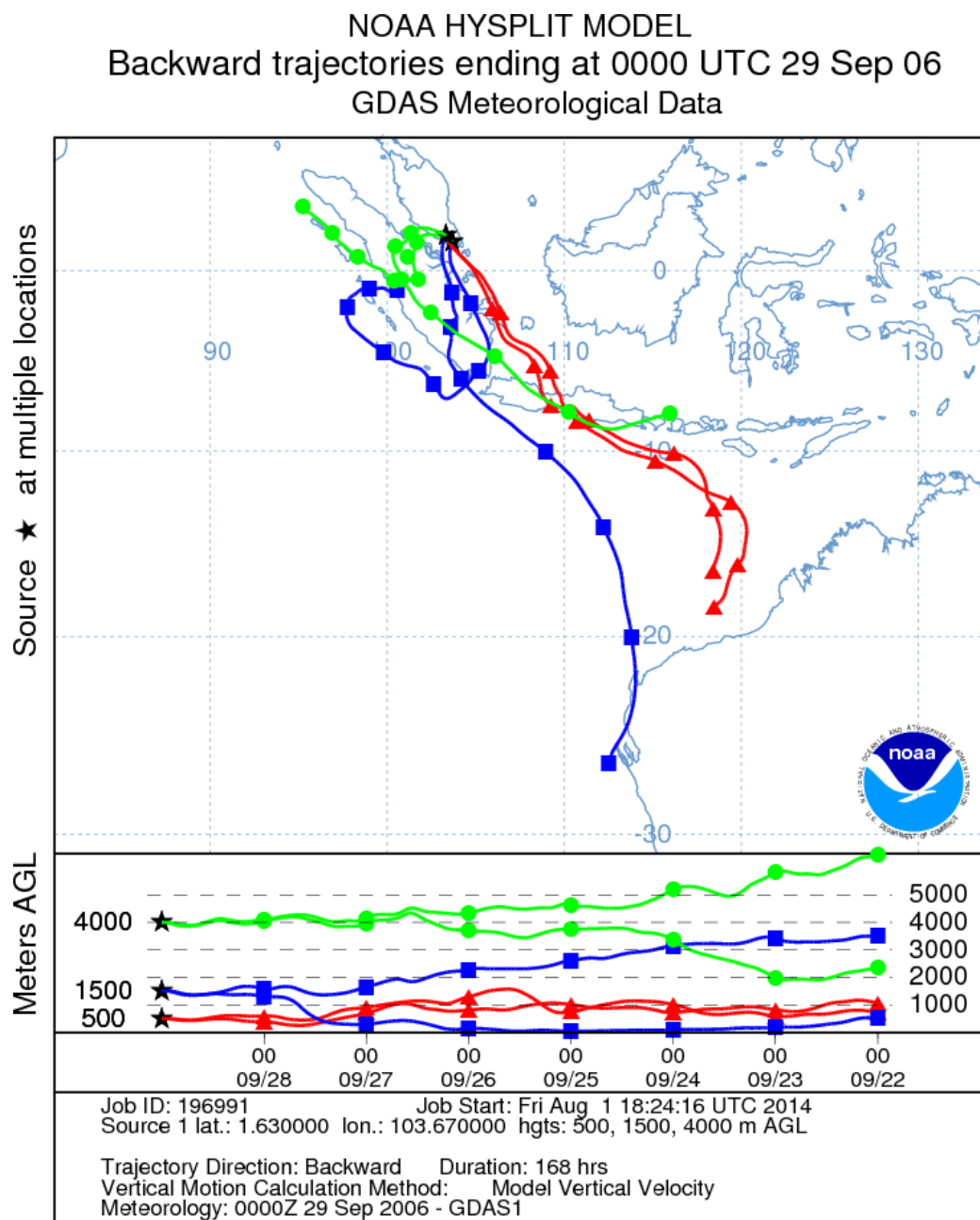
Tracking the route of aerosols transported from the source region to other locations is very important due to various far-reaching impacts of aerosols and associated microorganisms on human health and ecosystems, terrestrial and oceanic biogeochemical cycles, weather systems and climate (e.g., Kaufman et al., 2005; Yu et al., 2015). Such studies are essential in Malaysia that receives significant amount of aerosols and transboundary pollution as part of

the Southeast Asian pollution outflow (Lawrence and Lelieveld, 2010). One common way to identify the aerosol source regions and transport height of the polluted air masses is by using the HYSPLIT (Hybrid Single Particle Lagrangian Intergrated Trajectory) model (e.g. Kosmopoulos et al., 2008; Sinha et al., 2013; Lin et al., 2014; Lv et al., 2015; Pérez et al., 2015).

