Estimating Particulate Matter using satellite based aerosol optical depth and meteorological variables in Malaysia

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ABSTRACT

The insufficient number of ground-based stations for measuring Particulate Matter < 10 µm (PM₁₀) in the developing countries hinders PM₁₀ monitoring at a regional scale. The present study aims to develop empirical models for PM₁₀ estimation from space over Malaysia using aerosol optical depth (AOD₅₅₀) and meteorological (surface temperature, relative humidity and atmospheric stability) data (retrieved or estimated) from Moderate Resolution Imaging Spectroradiometer (MODIS) during the period 2007–2011. The MODIS retrievals are found to be satisfactorily correlated with ground-based measurements at Malaysia. Multiple linear regressions (MLR) and Artificial Neural Network (ANN) techniques are utilized to develop the empirical models for PM₁₀ estimation. The model development and training are performed via comparison with measured PM₁₀ at 29 stations over Malaysia and reveal that the ANN provides slightly higher accuracy with $R^2 = 0.71$ and RMSE = 11.61 µg m⁻³ compared to the MLR method ($R^2 = 0.66$ and RMSE = 12.39 µg m⁻³). Stepwise regression analysis performed on the MLR method reveals that the MODIS AOD₅₅₀ is the most important parameter for PM₁₀ estimations ($R^2 = 0.59$ and RMSE = 13.61 µg m⁻³); however, the inclusion of the meteorological parameters in the MLR increases the accuracy of the retrievals ($R^2 = 0.66$, RMSE = 12.39 µg m⁻³). The estimated PM₁₀ concentrations are finally validated against surface measurements at 16 stations resulting in similar performance from the ANN model ($R^2 = 0.58$, RMSE = 10.16 µg m⁻³) and MLR technique ($R^2 = 0.56$, RMSE = 10.58 µg m⁻³). The significant accuracy that has been attained in PM₁₀ estimations from space allows us to assess the pollution levels in Malaysia and map the PM₁₀ distribution at large spatial and temporal scales.

1. Introduction

Air pollution has become a serious environmental problem in the developing southeast Asian countries, especially in Malaysia that is aiming to become an industrial nation by 2020. Malaysia is ranked as the 117th worst country among 180 nations worldwide in terms of air quality (EPI, 2016) with the major sources of air pollutants to be open biomass burning, motor vehicles and industries (Afroz et al., 2003; Hyer et al., 2013; Alias et al., 2014; Khan et al., 2016a). Besides the local sources, biomass-burning aerosols from wildfires in Indonesia are also transported over Malaysia particularly during the dry season and southwest monsoon (Kanniah et al., 2014a; Khan et al., 2016b). Furthermore, Malaysia 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. The air pollution levels in Malaysia are expressed via the Air Pollution Index (API) that is calculated using surface concentrations of carbon monoxide (CO), tropospheric ozone (O₃), nitrogen dioxide (NO₂), sulphur dioxide (SO₂) and Particulate Matter < 10 µm (PM₁₀) (Awang et al., 2000; Dominick et al., 2012).

PM₁₀ is an important component of air-pollution monitoring networks since it affects human health by causing respiratory problems (Balakrishnan et al., 2002; Pope et al., 2011a, b; Ha Trang and Tripathi, 2014), while inhaling PM₁₀, and most particularly Particulate Matter < 2.5 µm (PM_{2.5}), can cause cardiovascular diseases (Dominici et al., 2006), birth defects and premature death (Ballester et al., 2010). PM₁₀, as aerosol particles, can also affect the climate system by scattering and absorbing the incoming solar radiation resulting in heating and/or cooling effects (Ramanathan and Carmichael, 2008) depending on the type of particles (i.e. dust, soot or anthropogenic pollution), optical and

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chemical properties.

Satellite remote sensing has been increasingly available for PM₁₀ monitoring studies over a large spatial domain. Remote-sensing retrievals of aerosol optical depth (AOD) from multiple satellite sensors, such as Multiangle Imaging Spectroradiometer (MISR) (Liu et al., 2007; Van Donkelaar et al., 2010; Dey et al., 2012; Sotoudeheian and Arhami, 2014), Spinning Enhanced Visible and Infrared Imager (SEVIRI) (Emili et al., 2010), Moderate Resolution Imaging Spectroradiometer (MODIS) (Gupta et al., 2006; Van Donkelaar et al., 2010; Jamil et al., 2011; Nordio et al., 2013; Yap and Hashim, 2013; Chitranshi et al., 2014), Medium Resolution Imaging Spectrometer (MERIS) (Kaskaoutis et al., 2010; Kanniah et al., 2014b; Belocconi et al., 2016) and Landsat (Nguyen and Tran, 2014; Nadzri et al., 2010) are commonly used to estimate PM₁₀ and/or PM_{2.5} from space.

Among the available remote-sensing data, MODIS AOD is found to have high retrieval accuracy over land (i.e. $\pm 0.05 \times \text{AOD}$ under clear skies and $\pm 0.15 \times \text{AOD}$ under moderately cloud-contaminated atmospheres), as well as nearly daily global coverage (Remer et al., 2008). Validation of MODIS AOD retrieved from both Terra and Aqua satellites showed high correlation ($R^2 = 0.9$) with AERONET AODs worldwide (Levy et al., 2010) and, over Malaysia ($R^2 = 0.55$) in particular (Kanniah et al., 2014a). Nevertheless, a main drawback for usage of MODIS AOD is its coarse spatial resolution (10 km) that may tend to generalize AOD information spatially. Therefore, especially over urban environments that are characterized by heterogeneous surfaces, satellite AOD of higher spatial resolution is preferred for PM₁₀ estimations. In this respect, MERIS AOD and synergized MERIS/AATSR data (North et al., 2009) have been recently used although MERIS's relatively poor performance compared to MODIS (Kaskaoutis et al., 2010; Benas et al., 2013; Kanniah et al., 2014b). Furthermore, MODIS-AOD retrievals at 3-km spatial resolution are also considered as not so suitable for air-quality studies due to improper characterization of the underlying urban surfaces that resulted in significant bias for AODs > 0.1 (Levy et al., 2013; Munchak et al., 2014; Xie et al., 2015).

Several studies have used only the satellite AOD via simple linear regressions for PM estimations from space resulting in correlation coefficients around 0.52 to 0.77 between measured and estimated PM₁₀ concentrations (Gupta and Christopher, 2008; Schaap et al., 2009; Kanniah et al., 2016). According to Gupta and Christopher (2009a, b), AOD alone is not a good predictor of PM, since AOD corresponds to the total columnar aerosol, whereas PM refers to particle concentration at the surface; therefore, the correlation between PM and AOD strongly depends on the vertical distribution of aerosols and several meteorological factors, i.e., profiles of potential temperature and wind regime (Chitranshi et al., 2014; Sinha et al., 2015). On the other hand, the satellite snap-shots usually cover large spatial areas and are subject to cloud contamination (Zhang et al., 2005; Gupta et al., 2006). Therefore, incorporation of other meteorological parameters, such as ambient relative humidity (RH), fractional cloud cover and mixing layer height, improved the prediction of PM, since weather conditions can greatly affect the aerosol dilution and dispersion processes (Gupta and Christopher, 2009b; Benas et al., 2013; Chitranshi et al., 2014). More specifically, RH influences the hygroscopic growth of the particles (Benas et al., 2013), while strong wind speeds affect the PM dispersion (Xiao, 2011) or contribute to additional emissions over arid terrains (Rashki et al., 2012). Furthermore, inclusion of surface temperature for PM estimates has been found to significantly improve the results (Chitranshi et al., 2014; Benas et al., 2013), since temperature may modulate the photochemical reactions and play a major role in the boundary-layer dynamics that control the PM concentrations at the ground via dilution and/or trapping of pollutants (Dumka et al., 2015). In this respect, the atmospheric stability index and boundary layer height (BLH) have been used as surrogates for atmospheric stability resulting in more accurate estimates of PM since the vertical variability of aerosol can be determined (Rohen et al., 2010; Emili et al., 2010). A literature overview (e.g. Hoff and Christopher, 2009) shows that

various techniques, such as simple linear regressions (Wang and Christopher, 2003; Engel-Cox et al., 2004), multiple linear regressions (Kanniah et al., 2014b; Chitranshi et al., 2014; Gupta and Christopher, 2009a), nonlinear regressions (Sotoudeheian and Arhami, 2014; Benas et al., 2013), mixed effects models (Kloog et al., 2012; Nordio et al., 2013; Yap and Hashim, 2013; Hu et al., 2014; Xie et al., 2015; Belocconi et al., 2016), statistical and chemical transport models (Van Donkelaar et al., 2010), as well as complex nonlinear regressions such as Artificial Neural Network (ANN) (Kanniah et al., 2014b; Gupta and Christopher, 2009b) have been applied for PM estimations from space at local, regional and global scales. These advanced statistical techniques significantly improve the prediction capability of the simple AOD-PM relationships that were initially used for PM estimation from space. The continuous renewal and improvement of the databases, methods and techniques for PM monitoring from space justifies the high importance that such studies have for air-pollution mitigation efforts and human-health perspectives, especially in the developing countries (Snider et al., 2016).

