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Spatial Assessment of Land Use Impact on Air Quality in Mega Urban Regions, Malaysia

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Highlights

- Impact of land use changes towards air quality at mega urban were investigated.

- All air pollutants showed a rising trend, except SO₂ concentrations.
- The built-up lands increase with a reduction in vegetation and water bodies area.
- The increase of built-up lands responsible for motor vehicle and industrial emissions.
- The land use categories have a significant impact on NO, NO₂, NO_x, CO and SO₂.

Abstract

This study aims to determine the impact of land use changes on air quality at the largest conurbation area in Malaysia, Kuala Lumpur Extended Mega Urban Regions (KLEMUR) from 2000-2015. Statistical and geostatistical analysis were used to analyse air quality and land use data. The results showed that only the daily average concentration of PM₁₀ at several stations exceeded the concentration suggested by the World Health Organization (WHO). All air pollutants measured showed a rising trend, with the exception of the SO₂. The land use trends during the 16-year research period showed an increase in built-up lands (4.0%) and a reduction in vegetation (3.3%) and water bodies (2.3%) which caused the spatial distribution of air pollutants to expand from the centre of KLEMUR to the north and south. Principal Component analysis (PCA) demonstrated that 52.68% of the air pollution was influenced by vehicular and industrial emissions given that the strong factor loadings (> 0.70) consisted of NO, NO₂, NO_x, CO and SO₂. The land use changes have a significant impact ($p < 0.10$) on air pollution in KLEMUR since NO, NO₂, NO_x, CO and SO₂ were positively correlated with the built-up lands and negatively correlated with vegetation and water bodies.

Keywords:

Mega urban regions; Urban air pollution; Land use changes; Sustainable urban land use; Geostatistical

1 Introduction

Air pollution from various sources such as industrial activity and vehicle emissions has become a major threat to human health. Air pollution exposure increases the risk of people suffering from health complications that affect them both psychologically (through stress, mood swings, anxiety, major depression, dementia, bipolar and schizophrenia) and physically (through premature birth, lung cancer, cardiovascular and respiratory diseases) (Attademo et al., 2017; Gurjar et al., 2010; Khan et al., 2019; Kim et al., 2018; Rajagopalan et al., 2018; Rajper et al., 2018; Thomson, 2019). Moreover, the World Health Organization (WHO) has reported that approximately 4.2 million people have died due to health issues resulting from air pollution (World Health Organization, 2020). They also assert that about 93% of children around the world breathe toxic air every single day (World Health Organization, 2018). Air pollution, especially in urban areas, has become a major concern due to the growing size of populations and pollution sources. Data on air pollution is quite disturbing and shows that 97% of cities around the world have, in recent years, failed to meet the WHO air quality guidelines (World Health Organization, 2018).

In urban areas, air pollution is predominantly determined via anthropogenic sources, such as traffic and industrial emissions. These are expected to increase as the size of urban

areas expand (Metia et al., 2020; Yang et al., 2019). Based on Suzuki et al. (2010), urbanization is predicted to triple the size of urban areas within the next two decades since the rise in the world's resident population in urban areas is expected to increase from 52% in 2011 to 67% in 2050 (United Nations, 2012). Rapid urbanization also results in land use patterns growing outwards from small urban centers to their original individual boundaries, seeing them then merge into each other to form huge conurbations of urban centers. Although the major sources of urban air pollution are considered to be from traffic and industrial emissions, changes in land use have also been identified as having a close link with air quality in urban areas (Huang et al., 2013; Romero et al., 1999; Weng & Yang, 2006; Xu et al., 2016; Zahari et al., 2016). Land use changes have, in effect, a close connection with air quality due to the effects of the distribution of the air pollutants that have been emitted from different kinds of sources which depend on the activities of the land use. Land use changes therefore affect air quality by influencing the spatial distribution of land use and human activities (Wei & Ye, 2014). Urban land use patterns are usually reflected via the dispersion of air pollutants and air quality in urban environments (Huang & Du, 2018). Land use changes can either be detrimental to the environment or improve the air quality. Good planning of land use can provide residents of an area with a liveable community and positively impact economic development. However, the deterioration of urban air quality is the sign of enormous stress led by the rising patterns of land use changes which are driven by freakishly fast rates of urbanization (Du et al., 2010; Hien et al., 2020; Huang & Du, 2018; Tao et al., 2015; Wei & Ye, 2014). Anthropogenic sources of air pollution are released both directly and indirectly from built-up land whereas the majority of socio-economic activities in urban areas take place. According to Irga et al. (2015), natural land cover surfaces such as forests, natural vegetation and water bodies have a positive impact and improve air quality in urban areas. Unfortunately, the transformation of natural vegetation, and the burial of water bodies into built-up areas through the process called 'rapid