The aerosol sources in Peninsular Malaysia were also identified by means of the HYSPLIT trajectory model (Kanniah et al., 2014a). 5-day air-mass back trajectories **showed that the** aerosols during the dry season (June to September) were mainly originated from Sumatra, Indonesia. The southwest monsoon during the dry season carries mainly biomass-burning aerosols from Sumatra to Peninsular Malaysia within 24 hours, while during the northeast monsoon (November to March) the aerosols were mostly associated with the northeast winds blowing from the southern China Sea (Kanniah et al., 2014a). The western and southern parts of the Peninsular Malaysia are the mostly affected areas by the biomass burning in Sumatra, since the polluted air masses initially cross these regions. At higher altitudes (>1500 m) the trajectories mainly originate from the equatorial Indian Ocean, thus being mostly clean at elevated layers. HYSPLIT 7- days back trajectories (Kanniah et al., 2014a) showed that the lower (500 m) air masses were found to originate from the Java Sea and northwest Australia (Figure 4), thus further contributing to the aerosol loading over Malaysia via transportation of biomass smoke during the dry season (Bouya et al., 2010).

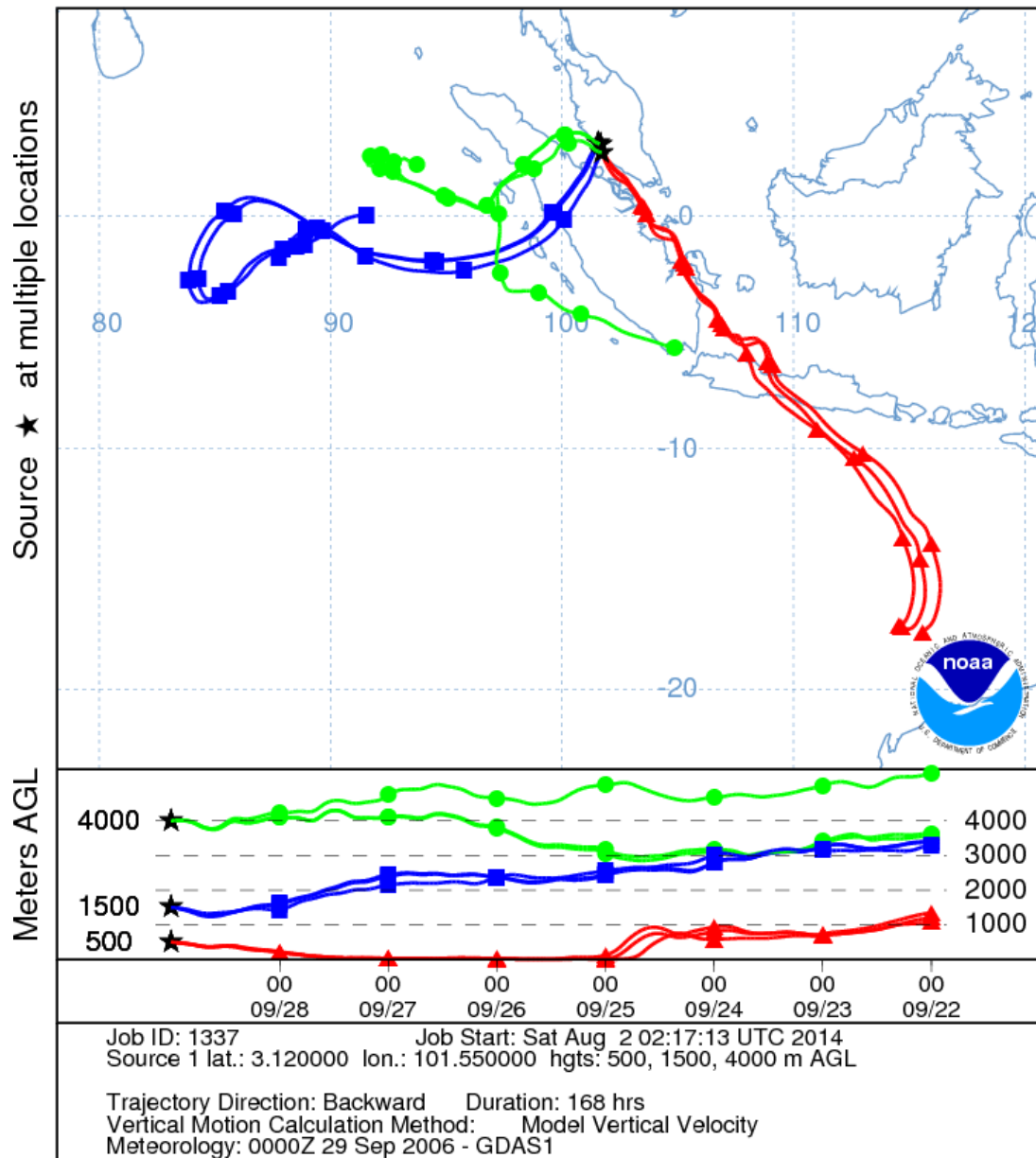
As far as the wet season (November to March) is concerned, the air masses in the mid troposphere) remain from the equatorial Indian Ocean. The main difference from the southwest monsoon is detected at the lower air masses, which come from northeastern directions (southern China Sea) due to changes in the wind pattern and are usually less aerosol loaded. Despite the fact that the southern China Sea cannot be characterised as a clean marine environment due to significant continental outflow, the lower aerosol loading during the wet season and the higher precipitation over the area prevent significant aerosol transportation over Peninsular Malaysia, thus resulting in lower AODs (Kanniah et al., 2014a). Over the urban/industrialised regions (western part of Malaysia) the aerosols are mostly of local origin while transported aerosol plumes contribute on certain occasions leading to haze or even pollution smog environments, like during the dry season of 2009. In HYSPLIT backward trajectories analysis, the aerosols over Peninsular Malaysia are found basically transported from southwest directions (Indonesia islands like Sumatra and