In Malaysia, the most widely measured air-quality parameter is the PM₁₀. However, the PM₁₀ monitoring network is rather sparse accounting for 74 stations over the whole Malaysian territory of 330,290 km² and, therefore, PM₁₀ estimates from space are considered as absolutely necessary for warning the local population about the air-quality standards. Nevertheless, no studies covering the entire Malaysian territory have been conducted to map PM₁₀ from space. A recent study by Yap and Hashim (2013) attempted to estimate PM₁₀ over Peninsular Malaysia (only) via a mixed-effects model, which took into consideration the monthly variability of the coefficients (i.e. slope and intercept) in association with MODIS-AOD and PM₁₀. The use of daily measurements of AOD allows for better assessment of space-time interactions than models that only have spatially resolved time invariant land use terms (Kloog et al., 2012) because the relationship between AOD and PM_{2.5/10} varies on daily basis due to differences in mixing height, RH, particle composition, vertical profiles etc. (Lee et al., 2011; Sinha et al., 2015). Moreover, the PM₁₀ concentrations exhibit a remarkable intra-seasonal variation at bi-weekly frequencies of 10–20 days (Juneng et al., 2009) over Malaysia and, therefore, the existing model may discount the PM₁₀ daily variation and limits the possibility of studying the acute effects of air pollution. Furthermore, the mixed-effects model may introduce uncertainties with the assumption of linearity and using AOD as the single predictor (Xie et al., 2015). Finally, the model assumes that there is little spatial variability in the relationship, which is not necessarily true especially when the modelling domain gets larger (Hu et al., 2014) as in our study.

Thus, for improving the accuracy of PM₁₀ estimates, the present study integrates atmospheric/meteorological parameters along with advanced statistical methods, i.e., multiple linear regressions (MLR) and ANN, for PM₁₀ monitoring from space for the first time over the whole Malaysian territory. Although a better knowledge has been gained during the last years on the spatial and temporal evolution of aerosols and pollutants over Peninsular Malaysia (Khan et al., 2016a, b), monitoring the aerosol loading and composition over the East Malaysia (Borneo Island) remains a real challenge and not so well documented. There are only 15 ground monitoring stations in this region compared to its size of 198,080 km². Thus, PM₁₀ estimations via satellite remote sensing is very essential in this region, which is strongly affected by pollution especially during the southwest monsoon. The seasonality of the spatial distribution of PM₁₀ is provided at 10 × 10 km spatial resolution during 2007–2011 and is discussed against the potential sources of aerosol and pollutants. The PM₁₀ estimates are validated against ground-based measurements from several stations, while the results are useful for air quality, meteorological and health studies as well as for the calculation of the Environmental Performance Index (EPI) over Malaysia.

2. Study area

Malaysia is a developing country in southeast Asia that undergoes rapid growth in industrial and transportation domains resulting in increase in air pollution and PM₁₀ concentrations during the last decades (Afroz et al., 2003; Jamil et al., 2011). The regional meteorology is characterized by four seasons, namely dry season (June–September), wet season (November–March) and two inter-monsoon seasons (April–May and October, respectively), which are examined separately due to differences in aerosol loading and characteristics. During the dry season, south-westerly winds blow from Indonesia (Sumatra and Kalimantan) carrying biomass-burning aerosols due to extensive forest and agricultural fires (Abas et al., 2004). In contrast, lower levels of aerosol loading are recorded during the rainy season as a result of wet deposition and rainy washout. The winds are mostly from northeastern directions traversing the southern China Sea, while during the two transition seasons, shift of the wind occurs.

3. Data and methodology

3.1. Ground-based datasets

In Malaysia, the PM₁₀ measurements are collected, processed, analyzed and distributed by a private company known as Alam Sekitar Malaysia Sdn Bhd (ASMA) appointed by the Department of Environment (DOE) and the Malaysian Meteorological Department (METMalaysia). Currently, there are 74 air-quality monitoring stations nationwide (52 continuous air-quality monitoring (CAQM) stations operated by ASMA and 22 stations run by METMalaysia). For the purpose of this study, the following ground-based datasets were used:

3.1.1. PM₁₀ data

The PM₁₀ concentrations were obtained from the DOE Malaysia covering the period 2007–2011 and correspond to hourly data measured by the beta attenuation monitor (BAM1020) at the 52 stations operated by ASMA. However, only 45 stations (Fig. 1) were selected to be used in this study due to long-term data missing in the rest of the stations. BAM1020 is used for air-quality monitoring in Malaysia since it is capable to record PM₁₀ concentrations (in mg m⁻³ or µg m⁻³) on hourly basis by using the beta ray attenuation technique with beta source the ¹⁴C (< 2.22 × 10⁶ Bq) and half-lifetime of 5730 years (Md.Yusof et al., 2010). Besides that, BAM1020 provides highest level of performance since its accuracy exceeds the U.S. EPA Class III PM_{2.5} FEM standards for additive and multiplicative bias, while it has low maintenance cost. The instrument operates at a flow rate of 16.7 l per minute using a glass fiber filter tape while the measurement resolution

is 0.24 µg in 1 mg range (Met One Instruments, Inc., 2013). BAM1020 is subjected to errors due to unresolved leaks, such as flow leak or nozzle leak, which need to be kept clean especially in humid areas. The uncertainty of the BAM-1020 measurements is ~16%, which meets the requirement (25%) of the Guide to the Demonstration of Equivalence of Ambient Air Monitoring Methods (GDE). The hourly PM₁₀ concentrations were analyzed and screened to include the measurements within ± 30 min of the Terra satellite overpass (10.30 am Local Standard Time, LST), and then, were correlated with the MODIS-AOD₅₅₀ retrievals. The PM₁₀ measurements from 29 stations were used for the model (MLR and ANN) development and training, while the rest 16 stations were used for the validation of the PM₁₀ estimates from space. Both the “training” and “validation” stations were selected in a way to cover the whole Malaysian territory, i.e., Peninsular Malaysia and Island Malaysia – north part of Borneo – (left and right panels of Fig. 1, respectively), as well as the different meteorological zones of the country and atmospheric environments like urban, rural, industrial, coastal in order to be as representative as possible (see Supplementary Fig. 1 for more details about the land use types around each station). In this way, the group of the “training” stations that was used for the MLR and ANN model developments is as representative as possible for the whole country, while the same exists for the group of stations that used for the model validation. In this way, we also attempted to limit the method's performance and the model's dependence on the selected stations. Initially, the selection of the “training” and “validation” stations was region-specific, i.e., the stations in the Peninsular Malaysia were used for the model training and those in the island Malaysia for the model validation, resulting in similar statistics about model training but lower accuracies in model validation. Therefore, the current selection of the “training” and “validation” stations seems to be more representative for the whole Malaysian territory.

3.1.2. Meteorological data

Besides PM₁₀ concentrations, the CAQM stations also provide recordings of several other air pollutants such as CO, tropospheric O₃, NO₂, SO₂ and meteorological parameters like wind speed, wind direction, ambient temperature and RH. The meteorological data that were used in the analysis correspond to hourly values during the period 2007–2011. Since the ground-based measurements are not able to cover the entire Malaysia, even interpolation cannot provide a promising result (Peng et al., 2006), satellite observations were mostly used. Thus, the measured ambient temperature and RH data were used for validation of the satellite retrievals and to cross check the satellite estimations of the surface temperature and RH. Similarly to the PM₁₀ data, the ambient temperature and RH were hourly-averaged around the Terra overpass (± 30 min). Table 1 includes the meteorological

Fig. 1. Locations of the 45 PM₁₀ monitoring stations that were used for training (black dots) and validating (red dots) the empirical models developed in this study.

Table 1

Meteorological conditions, PM₁₀ concentrations (ground based stations) and Terra-MODIS retrievals of AOD at 45 meteorological stations in Malaysia for different seasons during 2007–2011. The range of the values are shown in parentheses.

Parameter	Dry season (Jun–Sept)	Wet season (Nov–March)	April–May	October
Temperature	29.65 ± 0.67 (28.21–30.86 °C)	28.07 ± 1.03 (25.61–29.90 °C)	29.64 ± 0.74 (28.10–31.15 °C)	28.70 ± 0.77 (26.97–30.98 °C)
Relative humidity	72.51 ± 4.12 (65.94–79.35%)	74.85 ± 6.13 (54.12–86.03%)	73.33 ± 4.38 (67.41–83.41%)	74.53 ± 3.47 (64.62–79.41%)
Wind speed	6.11 ± 1.05 (4.40–8.84 m/s)	6.06 ± 1.49 (4.59–9.18 m/s)	6.18 ± 1.44 (1.46–7.51 m/s)	5.99 ± 1.00 (3.17–7.16 m/s)
Wind direction	155.13 ± 50.28° (24.75–214.24°)	144.87 ± 41.95° (45.88–201.12°)	147.67 ± 37.03° (46.87–202.57°)	180.50 ± 32.74° (112.23–254.22°)
PM ₁₀	50.02 ± 23.25 (23.67–278.49 µg m ⁻³)	34.19 ± 14.03 (18.33–169.41 µg m ⁻³)	38.24 ± 13.36 (23.03–158.67 µg m ⁻³)	43.65 ± 11.39 (27.84–214.77 µg m ⁻³)
MODIS-AOD ₅₅₀	0.52 ± 0.10 (0.10–1.89)	0.41 ± 0.23 (0.06–1.81)	0.42 ± 0.21 (0.10–1.80)	0.59 ± 0.32 (0.10–1.90)
Stability (k-index)	305.22 ± 25.51 (282.35–320.64 K)	307.01 ± 21.59 (286.22–321.59 K)	310.74 ± 5.62 (284.31–320.64 K)	304.38 ± 43.24 (292.59–320.64 K)

conditions, along with PM₁₀ concentrations and Terra-MODIS retrievals above the 45 meteorological stations in the four seasons during 2007–2011, in order to reveal the seasonality in meteorological, atmospheric and pollution conditions over Malaysia. The measured wind direction is apparently different in every season and inclusion of this data in the model may improve the prediction of PM₁₀. However, spatial distribution of such data covering entire Malaysia is currently not available, thus only temperature, relative humidity, k index and MODIS AOD₅₅₀ were included in the model development.