urban expansion' has caused the positive effects of the natural land cover surfaces to gradually decline (Du et al., 2010; Xu et al., 2016).

Land use changes can have a direct or indirect impact on natural land cover surfaces which in other ways influence the transport/dispersion of air pollution and urban air quality. According to previous studies, built-up lands can be closely related to higher tiers of emissions and consequently cause air pollution (Wang et al., 2018; Weng & Yang, 2006; Xian, 2007; Zahari et al., 2016). A study conducted by Weng and Yang (2006) examined the impact of land use variables on air pollutants using buffer analysis and correlation analysis whereby the results showed that the spatial trends of air pollutants correlated positively with the density of built-up land in Guangzhou, China. A study by Superczynski and Christopher (2011) also found significant correlations between the PM_{2.5} concentrations and the urban land which surrounded the air quality monitoring sites in Birmingham, Alabama. Furthermore, findings by Wang et al. (2018) show there are significant enhancements of CO₂ emissions around the Pearl River Delta area in China through land conversion from non-built up to built-up areas where development was for industrial, commercial, residential and traffic purposes. In these previous studies, there was evidence of a relationship between the built-up land category and air pollutants, particularly in urban areas. A study conducted by Xu et al. (2016) was able to prove that different land use categories in Wuhan, China had invidious effects on a range of air pollutants. A similar study by Zahari et al. (2016) conducted in Iskandar, Malaysia, showed the diverse effects that different types of built-up lands resulted in, which strongly suggested that the concentration of PM₁₀ is affected by a greater degree of commercial land use and industrial land. These findings are in contrast with a study by Zhu et al. (2019) which suggested that the water bodies area in Wuyishan City, China was negatively correlated with PM₁₀ and PM_{2.5} while the cultivated land was positively correlated with O₃, CO, SO₂ and black carbon. Studies have shown that urbanization causes an increase in living discharges which not only

change the land use structure but also strongly impact air quality (Yang et al., 2019; Zhu et al., 2019).

As such, land use strategies should therefore be given more priority when strategizing plans and formulating policies to reduce air pollution (Xu et al., 2016), this is particularly important for the mega urban regions which are expected to undergo rapid expansion along with population growth. This study attempts to analyze the relationship between land use changes and air pollutant trends within the largest urban conurbation area in Malaysia, namely the Kuala Lumpur Extended Mega Urban Region (KLEMUR), which has undergone a series of urbanization phases since the 1980s. This study focuses on spatio-temporal aspects using air quality data obtained from eight continuous air quality monitoring stations, as well as KLEMUR land use data recorded over a period of 16 years (2000-2015). Three main objectives were set for the relationship analyses, the first was to determine spatio-temporal air pollutant variations using statistical and both interpolation and extrapolation analysis where this analysis helps to assess the long term-air pollutions trends in relation to space and time changes, as KLEMUR consists of emerging cities that have undergone rapid urbanization. The second was to determine the land use characteristics transformation using buffer analysis as this analysis helps to determine the rate of land use change and the types of land use affecting air pollution intensity. In order to verify these, the third objective was to investigate the impact of different land use categories and how changes in land use type impacted the intensity of different air pollutants. This was conducted using correlation analysis. Fundamentally, the aim of this study is to provide scientific evidence to help policymakers adopt sustainable land use policies in order to improve KLEMUR's air quality.