Kalimantan) and northwest directions (South China Sea) during dry and wet seasons, respectively (Kanniah et al., 2014a, Tan, et al., 2013, 2014).



(a)

NOAA HYSPLIT MODEL
 Backward trajectories ending at 0000 UTC 29 Sep 06
 GDAS Meteorological Data



(b)

Figure 4: HYSPLIT backward trajectories (7 days) calculated in the dry southwest season (29 September 2006) for southern (a) and western (b) Malaysian regions. Lower-to-mid troposphere trajectories may originate as far as northern and western Australia.

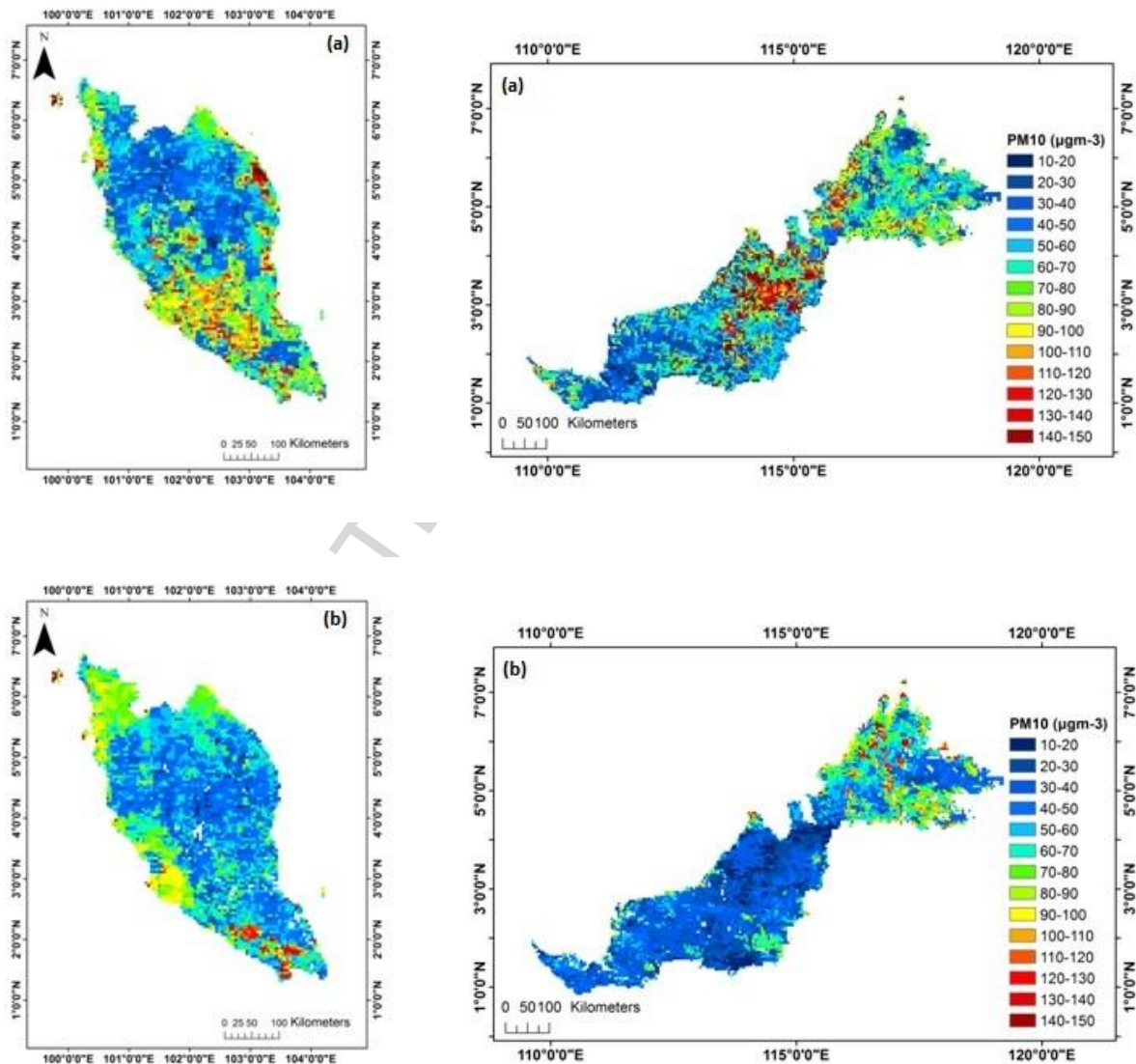
3.5 Particulate Matter modelling

The vast majority of the air-quality studies performed in Malaysia have been based on station-measured air quality data (i.e. PM_{10} data) that have limited spatial coverage. Therefore, there is a growing interest in using satellite-derived AOD as an intermediate tool to estimate PM_{10} concentrations in the ground, thus providing a spatial mapping over Malaysia that helps in improving air-quality forecasts (Yap and Hashim, 2013; Kanniah et al., 2014b). Van Donkelaar et al. (2010) developed a global model to predict $PM_{2.5}$ using AOD from both MODIS and MISR satellite sensors and the outputs are used by the global Environmental Performance Index (EPI) to measure the atmospheric pollution over each country in the world. Similar models have also been developed for the US (Kittaka et al., 2004; Al-Saadi et al., 2005; Liu et al., 2007; Gupta and Christopher, 2009; Saunders et al., 2014;), continental India (Dey et al., 2012), Athens, Greece (Kaskaoutis et al., 2010; Benas et al., 2013), Italy (Arvani et al., 2015), Agra city, India (Chitranshi et al., 2014), Tehran, Iran (Sotoudeheian and Arhami, 2014), and Alpine region (Emili et al., 2010).