3.2. Satellite datasets

Various products from Terra-MODIS observations over Malaysia were used for the objectives of the current work. MODIS sensor is flown on Terra and Aqua satellites since December 1999 and July 2002, respectively. Due to the wide swath of 2330 km, Terra and Aqua satellites cover the entire Earth surface every 1 to 2 days providing aerosol and atmospheric retrievals at 36 spectral bands from VIS to NIR (0.41–15.0 µm) with a spatial resolution ranging between 250 m and 1000 m by means of two separate algorithms over land and ocean (Levy et al., 2010; Munchak et al., 2014). Due to its acceptable accuracy (0.05 ± 0.15 × AOD), Level 2 (MOD04_L2, 10 × 10 km spatial resolution) Terra-MODIS AOD retrievals at 550 nm (AOD₅₅₀) from Collection 5 (version C005.1) downloaded from <http://ladsweb.nascom.nasa.gov/> on daily basis were used in this study during the period 2007–2011. The L2 Terra-MODIS AODs centered above the PM₁₀ stations were correlated with the measured PM₁₀ concentrations for developing models for PM₁₀ monitoring from space. Terra-MODIS atmospheric profile products (MOD07) at 5-km spatial resolution were also used on daily basis in order to obtain the atmospheric stability index (k-index) and surface temperature. The k-index was utilized to estimate the vertical temperature lapse rate and atmospheric static stability that can be used as proxy for the vertical mixing height (Benas et al., 2013). In general, the k-index was derived by using the infrared temperature and moisture profile retrievals in MOD07_L2 product and its values range from 268 K to 340 K (NASA, 2015). Especially over Malaysia, the range of k-index was found to be ~287–324 K. The surface temperature was used in the model development for PM₁₀ estimations, since it affects the photochemical reactions and controls the temperature inversions that influence the aerosol concentrations near the surface (Gupta and Christopher, 2009a). The MOD07_L2 product contains retrieved vertical profiles of atmospheric temperature and moisture at 20 vertical levels (Seeman et al., 2006). A clear-sky synthetic regression has been used to estimate the surface skin temperature, while the regression coefficients have been derived using a fast radiative transfer model and atmospheric characteristics from

global datasets of radiosonde and model profiles (Seeman et al., 2006). Regression between surface skin temperature measured at Atmospheric Radiation Measurement sites using the downward-looking infrared thermometer and surface air temperature measured using radiosonde was used to estimate/assign the surface skin temperature to all profiles.

The surface temperature data were also used as one of the input parameters for computing the saturation vapour pressure in a way to get the spatial distribution of RH (see Section 3.3). RH needs to be computed due to limitation of the meteorological data's spatial coverage and due to the fact that it is not a direct product from the satellite observations. The RH is included in the codes for PM₁₀ estimations in order to parameterize the hygroscopic growth of particles that modifies the aerosol scattering efficiency and, in turn, the AOD (Wang and Martin, 2007).

As stated above, RH cannot be obtained as a direct satellite product, thus a computation is required (Peng et al., 2006). In this respect, MODIS level 1B calibrated radiances (MOD021km) were used to retrieve the water-vapour content at NIR via observations at several bands, acting either as non-absorption bands (i.e. band 2 (0.865 µm) and band 5 (1.24 µm)) or as absorption bands (bands 17 (0.905 µm), 18 (0.936 µm) and 19 (0.940 µm)) (Gao and Kaufman, 2003). Band 17 is a weak absorption channel, which is suitable for retrievals of water-vapour content at very humid environments, while the strong absorption channel (band 18) is really suitable for dry areas. These five bands, along with surface temperature data, were used as inputs to compute RH using the formulas developed by Peng et al. (2006) as detailed out in the methodology Section 3.3. Similarly to AOD₅₅₀ and MOD07 retrievals, the MOD021km products were also obtained on daily basis during 2007–2011.

The surface-elevation data from the Shuttle Radar Topography Mission (SRTM) gridded product with 90 m spatial resolution was used to calculate the air pressure (P_a), since several air pressure data from MODIS (MOD07) retrievals were missing due to cloud cover. The air pressure data are necessary for the estimates of RH, as shown in the next section.

3.3. Methodology

3.3.1. Pre-processing

Fig. 2 shows the flow chart of the overall methodology adopted for the estimates of PM₁₀ concentrations from space over Malaysia. Initially, we geo-referenced the MODIS AOD₅₅₀ and the other inputs such as surface temperature, atmospheric stability and RH to geographic latitude/longitude (WGS84) coordinates. The coordinate system was used by the Global Positioning System and it is intended to be located at the equator.

Fig. 2. Flowchart of the overall methodology for PM₁₀ estimation from space over Malaysia.

3.3.2. Processing

3.3.2.1. AOD extraction. Initially, the AOD₅₅₀ values from Terra-MODIS retrievals were extracted at 10 × 10 km. Malaysia is a very challenging environment for PM₁₀ estimates from space because of extensive cloudiness and data missing. To resolve this problem, the spatial-averaged values over a 5 × 5 pixels domain (50 × 50 km) were used in order to fill the AOD₅₅₀ data series in the case of missing data (Ichoku et al., 2002); if none of the pixels in the 5 × 5 domain has values, these pixels were excluded from the analysis. This technique helped to improve the AOD₅₅₀ data availability (Kanniah et al., 2014a; Yap and Hashim, 2013).

3.3.2.2. Relative humidity computation. The RH was calculated by using the MOD021km product, particularly bands 2 (0.865 μm), 5 (1.24 μm), 17 (0.905 μm), 18 (0.936 μm) and 19 (0.940 μm), the MOD07 retrievals of surface temperature, and the elevation data from SRTM via the equations provided by Peng et al. (2006) (Eqs. (1)–(3)). The calculations were performed using a python script in ArcGIS environment.

$$RH = e/e_s \quad (1)$$

$$e_s = 611 \exp\left(\frac{17.27T_a}{237.3 + T_a}\right) \quad (2)$$

$$e = Q * P_a / 0.622 \quad (3)$$

where,

- RH: relative humidity
 e: vapour pressure
 e_s: saturation vapour pressure
 T_a: surface air temperature
 P_a: air pressure
 Q: specific humidity

As stated above, the MODIS-AOD₅₅₀ retrievals are in spatial resolution of 10 × 10 km, while the rest of the MODIS-derived meteorological parameters (i.e. surface temperature, RH, atmospheric stability) are considered at 5 × 5 km spatial resolution. Therefore, the meteorological parameters were resampled to 10 km to match the MODIS AOD₅₅₀ using the Nearest Neighbor resampling technique. This resampling technique was used because it selects the closest value to the center of the output cell and it does not alter the value of the image cells. Besides that, this simple technique is acceptable to be used to

aggregate the spatial resolution since both datasets have the same temporal resolution. For the RH estimations, the T_a was obtained from MODIS retrievals, while the P_a from the SRTM gridded product at 10 × 10 km resolution. The specific humidity (Q) depends on precipitable water vapour (PW) where PW was computed using the MOD021km product. Finally, we extracted the Terra-MODIS daily data of AOD₅₅₀, k-index, surface temperature and RH above the PM₁₀ and meteorological stations for validation of our retrievals against measured data. These datasets have been used as inputs to develop empirical models for the estimation of PM₁₀ via advanced statistical techniques. It should be noted here, that since RH has been estimated from the surface temperature retrievals, it is dependent on them and, therefore, cannot be considered as an Independent and Identically Distributed (IID) variable. However, it is used as input for the development of the MLR and ANN models although its inclusion in the multiple linear regressions may cause “co-linearity” phenomenon, but does not affect the model validation results.