2 Materials and methods

2.1 Study Area

Kuala Lumpur Extended Mega Urban Region (KLEMUR) is the largest urban conurbation region in Malaysia. The term KLEMUR is used, since Kuala Lumpur is the centre of the urban region and emerged into a dominant position based on its status as the national capital, as well as the centre for socio-economic growth. KLEMUR covers the area from the Bernam River Basin in south Perak to the Linggi River Basin in Negeri Sembilan, extending also to the Melaka River Basin in Melaka. The encircled area is about 11 982 km², and approximately 40 km east-west, from the mountain spine to the Straits of Melaka, and has been developed since 1980 to the point that it has emerged as a mega urban region. About 34% of the total population in Malaysia has been attracted to live in KLEMUR and the population is expected to grow in the future (Hadi & Idrus, 2017). Eight continuous air quality monitoring stations, all managed by the Malaysian Department of Environment (DOE), were selected for this study. Tg Malim (S1), Petaling Jaya (S2), Klang (S3), Shah Alam (S4), Nilai (S5), Seremban (S6), Bandaraya Melaka (S7) and Bukit Rambai (S8) continuous air quality monitoring stations are all located within the area of KLEMUR, situated in the northern region of the Malaysian Peninsular. All selected continuous air quality monitoring stations face the Malacca Straits to the west. S1 is a suburban station that is located in the residential area in the north of KLEMUR. Stations S2, S3 and S4 are in the central region of KLEMUR. Station S3 is located in a heavily industrialized area near to Port Klang, one of the main and busiest ports in Malaysia (Dominick et al., 2012). Stations S2 and S4 are located in highly populated residential areas. Furthermore, these stations are surrounded by industrial and commercial activities which are located near congested roads. Nilai (S5) and Seremban (S6) stations are situated in the south of KLEMUR. These two stations are based in a residential area that is facing rapid development. Bandaraya Melaka (S7) and Bukit Rambai (S8) stations are both suburban stations situated in the south of KLEMUR as well. Station S7 is situated near a popular touristic area so is surrounded by busy traffic while station S8 is situated in an

industrialized area. The location of the selected continuous air quality monitoring stations in KLEMUR, used for this study are shown in Fig. 1.

[Fig. 1]

2.2. Data Collection

The daily recorded air quality data for the sixteen-year-period, starting from January 2000 until December 2015, at the selected continuous air quality monitoring stations in KLEMUR for this study was obtained from the Air Quality Division of the Department of Environment (DOE), Malaysia. The air quality dataset consisted of ten variables for which the air pollutant variables included particulate matter with a diameter size of less than 10 μm (PM_{10}), carbon monoxide (CO), ground level ozone (O_3), nitrogen oxide (NO), nitrogen dioxide (NO_2), oxides of nitrogen (NO_x) and sulphur dioxide (SO_2) while the meteorological variables consisted of ambient temperature, relative humidity wind speed and wind direction. Table 1 shows the measurements of the variables. A Teledyne API instrument (Teledyne Technologies Inc., USA) was used to measure the gas pollutants (O_3 , CO, SO_2 , NO, NO_2 and NO_x) concentrations on an hourly basis. While, the Met One Instrument (Met One Instrument, Inc., USA) was used to measure the particulate matter (PM_{10}) concentrations and the meteorological variables (ambient temperature, relative humidity, wind speed and wind direction) (Latif et al., 2014).