For PM estimations from space, remotely-sensed AOD is correlated with ground-based PM using various statistical regression analysis (Wang and Christopher 2003; Engel-Cox et al., 2004; Gupta and Christopher, 2009a; Lee et al., 2011; Kloog et al., 2012; Benas et al., 2013; Nordio et al., 2013; Yap and Hashim, 2013; Chitranshi et al., 2014; Hu et al., 2014; Kanniah et al., 2014b; Sotoudeheian and Arhami, 2014; Xie et al., 2015; Beloconi et al., 2016) and statistical and chemical transport models (Van Donkelaar et al., 2010). **In some studies, additional information of vertical profiles of aerosols and meteorological parameters that affect the aerosol properties such as surface temperature, relative humidity, wind speed, and Boundary Layer Height (BLH) have been also included for more accurate estimations of PM from space** (Gupta and Christopher, 2009a; Emili et al., 2010; Rohen et al., 2010; Wang et al., 2010; Benas et al., 2013; Kanniah et al., 2014b).

Focusing on Malaysia, Yap and Hashim (2013) attempted to estimate PM_{10} over Peninsular Malaysia via a mixed-effects model, which took into consideration the monthly variability of the coefficients (i.e. slope and intercept) in association between MODIS-AOD and PM_{10} . That model provided more predictive accuracy relative to linear regression models. For improving the accuracy of PM_{10} estimates at daily time step, **Kanniah et al. (manuscript in preparation) used MODIS AOD data along with atmospheric stability, surface temperature and relative humidity derived from MODIS** along with advanced statistical methods, i.e. multiple linear regressions (MLR) and Artificial Neural Networks (ANN), for PM_{10} monitoring from space for the first time over the whole Malaysian territory (Figure 5).

The seasonal-mean PM_{10} mapping over Malaysia revealed significant spatio-temporal heterogeneities in the PM_{10} concentrations, as a result of the high heterogeneous aerosol and meteorology fields, especially in East Malaysia that has only 15 PM_{10} monitoring stations compared to its size of 198,080 km². The satisfactory performance of the ANN model for PM_{10} estimations (RMSE of 10.16 $\mu\text{g m}^{-3}$) and mapping over Malaysia allows for future estimates of $PM_{2.5}$ concentrations from space and monitoring of the EPI.