3.3.2.3. Regression analysis. Statistical analysis has been performed using both multiple linear regressions (MLR) and non-linear Artificial Neural Network (ANN) regressions in order to attain more accurate PM₁₀ estimations using AOD₅₅₀ and meteorological parameters as variables. In the linear regression technique, the stepwise approach was applied, which analyses the variables one-by-one and via this way each of the variable's performance or its contribution to the overall model can be easily determined (Benas et al., 2013; Chitranshi et al., 2014). Non-linear techniques are usually used when linear parametric models produce misleading results and high errors (Benas et al., 2013). On the other hand, the Multiple Layer Perceptron (MLP) approach with a back-propagation algorithm was adopted in the ANN non-linear technique. With the development of artificial intelligence technologies, several studies dealing with environmental issues have highlighted the capability of ANN to give accurate predictions of particulate pollution, dust storm detection and AOD retrievals (e.g. Gupta and Christopher, 2009b; Roy, 2012; Yao et al., 2012; Elossta et al., 2013; Han and Sohn, 2013; Xiao et al., 2015). In designing the ANN model, important parameters have to be carefully decided, especially the number of the neurons in the hidden layers, learning rate, momentum and stopping criteria. The ANN consists of interconnected neurons that simplify the non-linear mapping between each set of inputs, as shown in Fig. 3. The MLP is a feed-forward ANN model with at least one layer of hidden neuron whose activation function operates as universal function approximator. With enough

Fig. 3. Architecture of the Multi-Layer Perceptron (MLP) Artificial Neural Network model used for PM₁₀ estimations.

hidden neurons and training data, the ANN models are capable of simulating the mathematical formulas between inputs and outputs (Xiao et al., 2015). In our case, this model consists of three layers (i.e. input, hidden and output) and the sigmoid transfer function (TanH) was used in hidden and output layers. The input layer is composed of four nodes, AOD₅₅₀, RH, k-index and surface temperature derived from MODIS, as predictors, while the output layer is the estimated PM₁₀ (Fig. 3). In this study we used 3 hidden nodes to transform the inputs into output layers. Initially, we started with one hidden node and increased it until optimum results are obtained. The 3 hidden nodes seem to be adequate for the purpose of this study since we have only four inputs (Fig. 3). So far, there is no specific rule to create hidden node even one hidden node is enough for a simple task. However, the usage of many hidden nodes or more complicated models may lead to more precise outputs. In this study, the learning rate and the momentum were fixed at 0.1; the learning rate should be in the range of 0–1, but learning rate values close to 1 will lead to overfit of the data. Momentum is a parameter used to prohibit the model from converging to local minima and high momentum values are able to speed up the convergence of the model. But by setting the momentum too high will cause overshooting of the minimum, which means that the model becomes unstable. Moreover, we specified the number of maximum training iterations as the stopping criteria, meaning that training will stop once the maximum number of iterations is exceeded. The ANN model was trained with 500 iterations and 20 tours; tour is the number of times for restarting the fitting process, since each iteration uses a different random starting point. The algorithm used for the MLP training is based on the steepest descent gradient approach, which is applied in order to minimize the instantaneous error (Ham and Kostanic, 2001; Xiao et al., 2015). According to Gupta and Christopher (2009b), the MLP technique is commonly used in various atmospheric problems that showed directionality of information processing inside the neural network architecture as shown in Fig. 3. However, ANN is expected to provide better predictions because it is capable to analyse the pattern, and at the same time, it minimizes the error functions resulting in improved outputs. The developed MLR and ANN models were calibrated against PM₁₀ measurements at 29 stations distributed all over Malaysia during the period 2007–2011 (see Fig. 1 and Supplementary Fig. 1).

3.3.2.4. Model validation. At the final stage, the PM₁₀ estimations obtained from the developed MLR and ANN models over Malaysia during 2007–2011 were validated against measured PM₁₀ concentrations from 16 stations (Fig. 1). For assessing the model's accuracy in estimating the surface PM₁₀ concentrations, the linear regression analysis and statistical indicators like the Root Mean Square

Error (RMSE) and Mean Bias Error (MBE) were used. The RMSE shows the overall performance of the model in estimating the measured data, while the MBE determines the overestimation or underestimation of the measured PM₁₀ concentrations:

$$\text{RMSE in } \mu\text{g m}^{-3} = \sqrt{\frac{\sum_{i=1}^N \text{PM}_{10m} - \text{PM}_{10e}^2}{N}} \quad (4a)$$

$$\text{RMSE (in\%)} = \frac{\text{RMSE (in } \mu\text{g m}^{-3})}{\sum_{i=1}^N \text{PM}_{10m} / N} \times 100 \quad (4b)$$

$$\text{MeanBiasError (MBE)} = \frac{1}{N} \sum_{i=1}^N \frac{\text{PM}_{10e} - \text{PM}_{10m}}{\text{PM}_{10m}} \quad (4c)$$

$$\text{MBE (in\%)} = \frac{\text{MBE (in } \mu\text{g m}^{-3})}{\sum_{i=1}^N \text{PM}_{10m} / N} \times 100 \quad (4d)$$

where, the subscripts m and e stand for measured and estimated PM₁₀ concentrations respectively, while N is the total number of data used in the comparisons. Moreover, the Index of Agreement (Willmott, 1981, Willmott et al., 2012) was used to further ensure the model's performance against the surface PM₁₀ measurements as:

$$d = 1 - \frac{\sum_{i=1}^n (P_i - O_i)^2}{\sum_{i=1}^n (|P_i - \bar{O}| + |O_i - \bar{O}|)^2}$$

where P_i and O_i are the estimated and measured PM₁₀ concentrations, respectively, while the $\bar{O}$ is the mean value of the measured PM₁₀. Values close to 1 indicate perfect model performance.

4. Results and discussion

Before presenting the results of PM₁₀ estimates and validation efforts over Malaysia the spatial distributions (on seasonal basis) and the validation of parameters like AOD₅₅₀, RH and surface temperature that have been used in the intermediate stages for the PM₁₀ estimates (Fig. 2) are examined.

4.1. Terra-MODIS AOD₅₅₀

The seasonal-mean AOD₅₅₀ spatial distributions over Malaysia for the dry and wet seasons are shown in Fig. 4a–d. During the dry season, the AOD₅₅₀ exhibits high values (~0.5–1.8) over the highly-populated and industrialized regions of the west and south coast of Peninsular Malaysia, while even higher AOD₅₅₀ (0.8–1.8) is shown over the central-southern parts of Sabah and Sarawak in Island Malaysia (Fig. 4a) due to extensive forest and agricultural fires (Reid et al., 2013). These AODs are at the same magnitude with extreme values observed over environments strongly affected by seasonal forest fires, such as Amazonia (Kambezidis and Kaskaoutis, 2008), African savanna (Eck et al., 2003) northwestern Indo-Gangetic Plains (Kaskaoutis et al., 2014) and Russia (Chubarova et al., 2012). Very high AOD₅₅₀ values above several urban and suburban stations, such as Seri Manjung (0.96 ± 0.49), Perai (0.80 ± 0.31), Tawau (0.82 ± 0.21) were observed during the dry season (Supplementary Fig. 2), which are attributed to heavy traffic and industrial activities along with transported smoke plumes (Kanniah et al., 2014a). In general, the AOD₅₅₀ spatial distribution exhibits lower values during the wet season (Fig. 4b), except for the highly-polluted urbanized sites along the west coast and southern part of Peninsular Malaysia where the AODs remain very high, as well as over USM Penang (Fig. 1) with AOD₅₅₀ = 0.67 ± 0.16 . During the wet season, entire Malaysia faces extreme weather conditions with heavy rainfall, especially in the east coast of Peninsular Malaysia and in Sabah and Sarawak (Island Malaysia). However, some extreme AODs (> 1.4) are observed over

Fig. 4. Seasonal-mean spatial distribution of MODIS AOD₅₅₀ (10 × 10 km) over Malaysia in the dry (Jun–Sep) season (a), wet (Nov–Mar) season (b), and the two inter-monsoon seasons, April–May (c) and October (d) during 2007–2011.

certain areas in southern Sabah. It should be noted here that due to the lower data availability as a consequence of the extensive cloudiness and cloud contamination during the wet season, the uncertainty in AOD₅₅₀ increases compared to the dry season.

During the inter-monsoon (April–May) season (Fig. 4c), the spatial distribution of AOD₅₅₀ is somewhat similar to that observed in the wet season, with a tendency of increasing AOD₅₅₀ over some parts of Sabah and Sarawak due to lower moisture and beginning of the agricultural fires (Hyer et al., 2013). However, the most sites located in Sabah and Sarawak (see Fig. 1) exhibit seasonal-mean AOD₅₅₀ mostly below 0.5, especially the rural sites of Kota Samarahan (0.26 ± 0.15) and Kapit (0.17 ± 0.09) (Supplementary Fig. 2). In contrast, several sites around the industrial and metropolitan cities, such as Nilai, Balok, Petaling Jaya, Kuantan, USM Penang and Kota Bharu (Fig. 1) exhibit much larger AOD₅₅₀ values (0.6–0.9) compared to the rest of the stations (Supplementary Fig. 2). The spatial distribution of AOD₅₅₀ during October (Fig. 4d) indicates that almost the entire Malaysia is covered by a thick aerosol layer with AOD₅₅₀ of 0.6–> 1.0 due to accumulation of aerosol and pollutants, mostly originated from forest and agricultural fires and urban/industrial emissions, during the long dry season (Khan et al., 2016b). Some stations like Shah Alam recorded the highest AOD₅₅₀ value of 1.40 ± 0.09 followed by several stations with mean AOD above 0.8 like Tasek Ipoh (1.18 ± 0.39), Miri (1.00 ± 0.04), Tawau (0.98 ± 0.2830), Petaling Jaya (0.92 ± 0.02) and Klang

(0.83 ± 0.15), while numerous other stations exhibit AOD₅₅₀ in the order of 0.4–0.5 (Supplementary Fig. 2). Similarly to the other seasons, the stations with the highest AODs are located in urban and industrial areas, thus exposed to local sources like vehicle and industrial emissions and open biomass burning (Afroz et al., 2003). On seasonal-mean basis, the MODIS AOD₅₅₀ is considered as quite representative of the AERONET AOD. In a previous study, Kanniah et al. (2014a) validated the Terra-MODIS AOD retrievals against AOD data from five (Kuching, Tahir, Songkhla, USM Penang and Singapore) AERONET stations over Malaysia for the period 2007–2011 revealing a satisfactory agreement ($R^2 = 0.55$).