[Table 1]

The land use data for KLEMUR in the years 2000 to 2015 was obtained from the Federal Department of Town and Country Planning (PLAN Malaysia) and Kuala Lumpur City Hall (DBKL). This study focused on three main land use activities which are built-up lands, vegetation and water bodies. When it comes to land use type, built-up lands consist of

transportation, industrial, residential and facilities. The vegetation category consists of forest, vegetation and green areas and the water bodies category, comprises lakes, rivers and beaches.

2.3 Data Analysis

2.3.1 Univariate Statistical Descriptive Analysis

Univariate statistical descriptive analysis was performed to describe and summarize the distribution of air pollutants individually in order to determine the status of different air pollutants on KLEMUR within a long-term period as the results will be compared with the air quality standards (Cleff, 2019). The analysis was carried out using all hourly averages of the air pollutants concentrations, except for PM₁₀ as this is based only on daily average (24 h) concentrations in order to determine the measure of spread (minimum, maximum, mean and standard deviation). Air pollutant concentrations were compared to the allowable values suggested by the Malaysian Ambient Air Quality Standard (MAAQS) and World Health Organization (WHO). The MAAQS limit levels consist of three interim targets, starting from 2015 until 2018, with full implementation of the air quality guidelines by 2020. The results of this study were compared to the limit levels for the interim target 1 in 2015 (IT-1 2015).

2.3.2 Inverse Distance Weighted (IDW)

Predicting a new value through interpolation and extrapolation is a critical technique commonly used in geostatistical analysis. This technique's function is to estimate new data points within a range of discrete sample data points and a boundary area (Diao et al., 2019; Li et al., 2010; Tong, 2020). Based on Eq. 1, the value at a given point is constructed from a weighted sum of data values at surrounding points whereby the weight functions reduce with the increase in the distance of separation. The interpolation formula for Eq. 1 is as follows:

$$\hat{y}(x_0) = \sum_{i=1}^n \lambda_i \cdot y(x_i) \quad \text{and} \quad \lambda_i = \frac{1}{d_{0i}^p} \quad (\text{Eq. 1})$$

where n is the sampled point used for the estimation of the new data points, x_0 is the targeted point while x_i is the sampled point. The $\hat{y}(x_0)$ is the estimated value of the primary variable at the targeted point and $y(x_i)$ is the observed value at the sampled point. Then, λ_i is the weight assigned to the sampled point where the weight is the inverse function of the distance, d_{0i} (distance from the sampled point to the targeted point and p is the power) (Kumar et al., 2019; Tong, 2020). The interpolation and extrapolation method used in this study was Inverse Distance Weighted (IDW), which aims to examine the distribution of major air pollutants in KLEMUR over the long-term. This method works by using the Tobler's first law concept to estimate unknown cell point values based on known average points of cell values within the area (Chen & Liu, 2012; Lu & Wong, 2008). The IDW method is a popular alternative to the Kriging method in air pollution modelling studies, which can be used to give a fairly reliable performance when predicting new values of air pollutant concentrations (Kumar et al., 2016; Kumar et al., 2019; Vorapracha et al., 2015; Wong et al., 2004). The IDW method calculates all values of the points within the range of the boundary area. It estimates the value of a point from the distance between sampled points and the targeted points, improving the prediction when the minimum and maximum values of the surface are represented by the discrete sample data points (Kumar et al., 2019). The IDW method appears to be an appropriate navigational and averaging tool for the spatial distribution analysis of air quality because of its advantages. It enables the modeling of various scales, can lower the uncertainties prediction in exposure assessments, creates smooth surfaces of spatial distribution and offers a comparable performance to Kriging (Chen & Liu, 2012; Li et al., 2014; Susanto et al., 2016). In addition, the IDW method is a suitable and practical spatio-temporal prediction method for the spatial

modeling of air quality when the point observations are lower (Jumaah et al., 2019; Li et al., 2016; Li et al., 2019; Peshin et al., 2017).