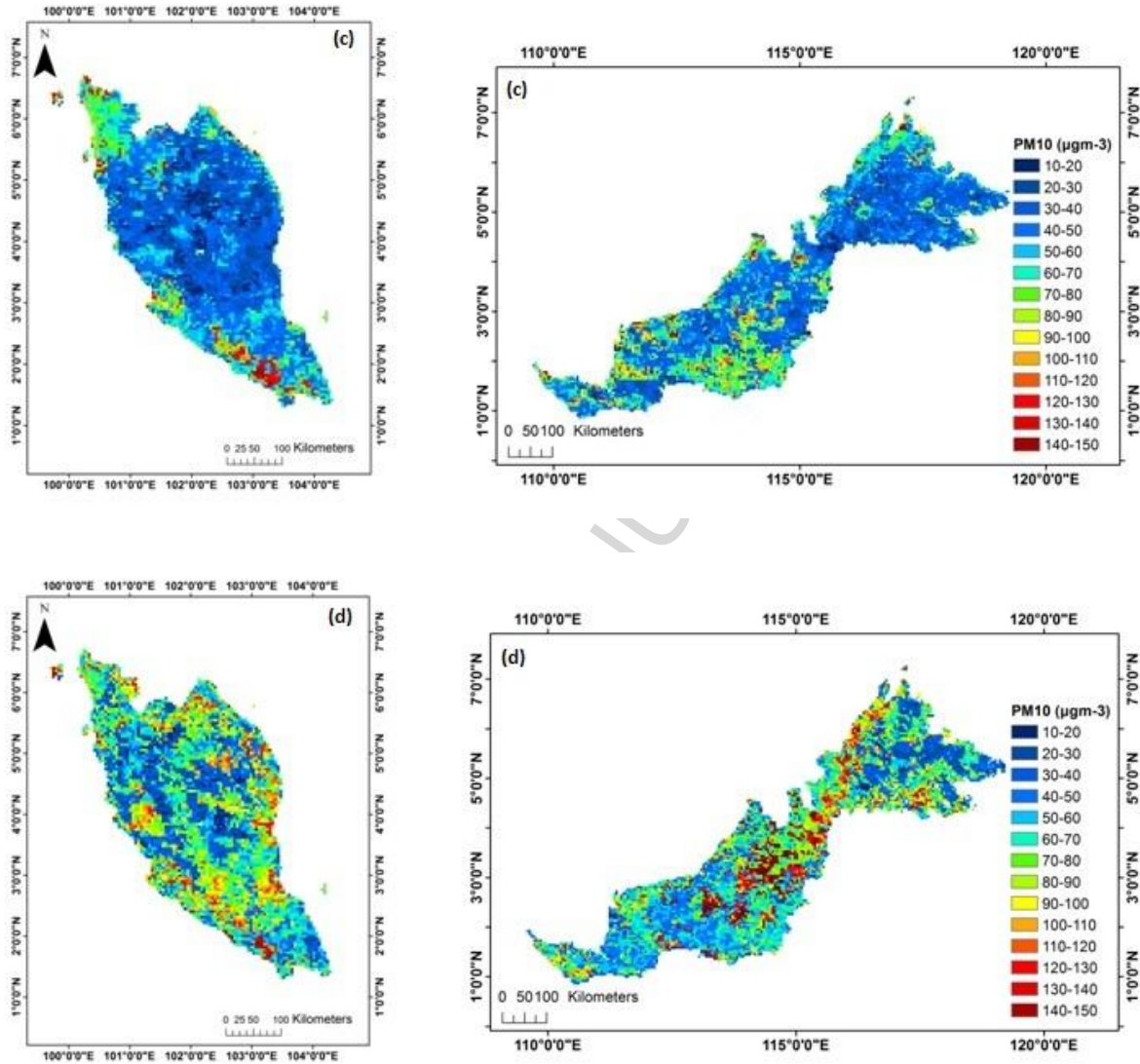


Figure 5: Spatial distribution of estimated PM_{10} concentrations over Malaysia during 2007-2011 for (a) dry season (June – September), (b) wet season (November – March), (c) inter-monsoon (April-May) and, (d) inter-monsoon (October), based on MODIS-AOD₅₀₀ and meteorological variables. The estimates were made by means of Artificial Neural Network technique with a spatial resolution of 10 x 10 km.

3.6 Relationship between PM_{10} , air pollutants and meteorological parameters

A preliminary study examining the relationship between atmospheric pollutants and meteorological variables in Malaysia showed that the PM_{10} concentrations were positively correlated with ambient temperature and negatively correlated with humidity (Azmi et al., 2010), while other variables such as wind speed and UV radiation were found to slightly affect the PM_{10} concentrations. Potential factors influencing the PM_{10} variations over the Klang Valley were also examined by Juneng et al. (2011), revealing that local meteorological

factors such as surface air temperature, humidity and wind speed dominated the variation of PM_{10} during the summer monsoon, while synoptic weather conditions were also found to contribute to the spatial and temporal PM_{10} variations. More recently, Tan et al. (2016) successfully developed a new ozone (O_3) algorithm based on CO_2 , O_3 , CH_4 , NO_2 , and H_2O atmospheric gases allowing predictions of O_3 variations via statistical methods. Results of the correlation analysis showed that increase in columnar ozone concentration was associated with an increase in NO_2 , since NO_2 is a precursor of O_3 , and a decrease in water vapor in the wet (November-March) and dry (May-September) seasons.

3.7 Health impacts of aerosols

Only very few studies have dealt with examining the health impacts of aerosols in Malaysia. The available studies (Awang et al., 2000; Nasir et al., 2000) mostly focus on the possible health effects during the 1997 haze crisis. Those studies found a direct positive relationship between the number of cases of diseases such as upper respiratory tract infections, asthma and conjunctivitis and the Air Pollution Index. The high health damage cost, which was estimated to be around RM 129 million, was linked to long duration of the haze (Nasir et al., 2000).