4.2. Relative humidity

The seasonal-mean spatial distributions of the RH (10 × 10 km spatial resolution) calculated using Eqs. (1)–(3) are shown in Fig. 5a–d. In the dry season, the RH spatial distribution generally exhibits values < 70%, except for forested and mountainous regions in the center of Peninsular Malaysia and in the mangrove areas in south and northern coastal regions (Fig. 5a). The RH remarkably increases during the wet season (Fig. 5b), since the east coast of Peninsular Malaysia, Sabah and Sarawak regions exhibit RH values usually above 70–80% due to heavy rainfall (Hamdan et al., 2010; Abdullah et al., 2011); Sabah and Sarawak (right plots) also exhibit high RH values in the

Fig. 4. (continued)

range of 60–90% in the forested areas even in the dry season. Light winds, which scarcely exceed 10 knots, and variable wind directions are responsible for the relatively high RH values that remained about ~70–80% in most of the areas over Malaysia during the inter-monsoon season (April–May, Fig. 5c) (Abdullah et al., 2011). The RH distribution during October (Fig. 5d) presents some similarities to that observed in April–May with values above 60% in Peninsular Malaysia, except for some areas in the west coast where RH is higher during October, and in the central part of Sabah and Sarawak where the RH in October is mostly below 50%. The estimated RH values using remote sensing data have been validated against the measured RH at the 29 “training” meteorological stations during 2007–2011 (Fig. 6). The comparison was performed by using the averaged RH values $\pm$ 30 min around the Terra overpass time (10:00–11:00 h, LST) in order to avoid any uncertainty from the diurnal variation of RH, while the satellite-derived RH was centered above each station at 10×10 km spatial resolution. The regression analysis shows a satisfactory agreement between measured and estimated RH values with $R^2 = 0.78$, RMSE = 5.14% and MBE = -0.06% . The data points follow the 1–1 line, while nearly all are within the observation uncertainties. The satisfactory agreement between estimated and measured RH values gives credit to the accuracy of the MODIS products and the methodology used for the RH estimations. As stated before, the calculated RH values from the MODIS retrievals do not guarantee IID characteristic for satellite-derived RH. However, the satisfactory agreement with the surface measurements

and the same range between measured and estimated RH values (Fig. 6) give further credential to the estimations.

4.3. Surface temperature

The seasonal-mean spatial distributions of the surface temperature derived from the MODIS atmospheric profile product are shown in Fig. 7a–d, revealing a range between 21.8 °C and 38.8 °C. In general, the urban and highly-populated sites in the west coast of Peninsular Malaysia (Klang, Petaling Jaya, Shah Alam and Penang) as well as the urban/industrialized centers in Sabah and Sarawak (Bintulu, Miri, Keningau, Sandakan and Kuching) exhibit higher values (35.8–38.8 °C) during the dry season compared to the rural areas and mountainous regions (Fig. 7a). Overall, the influence of the inserted surface-elevation data from SRTM is obvious in the temperature retrievals. The surface temperature during the wet season is remarkably lower than that observed in the dry season due to heavy rainfall, with values ranging between 21.8 °C and 33 °C (Fig. 7b). The lowest temperatures in all the seasons are observed at the southern parts of the Island Malaysia, along the borders with Kalimantan (Indonesia), due to elevated terrain and forested-mountainous areas. The maximum surface temperatures during the inter-monsoon (April–May and October) seasons are about 35.8–38.8 °C, with the urban regions to exhibit the highest temperatures of ~36–37 °C (Fig. 7c, d). In contrast, the mountainous areas in Peninsular Malaysia (24.8–27.8 °C) and the rural

Fig. 5. Seasonal-mean spatial distribution of estimated RH during 2007–2011 over Malaysia for (a) dry season (June–September), (b) wet season (November–March), (c) April–May and (d) October.

areas in Island Malaysia (21.8–29.8 °C) exhibit the lowest temperatures.

The MODIS-derived surface-temperature values have been validated against measured ambient temperatures at the 29 meteorological stations (Fig. 8). The correlation shows a moderate coefficient of determination ($R^2 = 0.49$), RMSE of 2.68 °C (8.73%) and MBE of -0.03 °C, and is considered quite satisfactory taking into account that the MODIS-derived surface temperature largely depends on the emissivity of the Earth's surface, whereas the measured ambient temperature (~ 2 m above ground) represents the temperature of the environment. Therefore, the surface temperature that estimated using satellite data may comprise of emissions and thermal absorption from the surface materials, surface moisture, and radiative transfer (thermals) between the surface and the atmosphere (Voogt and Oke, 2003). However, the satellite-derived atmospheric surface temperature may be comparable to the measured ambient temperature under clear skies and/or during atmospheric mixing of any surface-based inversion layers (Kensten et al., 2011). The satisfactory correlation and the fact that $\sim 80\%$ of the data points lie within the observational uncertainties support the accuracy of the methodology for satellite-derived temperature data over Malaysia and their use as inputs in the developed models.

4.4. Empirical models for PM_{10} estimation

Empirical models have been developed for PM_{10} estimations from space using MODIS AOD₅₅₀, meteorological (surface temperature, RH) and atmospheric stability parameters by means of MLR and ANN techniques. The total number of cases that satisfy the criteria (i.e.

PM_{10} measurements within ± 30 min of the Terra-MODIS overpass) for AOD₅₅₀ vs PM_{10} correlation is 1129 during 2007–2011 (463, 337, 224 and 105 during the dry season, wet season, April–May and October, respectively). The relationship between surface and columnar aerosol properties is a real challenge and can vary substantially due to variations in the aerosol source regions, emission rates and the presence of distinct aerosol layers aloft from different origin and composition (Gupta et al., 2006; Sinha et al., 2015). Furthermore, changes in the mixing-layer height that control the dilution processes may influence the accumulation of near-surface aerosols with negligible effects in the columnar (Sinha et al., 2013; Dumka et al., 2015). The correlation between PM_{10} and AOD₅₅₀ (Fig. 9) over the 29 “training” stations is considered satisfactory with $R^2 = 0.59$ suggesting that satellite AODs can be used for PM_{10} estimates over Malaysia with an acceptable accuracy. However, for AOD₅₅₀ values above ~ 0.6 and PM_{10} larger than $\sim 80 \mu g m^{-3}$ the scatter of the data points increases indicating higher uncertainty in the PM_{10} estimations. It should be noted here that similar correlation associated with 53% of the variance ($R^2 = 0.53$) was found between the MODIS AODs and the PM_{10} over the 45 stations belonging to Peninsular Malaysia, thus indicating similar relationships between PM_{10} concentrations at the ground and columnar AODs, nearly independently from the group of the selected stations.

High correlations between the surface PM_{10} concentrations and columnar AOD₅₅₀ were observed in each season, with R^2 values ranging from 0.51 (wet season) to 0.71 (October), which are much above those (R^2 between 0.16 and 0.71 and mostly around 0.3) found between surface PM_2 (PM below $2 \mu m$, measured by a Quartz Crystal

Fig. 5. (continued)

Fig. 6. Correlation between estimated and measured RH values at the 29 meteorological stations that used for model calibration over Malaysia during 2007–2011.

Microbalance instrument) and columnar (MODIS and Microtops-II) AODs over Hyderabad, India (Sinha et al., 2015). Furthermore, the $R^2 = 0.59$ (Fig. 9) is comparable or even higher than respective values obtained from several studies over the globe dealing with AOD vs PM relationship (see Table 2). Table 2 summarizes the satellite datasets and

other input parameters as well as the various techniques that have been used at several locations worldwide for PM estimations from space. Note that the accuracy of the PM_{10} estimations via both MLR and ANN techniques over Malaysia is found to be comparable or even larger compared to other studies. On the other hand, changes in the physical-chemical properties of aerosols may also affect the PM_{10} vs AOD relationship, since these properties control the scattering and absorption processes in the atmosphere and, therefore, the AOD and its spectral variation (Valenzuela et al., 2015). Especially over Malaysia, the elevated aerosol plumes (1–2 km), mostly composed by biomass-burning smoke and anthropogenic pollution transported either from southeast Asia or Indonesia (Campbell et al., 2013; Wang et al., 2013), may weaken the relationship between near-surface and columnar aerosols, thus considering the PM_{10} estimates from space a real challenge.

Although Fig. 9 revealed a satisfactory correlation between MODIS AOD_{550} and surface PM_{10} concentrations over the 29 stations used for model training, the inclusion of the meteorological parameters (surface temperature, RH, k-index) in the developing models may improve the PM_{10} estimations, as has been also seen in several previous studies (see Table 2). The developed MLR model for the PM_{10} estimations over Malaysia based on the 29 selected “training” stations is:

$$PM_{10} = 268.51 + (56.19 \cdot AOD_{550}) + (1.30 \cdot \text{surface temperature}) + (-0.86 \cdot k \text{ index}) + (-0.26 \cdot RH) \quad (5)$$

It should be noted that the selection of different group of stations (e.g., stations only over the Peninsular Malaysia) results is slight

Fig. 7. Seasonal-mean spatial distribution of surface temperature derived from MODIS during 2007–2011 over Malaysia for (a) dry season (June–September), (b) wet season (November–March), (c) inter-monsoon (April–May) and, (d) inter-monsoon (October).

modifications in the constant terms for each variable, but the final evaluation against measured PM_{10} leads to similar statistics (R^2 , RMSE, MBE). Furthermore, it should be reminded that the RH cannot be considered as IID variable, and therefore, via Eqs. (1) and (2), this model (Eq. (5)) is not linear for the surface temperature. The evaluation of this model has been performed against the measured PM_{10} concentrations at the 29 “training” stations with total availability of $N = 723$ data. The correspondence of each variable included in Eq. (5) on PM_{10} estimations was determined by the stepwise technique and the results are summarized in Table 3. The MLR technique reveals that the accuracy of the PM_{10} estimates increases when all the four variables are included in the parameterization scheme ($R^2 = 0.66$ and $RMSE = 12.39 \mu g m^{-3}$, $RMSE (\%) = 25.95\%$, $MBE = -0.0004 \mu g m^{-3}$) compared to the cases that a single variable was included (Table 3). Stepwise analysis with the individual RH values was avoided, since it depends on surface temperature.