2.3.3 Principal Component Analysis (PCA)

Principal Component Analysis (PCA) is used in this study in order to determine the major sources of the air pollution in the KLEMUR as a result of land use activities. This technique has been widely applied in previous air quality studies (Dominick et al., 2012; Eder et al., 2014; Elorduy et al., 2016; Franceschi et al., 2018; Halim et al., 2018). PCA is an orthogonal transformation method that is able to identify the interrelation within the observational variables (Voukantsis et al., 2011). PCA provides the most significant and meaningful variables that reduce the dimensionality of a number of interrelated variables while retaining the maximum variability present in the data (Franceschi et al., 2018; Shiva Nagendra & Khare, 2003). PCA in this study used air pollutants and meteorological variables data, arranged on an hourly average time basis. Small and slightly-sized data is not suitable for the PCA method because the main function of this analysis is to reduce the size of the data being tested to just a few components. The principal components (PCs) were calculated by ordering the variables in such a way that the first variable explains the largest proportion of variability within the original data. This study chose to use PCA analysis with varimax rotation based on Kaiser normalization. Varimax rotation ensures that each variable is maximally correlated with only one component and has a near zero association with the other components. PCA is composed, based on Eq. 2, as follows:

$$PC_i = l_{1i}X_1 + l_{2i}X_2 + \dots + l_{ni}X_n \quad (\text{Eq. 2})$$

where PC_i is the i^{th} principal component and $l_{ni}X_n$ is the loading of the observed variable X_n (Abdullah et al., 2018). PCA produced several PC from the observed data. The Kaiser-Meyer-

Olkin values of the PCs, which should be greater than 0.5, were considered adequate (Eyduran et al., 2010; Voukantsis et al., 2011). The factor loading values after the varimax rotation can reflect how much the variables (air pollutants or meteorological) contribute to that PC where the higher the factor loading value of that variable (near to 1.0) for that particular PC, the more the variable contributes to the variability in the PC which shows the strongest contributor to the sources of the air pollution (Chavent et al., 2009; Dominick et al., 2012; Jolliffe, 2002).

2.3.4 Buffer Analysis

Buffer analysis is one of geospatial analyses used to determine the air quality spatio-temporal response towards land use surrounding the air quality monitoring stations in KLEMUR, where the responses are varied by the spatial scales. A series of buffers with different spatial scales were created at each of the air quality monitoring stations in KLEMUR in order to obtain land use variables information. In this study, the series of buffers was set to five radiuses: 1 km, 2 km, 3 km, 4 km and 5 km (Fig.1). According to Xu et al. (2016), if the radius of the buffer is too far out from the monitoring stations, the discrepancies in land use categories shall not be clearly distinguished. The built-up lands, vegetation and water bodies were the three main land use categories that have been selected to be studied. In this buffer analysis, the areas within each buffer of the three land use categories were calculated and they were classified as land use variables and the year of land use was noted. The areas of land use variables were averaged for the year 2000 and 2015.

2.3.5 Bivariate Correlation Analysis

Bivariate correlation analysis was used to determine the magnitude of correlation between land use categories in KLEMUR and air pollutant concentrations from the year 2000 to 2015 at different spatial scales (series buffers radiuses). This study used Pearson's correlation analysis since the data is normal distribution. Correlations between the land use

categories (built-up lands, vegetation and water bodies) and the concentrations of PM₁₀, O₃, NO, NO₂, NO_x, CO and SO₂ were measured based on the calculations of the Pearson's correlation coefficient in Eq. 3:

$$r_{a,b} = \frac{\sigma_{ab}}{\sigma_a \sigma_b} \quad (\text{Eq. 3})$$

where $r_{a,b}$ represents the covariance between the two variables, a represents the land use categories, and b represents the air pollutants' concentration. The σ_a in the equation represents the standard deviation of a while σ_b represents the standard deviation of b (Zhu et al., 2019). When the covariance between the two variables is divided by the two standard deviations, the range of the covariance is rescaled to the value of -1 and 1. From the correlation analysis, a correlation coefficient value of -1 and 1 suggested the variables have a significant negative correlation and a significant positive correlation while a value of 0 indicated no significant relationship between the variables (Rodgers & Nicewander, 1988; Zhu et al., 2019). The highest Pearson's correlation coefficient was chosen as an optimum radius that shows a significant relationship between a selected land use category and a selected air pollutant.