A recent study by Othman et al. (2014) analysed the inpatient cases due to haze occurrence in the Klang valley, Malaysia area and assessed the economic value of health impacts. The results revealed that transboundary haze pollution increased inpatient cases by 31% from the normal days. The average annual economic loss due to the inpatient health impact of haze was valued at ~\$91,000 USD. According to Sahani et al. (2014), haze events were found to be significantly associated with natural and respiratory mortality at various lags. For natural mortality, haze events at lagged 2 showed significant association with children less than 14 years old (Odd Ratio (OR) = 1.41; 95% at Confidence Interval (CI) = 1.01to1.99), while the respiratory mortality was significantly associated with haze events for all ages at lagged 0 (OR = 1.19; 95% CI = 1.021.40). Nevertheless, Khan et al., (2016) reported that the noncarcinogenic cancer risk posed by the exposure of $PM_{2.5}$ was at a considerably safer level (3–4 per 1 000 000 people) in Malaysia compared to other areas of South and East Asian region.

4. Knowledge Gaps (the unknown)

4.1 Spatial aerosol patterns around the urban areas

Although a better understanding has been gained over the years on the spatial and temporal evolution of aerosols over Peninsular Malaysia via ground measurements and satellite remote sensing, monitoring the aerosol loading and composition over the East Malaysia in the Borneo Island and over urban areas remains a real challenge and not so well documented. Urban centres in Malaysia have significant aerosol loading and pollution levels that are far beyond the air quality standards (Pinto et al., 1998; Ee Ling et al., 2015; Amil et al., 2016; Khan et al., 2016). Therefore, routine monitoring of air quality in cities becomes very crucial. The air-quality monitoring networks in most of the cities have relied almost exclusively on ground-based stations, which do not provide adequate spatial coverage. Higher resolution remote sensing data will be helpful to describe the detailed distribution of the pollution plumes in and around the cities that are characterized by heterogeneous surfaces, and aid in air-quality monitoring and forecasting. Currently, there are no satellite sensors providing AOD and other aerosol parameters at a spatial resolution better than 1.2 km, except for MERIS sensor. Nevertheless, MERIS was found to perform poorly compared to MODIS (Kaskaoutis et al., 2010; Benas et al., 2013; Kanniah et al., 2014b) and it has now stopped its operation. MODIS AOD retrieved at 3km spatial resolution was also found not to be so suitable for air quality studies due to improper characterization of urban surfaces that resulted high bias at $AOD > 0.1$ (Levy et al., 2013; Munchak et al., 2014). Therefore, the high-resolution spatial aerosol mapping around the main cities and industrialized regions along the west Malaysian coast remains an issue for further research.

4.2 Aerosol size

The analysis of Kanniah et al. (2014a) revealed that many data points did not exist to a specific aerosol type and they were identified as mixed (mixing between two or three aerosol types). In their study, Kanniah et al (2014a) identified 17-40% (FMF values below 0.6 and $AOD_{550} > 0.3$) of aerosols as dust on seasonal basis. The data points that were identified as dust aerosols correspond to coarse-mode particles and may have a significant marine influence during the northeast monsoon season. In addition, they may mix with continental/urban and/or biomass-burning aerosols and form a turbid atmosphere with moderate- to coarse-sized particles after coagulation and humidification under a humid tropical environment. Thus, further analysis is needed in order to examine the possible

influence of dust over Peninsular Malaysia, via studying the spectral distribution of SSA and refractive index. Increased absorption in the UV range corresponding to increase in SSA with wavelength is a fingerprint of dust presence (Dubovik et al., 2002). This information from the ground-based instruments is vital to capture the local effects of aerosols and can be obtained when long-term AERONET data from the Malaysian sites become available in the future.

4.3 Aerosol climate implications

Aerosol particles affect the climate by scattering and absorbing the solar radiation (direct effect) and modifying the clouds' properties (indirect effect). For the direct effect, the solar radiation (shortwave and longwave) is scattered and absorbed by aerosols, thereby altering the radiative balance of the Earth-atmosphere system (Jose et al., 2015). A net negative direct aerosol radiative forcing (ARF) will be exerted by the scattering aerosols, while partially absorbing aerosols may result in different effects over dark and bright surfaces (Satheesh et al., 2010). More specifically, a negative top-of-atmosphere (TOA) direct ARF will be exerted over dark surfaces (e.g. ocean and dark vegetated surfaces) and a positive TOA ARF will be exerted over bright surfaces (e.g. deserts, snow and ice) from moderately absorbing aerosols due to multiple reflections between surface and atmosphere that enhance the atmospheric absorption (Hatzianastassiou et al., 2004, 2005; IPCC, 2007). Aerosol radiative forcing studies are yet to be conducted over Malaysia, since the region receives considerable amounts of smoke aerosols almost every year. It is also crucial to understand whether the radiative forcing due to biomass-burning smoke, which is a mixture of organic and black carbon, is positive or negative for its climatic implication (Andreae and Ramanathan, 2013). Kanniah et al. (2014a) provided a preliminary analysis on the relationship between solar radiation and aerosols over Malaysia revealing that aerosols attenuate the total incoming solar radiation by 0.8% for a 0.1 increase in AOD.