As expected, AOD_{550} is found to have the highest correlation ($R^2 = 0.59$) and lowest RMSE ($13.61 \mu g m^{-3}$) among the variables regarding the PM_{10} estimates. The MODIS AOD_{550} results in better prediction of the PM_{10} concentrations compared to previous analysis using MERIS AOD over Malaysia (Kanniah et al., 2014b), suggesting that the coarser spatial resolution of MODIS is more suitable for estimating PM_{10} for altitudes < 2 km (Munchak et al., 2014; Levy et al., 2013). The finer resolution of AOD retrievals provides errors in the pixel selection, especially over the urban areas due to the heterogeneity of land surface, although it is capable to monitor PM_{10} at local scales (Munchak et al., 2014). The second best predictor of PM_{10} using

the MLR technique is the atmospheric stability (k-index) with $R^2 = 0.16$ and $RMSE = 19.44 \mu g m^{-3}$. The inclusion of this parameter considerably improves the PM_{10} estimations, similarly to the findings by Gupta and Christopher (2009a) who used the BLH as a surrogate to estimate the height of the aerosol layer, resulting in a slight improvement of the correlations from $R = 0.66$ to $R = 0.67$. Similarly, Benas et al. (2013) found that the inclusion of the atmospheric stability in the parameterization scheme slightly increased the R^2 from 0.67 to 0.71. According to Benas et al. (2013), the k-index considers both the horizontal and vertical aerosol mixing within the $10 km \times 10 km$ of MODIS spatial resolution, since the surface layer is generally affected by turbulent movement (unstable conditions). However, other studies (Seo et al., 2015; Emili et al., 2010) found that the BLH, which strongly affects the aerosol vertical profile, showed a better agreement with PM_{10} estimations. The RH takes place in the aerosol hygroscopic growth factor and may influence the particle composition and optical properties of aerosols, mainly the scattering process (Malm et al., 2000; Titos et al., 2014), while a negative correlation with PM_{10} was found according to the developed MLR model (Eq. (5)), similarly to that obtained by Seo et al. (2015) and Emili et al. (2010). However, as a calculated parameter via MODIS surface-temperature, the RH values may not fully explain the impact of “real RH” on PM_{10} estimations. The surface temperature may influence the photochemical reactions in the atmosphere and favors the formation of secondary aerosol (Sheehan and Bowman, 2001; Takekawa et al., 2003). Although the surface temperature exhibits very low correlation with PM_{10} ($R^2 = 0.11$, $RMSE = 20.08 \mu g m^{-3}$), other studies found significant influence of

Fig. 7. (continued)

Fig. 8. Correlation between the MODIS-derived surface-temperature product and measured temperatures at 2 m above ground at the 29 “training” meteorological stations over Malaysia during 2007–2011.

surface temperature on the PM_{10} estimates (Benas et al., 2013; Bampadimos et al., 2011). Besides that, the very high humidity levels in southeast Asia may reduce the effect of the surface temperature. In summary, the analysis reveals that the AOD_{550} exhibits a satisfactory correlation with the PM_{10} measurements, since aerosols are usually well mixed within the lower boundary layer in the absence of distinct

Fig. 9. Correlation between Terra-MODIS AOD_{550} and ground-based PM_{10} concentrations obtained at the 29 “training” stations over Malaysia.

aerosol layers aloft; however, the inclusion of the meteorological parameters and stability index improves the accuracy of the PM_{10} estimates.

As far as the ANN technique is concerned, the empirical model developed for estimating the PM_{10} concentrations based on data over the 29 “training” stations is:

$$PM_{10} = 72.599 + (39.399 \cdot H1) + (-31.944 \cdot H2) + (-30.735 \cdot H3) \quad (6)$$

Table 2
Previous studies using satellite-based AOD and other meteorological data for PM estimation from space.

Study area	Input data		Technique	R ² /R	Accuracy	Reference
	Source of AOD	Other parameter	Output			
Malaysia	Terra-MODIS	Relative humidity, surface temperature, atmospheric stability index	PM _{1.0}	Multi-linear regression	R ² = 0.66 RMSE = 12.39 $\mu\text{g m}^{-3}$ MBE = -0.0004 $\mu\text{g m}^{-3}$ d = 0.89	Current study
Malaysia	Terra-MODIS	Relative humidity, surface temperature, atmospheric stability index	PM _{1.0}	Artificial Neural Networks	R ² = 0.71 RMSE = 11.61 $\mu\text{g m}^{-3}$ MBE = -0.03 $\mu\text{g m}^{-3}$ d = 0.90	Current study
Agra, India	MODIS	Relative humidity, wind speed, atmospheric temperature	PM _{1.0}	Linear, Log linear and multiple regression	R = 0.75–0.81 Relative standard error (RSE) = 54.5–8.86%	Chitranshi et al. (2014)
Tehran	MODIS and MISR	–	PM _{1.0}	Linear and non-linear multi regression model	R ² RMSE = 22.62 $\mu\text{g m}^{-3}$ RMSE = 0.32 $\mu\text{g m}^{-3}$ RMSE = 21.70 $\mu\text{g m}^{-3}$ RMSE = 0.32 $\mu\text{g m}^{-3}$	Sotoudehian and Arhami (2014)
Delhi-Kanpur, India	MISR	–	PM _{2.5}	Linear regression	R = 0.71	Dey et al. (2012)
Hyderabad, India	Terra and Aqua MODIS	–	PM ₂	Linear regression	R ² = 0.16–0.71	Sinha et al. (2015)
Athens	MODIS and MERIS/AATSR	Relative humidity, surface temperature, k index	PM _{1.0}	Linear regression and non-linear	R ² RMSE = 9.4 $\mu\text{g m}^{-3}$ RMSE = 11.1 $\mu\text{g m}^{-3}$ RMSE = 8.29 $\mu\text{g m}^{-3}$ RMSE = 8.25 $\mu\text{g m}^{-3}$	Benas et al. (2013)
Peninsular Malaysia	MODIS	–	PM _{1.0}	Linear mixed effect	R = 0.88	Yap and Hashim (2013)
European Alpine region	MODIS and SEVIRI	Relative humidity, boundary layer height (BLH)	PM _{1.0}	Linear model	R = 0.60 0.70	Emili et al. (2010)
Athens	MERIS	–	PM _{1.0}	Differential textual analysis (DTA)	R ² = 0.85	Kaskaoutis et al. (2010)
North America	MODIS/MISR	–	PM _{2.5}	Chemical transport model (GEOS-CHEM)	R = 0.77 Bias = -1.75 $\mu\text{g m}^{-3}$	Van Donkelaar et al. (2010)
Eastern United States	MISR	GEOS-3 meteorology (relative humidity and HPBL)	PM _{2.5}	Linear model	R ² = 0.48 p = 0.01	Liu et al. (2005)
Birmingham	MODIS	–	PM _{2.5}	Linear model (daily and hourly)	R = 0.52	Gupta and Christopher (2008)
Alabama	MODIS	–	PM _{2.5}	Two-variate	R = 0.62	Gupta and Christopher (2008)
South-eastern United States	MODIS	Wind speed, relative humidity, surface temperature, cloud fraction and HPBL	PM _{2.5}	Multivariate	R = 0.60 Slope (value close to 1 indicates improvement) 0.37 0.46	Christopher (2009a)
Florida	MODIS	Latitude, longitude, month, wind speed, relative humidity, surface temperature and HPBL	PM _{2.5}	Two-variate, multivariate ANN	R = 0.67 0.77 0.83	Gupta and Christopher (2009b)
Mid Atlantic states, USA	MODIS	Temperature, wind speed, visibility, relative humidity, open space, elevation, tree density	PM _{2.5}	Mixed models regressing PM _{2.5} with AOD, meteorology and land use data	R ² = 0.81	Kloog et al., 2012
London, UK	MERIS/AATSR	Relative humidity, temperature, k index (atmospheric stability)	PM _{1.0} and PM _{2.5}	Linear mixed effects model	Mean absolute error 2.14 $\mu\text{g m}^{-3}$ 2.24 $\mu\text{g m}^{-3}$	Bellocchi et al., 2016
Lombard, Italy	MODIS	Temperature, wind speed, relative humidity, visibility, sea level pressure, open space, elevation, roads, and population density	PM _{1.0}	Mixed models regressing PM _{2.5} with AOD, meteorology and land use data	R ² = 0.95 0.86 R ² = 0.79	Nordio et al., 2013
Beijing, China	MODIS AOD (3 km)	–	PM _{2.5}	Mixed effects model	RMSE = 16.63 $\mu\text{g m}^{-3}$	Xie et al., 2015
South-eastern United States	MODIS AOD retrieved from Multi-Angle Implementation of Atmospheric	Surface temperature, relative humidity, wind speed, boundary layer height forest	PM _{2.5}	Linear mixed effects model	Root mean squared prediction error = 2.73 $\mu\text{g m}^{-3}$	Hu et al., 2014

(continued on next page)

Table 2 (continued)

Study area	Input data	Technique	R ² /R	Accuracy	Reference
	Source of AOD				
	Other parameter				
	Correction algorithm	cover, road length, elevation and point emissions			

Table 3

Determination coefficient (R^2), RMSE, MBE and index of agreement values using multiple linear regression (stepwise) implementation for PM_{10} estimations. The comparison has been made between estimated and measured PM_{10} concentrations at the 29 stations used for model training.