3 Results and Discussion

3.1 Spatial Variation of KLEMUR's Air Quality

3.1.1 Univariate Descriptive Statistics

Table 2 shows the statistical distribution of the air pollutant variables for KLEMUR which consist of the minimum, maximum, mean and standard deviation values based on hourly averages for gases and 24 h for PM₁₀. Overall, the air quality status in KLEMUR over the 16-year study period shows that the average concentrations of all air pollutants measured are below the Malaysian Ambient Air Quality Standards (MAAQS). However,

the daily average concentration of PM₁₀ at several stations located in the city centre and industrial areas such as Petaling Jaya (S2) station (51.81 µg/m³), Klang (S3) station (67.00 µg/m³), Shah Alam (S4) station (54.83 µg/m³), Nilai (S5) station (59.66 µg/m³) and Bukit Rambai (S8) station (66.02 µg/m³) exceeded the concentration suggested by the World Health Organization (WHO) guidelines for ambient PM₁₀ concentrations which is 50 µg/m³. The background concentrations of PM₁₀ in KLEMUR and Malaysia as a whole, which is located within a tropical environment contributed to by high temperature conditions and natural emissions such as soil dust and other natural sources, influence the concentration of PM₁₀ in ambient air. Anthropogenic sources such as emissions from motor vehicles, industrial activity and regional biomass burning also contribute to the higher PM₁₀ level in these areas. The high concentration of PM₁₀ during biomass burning episodes, especially during the southwest monsoon which occurs between June to September every year, usually contribute to the maximum value of PM₁₀ concentrations in KLEMUR (Azmi et al., 2010; Juneng et al., 2009; Latif et al., 2018; Wui et al., 2018).

[Table 2]

The gas concentration results show that the maximum value of O₃ exceeded the standard concentration suggested by MAAQS (100 ppb) at all stations in KLEMUR. This can be attributed to high photochemical reactions which occurred due to the presence of high ultraviolet (UV) radiation, which reacted with O₃ precursors such as NO_x and volatile organic compounds (VOCs) to form O₃ (Vermeuel et al., 2019; Wałaszek et al., 2017; Wei et al., 2014). In some areas, O₃ is known as a secondary gas with high concentration values after PM₁₀ due to the concentrations of O₃ being higher during the afternoon and due to the fact that it dominates the Air Pollution Index (API) (Department of Environment Malaysia, 2013; Halim et al., 2018; Orru et al., 2017). Meanwhile, there were two continuous air quality monitoring stations where the SO₂ concentration exceeded the hourly maximum of

allowable values by MAAQS (135 ppb) which are the Klang (S3) station (179.00 ppb) and Shah Alam (S4) station (139.00 ppb). Both of these stations are located in heavily industrialised areas where the SO_2 is derived from industrial activity and heavy-duty vehicle emissions (Mohamad et al., 2015; Pereira et al., 2007).

The mean of NO , NO_2 and NO_x concentrations in KLEMUR were found to be within the range 4.59–39.70 ppb, 6.99–30.82 ppb and 12.29–70.16 ppb, respectively. The mean values of the NO_2 concentration at Petaling Jaya (S2) station (30.82 ppb) and Klang (S3) station (21.67 ppb) exceeded the guideline concentrations suggested by WHO (21.26 ppb). In addition, S2 station recorded an hourly maximum NO_2 concentration (216.00 ppb) above the value recommended by MAAQS (170 ppb). Stations S2 and S3 are surrounded by congested roads where the main source of NO_2 is from traffic emissions (Latif et al., 2014; Leh et al., 2014). Although the CO concentration (4460–15 510 ppb) in KLEMUR over the 16 years was still below the maximum concentration allowed by MAAQS (30 000 ppb), the air quality status in KLEMUR is quite worrying as most of the air pollutants exceeded the healthy levels recommended by air quality standards. In fact, as it is predicted that the urban population will have grown by more than 50% by 2050, this will increase both the urban size and also the concentration of air pollutants in the future (United Nations, 2012; Yang et al., 2019).