Remote sensing observations, in situ measurements and a better knowledge of bulk aerosol optical properties make the assessment of ARF more robust during the last years. Estimates of ARF are either taken directly from global aerosol models (Schulz et al., 2006; Myhre et al., 2013) or based mostly on observations, but using supplemental simulations from models (e.g., Myhre, 2009; Loeb and Su, 2010; Su et al., 2013). Numerous studies dealing with ARF and their effects on monsoon circulation, precipitation re-distribution, solar dimming and atmospheric stability have been performed over the developing countries of south and east

Asia (e.g. Ramanathan et al., 2005, 2007; Lau et al., 2006; Gautam et al., 2009, 2010, 2011; Lawrence and Lelieveld, 2010; Zhuang et al., 2013, 2014; Che et al., 2014; Lin et al., 2014). On the other hand, many studies have dealt with ARF estimations over urban areas in south and east Asia (e.g., Ramachandran and Kedia, 2010; Alam et al., 2011; Niranjana et al., 2012; Che et al., 2014; Kaskaoutis et al., 2013; Sinha et al., 2013; Zhuang et al., 2014; among many others). Aerosol data such as AOD, water vapour and albedo from satellite sensors (MODIS, MISR and Calipso) and AERONET measurements are usually used for ARF estimates. The vast majority of these studies used the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model along with sun-photometer retrievals of aerosols and MODIS observations of surface reflectance in order to calculate the ARF at TOA and surface.

Regional studies have also been conducted to quantify ARF under certain turbidity conditions. Focusing over Indochina (10–22°N and 95–107°E), Lin et al. (2014) simulated the effect of aerosols originated from biomass burning on radiative forcing at surface, atmosphere and TOA during the spring time. The ARF at surface was found to be -26.85 W m^{-2} , and at TOA -4.74 W m^{-2} , resulting in atmospheric heating of 22.11 W m^{-2} . A reduction in shortwave radiation of about 20 W m^{-2} at surface was also found in a zone stretching from southern China to Taiwan; such significant reduction may exert a great impact on the regional climate in East Asia (Lin et al., 2014). Furthermore, WRF-Chem model simulated high AOD (0.8–1.2) over Indochina (10–22°N and 95–107°E), where 34% of the AOD was attributed to organic carbon and 4% to black carbon.

Feng and Christopher (2013) assessed the spatial and temporal distributions of AOD and FMF acquired from MODIS and AOD at 558 nm and SSA obtained by MISR as well as the shortwave radiation (obtained by Clouds and the Earth's Radiant Energy System, CERES) in the southeast Asian region (10°S–25°N, 90°E–150°E) and quantified the regional cloud-free diurnally-averaged shortwave ARF at TOA over both land and ocean. The ARF at TOA was found to be $-6.4 \pm 1.2 \text{ W m}^{-2}$ and $-5.9 \pm 1.3 \text{ W m}^{-2}$ over land and ocean, respectively. The fine-mode aerosols, especially from biomass burning and anthropogenic emissions that dominated over the region were found to mostly contribute to ARF. However, that study was performed under cloudless conditions, while the cloud-cover fraction over this region is above 60%, thus preventing the usage of a complete aerosol database in order to examine the aerosol effects on regional climate. Therefore, further aerosol sampling and analysis of their

optical properties underneath the cloud layers are needed in future field campaigns for assessment of the ARF under cloudy conditions.

The quantification of the direct aerosol radiative forcing depends on several factors such as aerosol composition (i.e. relative abundance of various individual species like water-soluble scattering particles, absorbing carbonaceous particles, mineral dust, etc.), their mixing state, size and number concentrations (Sinha et al., 2013; Srivastava et al., 2014). Currently, information about the dominant aerosol types and their composition in different seasons and locations (i.e. urban, rural, and coastal), their mixing state etc. is not widely available over Malaysia. Nevertheless, we have gained some preliminary understanding on the types of aerosols present in Peninsular Malaysia and their seasonality based on AERONET and MODIS satellite data (Jalal et al., 2012; Alias et al., 2014; Kanniah et al., 2014; Tan et al., 2015). The availability of long-term AERONET data in the future will assist in characterising the size and types of aerosols in Malaysia, allowing us for a short-period climatology of aerosol properties and types that could be used in the radiative transfer models for long-term ARF estimates. The dominant aerosol types for different seasons and regions over Malaysia are highly desired for assessing the radiative effects of aerosols and for climate change studies. On the other hand, ARF estimations using ground-based measurements have been limited only over the AERONET sites due to lack of other sun-photometer measurements over Malaysia. Except of the few studies that cited above, there are no other results concerning ARF estimations, even for short pollution events or case studies, over Malaysia and this gap of knowledge should be healed in the future.