Parameter(s) used for PM_{10} estimation	R^2	RMSE ($\mu g m^{-3}$)	MBE ($\mu g m^{-3}$)	Index of agreement	p-Value
AOD ₅₅₀	0.59	13.61	− 0.002	0.85	1.55E − 96
Surface temperature	0.11	20.08	0.0002	0.32	1.63E − 13
Atmospheric stability (k-index)	0.16	19.44	0.003	0.44	1.64E − 20
AOD ₅₅₀ , RH, surface temperature	0.63	12.94	− 0.035	0.87	1.66E − 108
AOD ₅₅₀ , RH, surface temperature, atmospheric stability	0.66	12.39	− 0.0004	0.89	1.55E − 117

where: H = hidden layers and

$$H1 = \text{TANH}(0.5 * ((-67.612) + (7.216 * \text{AOD}) + (-0.243 * \text{surface temperature}) + (0.214 * k \text{ index}) + (0.058 * RH)))$$

$$H2 = \text{TANH}(0.5 * ((-76.084) + (3.464 * \text{AOD}) + (-0.319 * \text{surface temperature}) + (0.254 * k \text{ index}) + (0.057 * RH)))$$

$$H3 = \text{TANH}(0.5 * ((-32.739) + (-0.667 * \text{AOD}) + (-0.169 * \text{surface temperature}) + (0.114 * k \text{ index}) + (0.075 * RH)))$$

The ANN technique produces similar or slightly better estimations of the PM_{10} concentrations compared to MLR, as shown from the statistical indicators in Table 4, which were computed from the comparison between measured and estimated PM_{10} concentrations at the 29 “training” stations (Fig. 1). Therefore, the ANN model slightly improves the R^2 from 0.66 (Table 3) to 0.71 (Table 4) and the RMSE from $12.39 \mu g m^{-3}$ in the MLR model to $11.61 \mu g m^{-3}$ (24.32%) in the ANN. A slightly different ANN model developed based on the 45 stations over the Peninsular Malaysia resulted in similar $R^2 = 0.73$ after comparison with the measured PM_{10} .

4.5. Model validation

The ability of both developed models (MLR and ANN) in predicting the PM_{10} concentrations over Malaysia was examined via comparisons against measured PM_{10} concentrations at the 16 stations used for model validation. The correlation between measured and estimated PM_{10} via the MLR model (Eq. (5)) exhibits a moderate but statistically significant correlation at 95% confidence level ($R^2 = 0.56$, p-value < 0.05, RMSE = $10.58 \mu g m^{-3}$ (24.28%), and MBE = $0.04 \mu g m^{-3}$) (Fig. 10a). The ANN model (Eq. (6)) very slightly improves the PM_{10} estimations compared to MLR with $R^2 = 0.58$, RMSE = $10.16 \mu g m^{-3}$ (23.31%) and MBE = $0.05 \mu g m^{-3}$. Similarly to our findings, Gupta and Christopher (2009b) found that ANN model improves the PM estimates compared to general linear models, such as the multivariate model. Nearly all the data points are well within the 95% confidence

Table 4

Statistical results of the ANN model used for PM_{10} estimations at the 29 training stations over Malaysia.

Parameter	Training data
R^2	0.71
RMSE ($\mu g m^{-3}$)	11.61
MBE ($\mu g m^{-3}$)	− 0.03
Index of agreement	0.90
N	723

Fig. 10. Correlation between PM_{10} concentrations estimated using (a) multiple linear regression and (b) Artificial Neural Network models and measured PM_{10} concentrations at 16 stations over Malaysia during 2007–2011.

interval of the observations, indicating a satisfactory accuracy in predicting the PM_{10} concentrations from satellite datasets, even over a tropical environment with large satellite data missing and perturbed data due to cloud contamination. Meanwhile, [Yap and Hashim \(2013\)](#) used PM_{10} concentrations from only 3 stations in Peninsular Malaysia for validation of the estimated monthly PM_{10} using the mixed effects model (the model was calibrated using data from 31 stations), obtaining an R^2 of 0.80. In our study, we used PM_{10} measurements from 29 stations for the models training/calibration and another 16 stations for the models validation, resulting in satisfactory R^2 values of 0.56 and 0.58 for MLR and ANN models, respectively after the validation procedure ([Fig. 10](#)). The validation results give credit to the accuracy of the developed MLR and ANN models for the PM_{10} estimations over Malaysia. It should be noted that the MLR and ANN models developed based on “training” stations only over the Peninsular Malaysia, resulted in larger errors in the PM_{10} estimations over the island Malaysia. Therefore, the current selection of stations over both regions and for various atmospheric and meteorological conditions seems to be the most intended way for model development. In a future step, different models will be developed for PM_{10} and $PM_{2.5}$ estimations separately over Peninsular Malaysia and Island Malaysia for more detailed predictions.

4.6. Sensitivity analysis

The AOD_{550} uncertainties were described in [Section 3.2](#), but have not been evaluated in the model development or for any other sensitivity study concerning the PM_{10} uncertainties. This is because the typical errors from the correlations between AOD_{550} and PM_{10} are larger than the AOD_{550} retrieval uncertainties, and the same is true for the other MODIS retrievals. Thus, any sensitivity study will result in PM_{10} uncertainties lesser than those obtained by the developed models. In this section, a sensitivity analysis was performed by changing the variables AOD_{550} , RH, surface temperature and k index as much as $\pm 5\%$ and $\pm 10\%$ and the results show that the estimated PM_{10} concentrations are well within the measurement uncertainties ([Table 5](#)). Comparison between the measured and simulated PM_{10} concentrations over the stations used for the model training showed that the RMSE varies between 10.51 and $10.70 \mu g m^{-3}$ for a $\pm 10\%$ change in AOD_{550} or in any other parameter, suggesting negligible changes in simulated PM_{10} concentrations due to uncertainties in the input variables. It should be noted that negligible differences in the statistical indicators (R^2 and RMSE) for a $\pm 5\%$ – 10% change in the input parameters were also observed for the MLR model.

4.7. Spatial distribution of estimated PM_{10}

The seasonal-mean spatial distributions of the estimated PM_{10} concentrations over entire Malaysia during 2007–2011 are shown in [Fig. 11a–d](#) at 10×10 km spatial resolution, using the ANN model. The seasonal-mean spatial mapping was performed by averaging the daily PM_{10} concentrations in each season. Very similar seasonal maps have been obtained using the MLR model (not shown).

As expected from the AOD_{550} spatial distribution, the dry season is associated with high PM_{10} concentrations reaching about 90 – $110 \mu g m^{-3}$ over the densely-populated southwestern part of the Peninsular Malaysia ([Fig. 11a](#)). Even higher PM_{10} concentrations are revealed over parts in the central Sabah and Sarawak, reaching above $\sim 140 \mu g m^{-3}$, due to the influence of severe biomass burning during the dry season. These high PM_{10} concentrations are also attributed to the extreme haze phenomenon occurred in 2009. According to [Hamdan et al. \(2010\)](#), severe haze conditions covered Sarawak in 2009 (August to early October) due to a strong El Nino event that caused dry weather in the Borneo island and lasted for about 3 months. According to DOE guidelines, PM_{10} concentrations of about 0 – $50 \mu g m^{-3}$ represent a relatively-clean “background” environment, while values of 51 – 100 , 100 – 200 , 200 – 300 and $> 300 \mu g m^{-3}$ correspond to moderate, unhealthy, very unhealthy and hazardous atmospheres, respectively. According to this classification, the atmosphere over the Peninsular Malaysia during the dry season is mostly considered as “moderate”, while over many parts of the Island Malaysia the atmospheric conditions are classified as “unhealthy”. During the wet season ([Fig. 11b](#)), the estimated PM_{10} are much lower due to rainy washout ([Juneng et al., 2009](#)) and only over the urban regions and northernmost areas of the Peninsular Malaysia and Sabah & Sarawak may reach up to ~ 90 – $110 \mu g m^{-3}$. In April–May ([Fig. 11c](#)), the PM_{10} spatial distribution exhibits great similarities with that of the wet season, with maximum concentrations over the urban centers and central-south parts of the Island Malaysia due to beginning of forest fires. The main sources of aerosols during this inter-monsoon season are vehicles emissions, industries and open burning activities ([Jamil et al., 2011](#)). Finally, October ([Fig. 11d](#)) exhibits higher PM_{10} concentrations over many parts of the Peninsular Malaysia as well as in Sabah and Sarawak resulting in high heterogeneity in the PM_{10} mapping, similar to that observed during the dry season.