4.4 PM monitoring

Most of the particulate matter studies in Malaysia are currently based on ground PM_{10} measurements and to a smaller extent on satellite observations. On the other hand, the $PM_{2.5}$ concentration measurements are limited over Malaysia due to the unavailability of $PM_{2.5}$ in the ambient air quality standards. However, since 2015 a new air-quality standard which includes $PM_{2.5}$ has been formulated (<http://www.doe.gov.my/portalv1/wp-content/uploads/2013/01/Air-Quality-Standard-BI.pdf>). Malaysia DOE and MetMalaysia still measure the PM_{10} concentrations at all their sampling stations, while the $PM_{2.5}$ concentrations and chemical compositions are analysed by individual groups such as Ee-Ling et al. (2015) and Khan et al. (2016) Therefore, the limited continuous $PM_{2.5}$ database reduces the

capability of fine-particulate modelling over Malaysia, although the fine particles are very important on examining the aerosol's impact on human health and climate during hazy conditions. Nevertheless, Beh et al. (2012) studied the $PM_{2.5}$ and $PM_{1.0}$ distribution in three cities (Georgetown, Batu Ferringhi and Permatang Damar Laut) located in the northern part of Peninsular Malaysia. These preliminary results indicated that the highest 6-hour averaged PM concentrations in the Batu Ferringhi city were found to be $200 \mu\text{gm}^{-3}$ for PM_{10} , $194 \mu\text{gm}^{-3}$ for $PM_{2.5}$ and $185 \mu\text{gm}^{-3}$ for $PM_{1.0}$, thus highlighting an extremely large contribution of fine particulate pollution during turbid conditions in Malaysian cities. With the larger availability of $PM_{2.5}$ concentrations at several sites over Malaysia in the near future, the spatial and temporal analysis would become available and would be a useful tool for monitoring and forecasting of air pollution.

In synopsis, the limited data of fine-particle concentrations, composition, toxicity, and columnar aerosol properties is the major issue for lack of studies dealing with the impact of fine particles on human health and climate, especially during haze episodes. Moreover, lacking of data gathering from environmental epidemiological analysis is also a major issue for studying the health impacts of aerosols in Malaysia. Thus, much more endeavour is needed in order to examine the aerosol/pollution effect on climate, human health and ecosystems on long-term basis.

4.5 Aerosol transport, air quality and atmospheric parameters

Quantification of the amount of aerosols transported from Sumatra to Malaysia will enable studies on the impact of aerosols on human health, radiative forcing, agricultural activities etc. A long-term record of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) measurements of the three-dimensional distribution of aerosol backscatter, extinction and depolarization ratio for both cloud-free and cloudy conditions as well as observations from remote sensing satellite sensors as shown in Table 1 can be used for this purpose. Furthermore, studies examining the air quality and atmospheric parameters have not directly considered the meteorological conditions that could affect the air-quality levels (e.g., temperature, humidity, and wind speed). Further research that focuses on the link between air quality concentrations and meteorological conditions (i.e., humidity/rainfall, wind speed and temperature) is necessary in order to shed light on the meteorological conditions that favour or not the accumulation of aerosol and pollutants over Malaysia. Meanwhile, some studies have identified the influence of atmospheric circulation patterns on seasonal air-quality

variations over Malaysia (Tan et al., 2015), revealing that the long-range transport of air pollutants does affect significantly the air quality over Peninsular Malaysia. Further research is needed in order to quantify the relative contribution of long-range transport, source strength variations and local factors that favour the pollution formation and concentrations.

4.6 Satellite data/AOD retrievals

Although during the last years several studies have used satellite remote sensing observations for aerosol monitoring over Malaysia, these observations have several limitations due to the extensive cloudiness and orbital gaps of satellite tracks. Extensive cloud cover is a common phenomenon in Southeast Asia especially during the winter monsoon season (Feng and Christopher, 2013) that reduces the availability of cloud-free images for AOD retrievals and further analysis (Kanniah et al., 2014a). Future studies should be made by integrating aerosol products delivered by various satellite sensors (Table 1) in order to obtain temporarily continuous remote sensing data, which can cover larger regions for aerosol and PM monitoring from space, also providing useful information for air-quality mapping and forecast.

5. Conclusions

In synopsis, atmospheric aerosols are of global importance because they affect the climate via direct and indirect radiative forcing and adversely impact the human health and ecosystems. The quantification of the aerosol climate implications is very uncertain due to the large spatial and temporal variability of the atmospheric aerosols, the great variety of their types, sources and sinks and mixing processes in the atmosphere. This is especially true over Malaysia, which is a crossroad of various aerosol types and pollution plumes. Therefore, comprehensive studies on the size distribution and the size-resolved chemical and mineralogical composition of aerosols over Malaysia are important i) to understand the processes governing the production, transport and removal of aerosols from the atmosphere, ii) to estimate the contribution of aerosols on the radiative forcing and to quantify their magnitude of the regional warming or cooling, and iii) to comprehend the impact of aerosols on the human health and wellbeing. Despite the fact that significant progress has been made during the last decade in aerosol/pollution monitoring over Malaysia, much more efforts are needed for a comprehensive understanding of aerosols on climate, human health and ecosystems focusing on the issues itemized above.

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Highlights:

- Aerosols affect human health, air quality and climate system of Southeast Asia
- Significant progress in aerosol monitoring over Malaysia in the last decade
- Quantification of aerosols' impact on climate is very uncertain in Malaysia
- Studies on size-resolved chemical and mineralogical composition are important

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