The seasonal-mean measured and estimated PM_{10} concentrations over the 45 stations during 2007–2011 are shown in [Figs. 12–15](#). During the dry season ([Fig. 12](#)), the estimated PM_{10} concentrations vary widely between the stations from $\sim 30 \mu g m^{-3}$ (Bachang and Kapit) to ~ 60 – $80 \mu g m^{-3}$ over the urban and industrial stations, such as Bukit

Table 5
R² and RMSE values obtained from the comparison between measured and estimated PM₁₀ concentrations over the 29 training stations by changing ± 5% and ± 10% the input parameters in the ANN model.

Parameter	AOD	Relative humidity	Surface temperature	k index
Original	R ² = 0.58 RMSE = 10.61 μg m ⁻³	R ² = 0.58 RMSE = 10.61 μg m ⁻³	R ² = 0.58 RMSE = 10.61 μg m ⁻³	R ² = 0.58 RMSE = 10.61 μg m ⁻³
Original parameter minus 5%	R ² = 0.56 RMSE = 10.68 μg m ⁻³	R ² = 0.56 RMSE = 10.65 μg m ⁻³	R ² = 0.56 RMSE = 10.66 μg m ⁻³	R ² = 0.55 RMSE = 10.60 μg m ⁻³
Original parameter plus 5%	R ² = 0.57 RMSE = 10.65 μg m ⁻³	R ² = 0.57 RMSE = 10.61 μg m ⁻³	R ² = 0.56 RMSE = 10.68 μg m ⁻³	R ² = 0.56 RMSE = 10.64 μg m ⁻³
Original parameter minus 10%	R ² = 0.54 RMSE = 10.70 μg m ⁻³	R ² = 0.57 RMSE = 10.61 μg m ⁻³	R ² = 0.56 RMSE = 10.63 μg m ⁻³	R ² = 0.56 RMSE = 10.67 μg m ⁻³
Original parameter plus 10%	R ² = 0.58 RMSE = 10.51 μg m ⁻³	R ² = 0.56 RMSE = 10.56 μg m ⁻³	R ² = 0.56 RMSE = 10.70 μg m ⁻³	R ² = 0.56 RMSE = 10.70 μg m ⁻³

Fig. 11. Spatial distribution of the estimated PM₁₀ 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).

Rambai ($58.4 \pm 8.1 \mu\text{g m}^{-3}$), Petaling Jaya ($95.7 \pm 18.4 \mu\text{g m}^{-3}$), Shah Alam ($71.7 \pm 22.7 \mu\text{g m}^{-3}$), Balok ($72.9 \pm 27.7 \mu\text{g m}^{-3}$), Klang ($76.0 \pm 10.1 \mu\text{g m}^{-3}$) and Tawau ($81.7 \pm 11.4 \mu\text{g m}^{-3}$). According to Mahmud and Iza (2010), Bukit Rambai experiences high air-pollution levels throughout the year, since it is located in an industrial area with high anthropogenic and industrial emission rates and is also strongly effected by seasonal haze transported from neighboring countries. The seasonal-mean measured and estimated PM₁₀ values maximize over the urban and industrial sites, while great consistency is revealed between measured and estimated PM₁₀ between the sites.

During the wet season (Fig. 13), the PM₁₀ concentrations decrease

and the most of the stations located along the east coast of the Peninsular Malaysia, like Kota Bharu, Tanah Merah and Kuantan exhibit the lowest PM₁₀ concentrations (in the order of $30\text{--}40 \mu\text{g m}^{-3}$) as a result of rainy washout (Juneng et al., 2009). In contrast, industrial stations like Klang, Putrajaya, Balok, Seremban, Shah Alam, Paka-Kertih, Kemaman and Petaling Jaya are the most PM₁₀-loaded with concentrations of about $50\text{--}80 \mu\text{g m}^{-3}$ due to the continuous emissions in the industrial zones.

The PM₁₀ concentrations during the inter-monsoon (April–May) season show a smaller variation between the stations with values generally ranging between 40 and $60 \mu\text{g m}^{-3}$ (Fig. 14). Stations like

Fig. 11. (continued)

Nilai and Johor Bahru exhibit the highest PM_{10} concentrations ranging between 65 and $85 \mu g m^{-3}$ while rural areas show the lowest PM_{10} concentrations which is $< 60 \mu g m^{-3}$. The PM_{10} concentrations during October (Fig. 15) exhibit a similar pattern to that observed during April–May but with higher values, which are usually above $40 \mu g m^{-3}$ except for Jerantut, Kangar, Labuan, Pegoh, Sandakan, Kuala Selangor, Limbang, Sri Aman and Tanah Merah. Some stations exhibit very high monthly PM_{10} concentrations reaching $100 \mu g m^{-3}$, like Shah Alam

($144.7 \pm 25.4 \mu g m^{-3}$), Tasek ($122.23 \pm 25.4 \mu g m^{-3}$) and Tawau ($98.6 \pm 22.7 \mu g m^{-3}$).

During non-haze episodes, air pollution over Malaysia is mostly due to motor-vehicle emissions, industry and biofuel burning (Afroz et al., 2003; Jamil et al., 2011), thus favoring the accumulation of aerosols around the urban and industrialized areas. Many other factors that are not included in the current models, like the spatio-temporal distribution of rainfall, seasonally-changed winds and influence of the El-Nino

Fig. 12. Seasonal-mean measured and estimated PM_{10} concentrations at the 45 stations in Malaysia during the dry season.

Fig. 13. Same as in Fig. 12, but for the wet season.

Southern Oscillation strongly affect the atmospheric dynamics (e.g. drought, boundary-layer height, dilution processes, transported aerosol plumes, rainy washout) and, in turn, the aerosol loading and PM₁₀ concentrations over Malaysia. Despite all these challenges, the large gaps in the satellite dataset due to extensive cloud cover and the large heterogeneity in the aerosol field, the current analysis shows that PM₁₀ monitoring from space over Malaysia can be achieved with high accuracy (via both MLR and ANN techniques), which allows future PM₁₀ forecasts of good quality for monitoring local and regional air pollution on daily basis. The techniques developed in this study for PM₁₀ estimations will be helpful to assess the pollution levels over Malaysia at large spatial scales, which is essential for developing the Malaysian Environmental Performance Index (EPI). Currently, the global EPI is computed via PM_{2.5} estimations using MODIS AOD at 10-km spatial resolution (Van Donkelaar et al., 2010). This index needs to be refined in order to represent the local variations, so global EPI can adopt local values in its calculations. Once PM_{2.5} data becomes available in long-term basis over Malaysia, similar to the current techniques and models may be developed to predict PM_{2.5} concentrations and distribution over the country.

5. Conclusions

The current study dealt with PM₁₀ estimations from space and mapping of the seasonal-mean PM₁₀ concentrations (10 × 10 km spatial resolution) over Malaysia during the period 2007–2011. Multiple Terra-MODIS retrievals like AOD₅₅₀, atmospheric profiles and calibrated radiances were used for computations of the spatial distributions of aerosol, RH, surface temperature and stability index that were involved in the developed MLR and ANN models for the PM₁₀

estimations from space. The estimated RH showed the potential of remote-sensing data to provide atmospheric parameters needed to run the model due to strong correlation and low bias against ground-based measurements ($R^2 = 0.78$, RMSE = 5.14% and MBE = −0.06%). Furthermore, the surface temperature calculated using MODIS data revealed moderate-to-good correlation with the ambient temperature from the meteorological stations ($R^2 = 0.49$, RMSE = 2.68 °C and MBE = −0.03 °C). Multiple linear regression (MLR) and Artificial Neural Network (ANN) models were developed using the MODIS AOD₅₅₀, atmospheric stability, RH and surface temperature as variables for better estimations of PM₁₀, instead of the simple linear regression between measured PM₁₀ concentrations and satellite AODs. Stepwise regression techniques examined the performance of each parameter in PM₁₀ estimates and the MODIS AOD₅₅₀ was found to yield the highest correlation with PM₁₀ ($R^2 = 0.59$), followed by atmospheric stability ($R^2 = 0.16$) and surface temperature ($R^2 = 0.11$). The inclusion of all parameters in the MLR model increased the accuracy of the predicted PM₁₀ ($R^2 = 0.66$). In future research, both wind speed and wind direction will be included in the MLR and ANN models attaining for more accurate estimations of the PM₁₀ concentrations, since the wind regime controls the air-mass trajectories, thus affecting the PM₁₀. The MLR and ANN models developed for PM₁₀ estimation from space were initially trained against measured PM₁₀ concentrations at 29 stations homogeneously distributed over Malaysia resulting in average RMSE values of 12.39 $\mu\text{g m}^{-3}$ and 11.61 $\mu\text{g m}^{-3}$, respectively indicating satisfactory accuracy in the estimates. The ANN model resulted in slightly higher accuracy compared to MLR and an RMSE of 10.16 $\mu\text{g m}^{-3}$ was found after model's validation against PM₁₀ measurements at 16 stations. The seasonal-mean PM₁₀ mapping over Malaysia revealed significant spatio-temporal heterogeneities in the

Fig. 14. Same as in Fig. 12, but for the inter-monsoon (April–May) season.

Fig. 15. Same as in Fig. 12, but for the inter-monsoon (October) season.

PM₁₀ concentrations, as a result of the high heterogeneous aerosol and meteorology fields. The high performance of the MLR and ANN models for PM₁₀ estimations over Malaysia allows for future estimates of PM_{2.5} concentrations from space and monitoring of the Environmental Performance Index by developing separate models over the Peninsular and Island Malaysia. When more daily data pairs (AOD and PM) become available over Malaysia we will use the mixed-effects model combined with meteorological parameters for PM estimations on a daily basis.

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