3.1.2 Spatial Distribution of Air Pollutants in KLEMUR

Inverse Distance Weighted (IDW) was employed to find the spatial distribution of air pollutants in KLEMUR. Fig. 2 shows the contour maps obtained by the IDW technique. The contour maps allow the spatial distribution of air pollutants, in places where no measurements of pollutants had been made, to be estimated.

[Fig. 2]

The spatial distribution for PM₁₀ in 2000 shows that the most-affected areas are those in the centre of KLEMUR. In 2015, the distribution of PM₁₀ started to expand from the centre of KLEMUR to south of the region. The PM₁₀ concentrations for the years 2000 and 2015 both exceeded the established annual average limit by WHO (20 µg/m³) and MAAQS (50 µg/m³) (green, yellow, orange and red). In 2000, the Tg Malim, Seremban and Melaka stations, were still below 50 µg/m³ but the PM₁₀ concentration showed a tendency to increase in 2015. Then, in 2015, the only station in KLEMUR that showed a PM₁₀ concentration below 50 µg/m³ was Tg Malim station. This related to the urban expansion that occurred in KLEMUR over the 16-year period, whereby a developed urban area with heavy industrial and commercial activities contributed to an increased concentration of PM₁₀ (Asmat et al., 2018). In addition, the geographical characteristics of the source area, an unfortunate flow of the wind direction and the effects of the biomass burning plume could also be responsible for enhancing the suspended particulate matter in the area (Afroz et al., 2003).

On the O₃ contour map, it can be seen that the distribution of O₃ in KLEMUR increased from 2000 to 2015. In 2000, the distribution of O₃ was recorded as being high at Shah Alam and Bukit Rambai stations. Both stations are located in large and busy industrial areas in KLEMUR. Then in 2015, the distribution started to expand to the south of KLEMUR. The expansion of O₃ distribution relates to the development and extension of the industrial area, particularly in the south of KLEMUR where water bodies were reclaimed to solve land shortage problems and provide additional space to meet industrial and housing demands (Mohamed & Razman, 2018). Industrial activities, as well as the additional number of motor vehicles, increase the emission of O₃ precursors such as NO_x, VOCs and CO (Banan et al., 2013). The chemical process of O₃ formation that occurs through reaction sequencing involves VOCs, CO and NO_x, which results in the conversion of NO to NO₂ and formation of O₃ from

NO₂ by photolysis reaction as shown by Sillman (1999), increased the possibility of O₃ distribution to larger areas in KLEMUR.

These maps (Fig. 2) show that in 2000, NO, NO₂ and NO_x distributions for the most affected areas, were all in the centre of the KLEMUR, where Petaling Jaya station is located. Petaling Jaya station is situated close to the capital city of Malaysia, Kuala Lumpur where the area is known to have a high volume of traffic (Leh et al., 2014). Within 16 years, the distribution of NO, NO₂ and NO_x had extended to both the north and south of the region, where the road networks had expanded by 2015 due to an increase in the human population in the urban areas (Almselati et al., 2011). The expansion of road networks involved main highways such as KL-Putrajaya Highway, SMART Tunnel, North-South Expressway, Kajang-Seremban Highway, South-Klang Valley Expressway (SKVE) and Kuala Lumpur-Kuala Selangor Highway (Malaysian Highway Authority, 2010). It is possible that this expansion correlates with the trends of CO spatial distribution in KLEMUR from 2000 to 2015. In 2000, the distribution of CO was mainly in the centre of KLEMUR while the spatial distribution of CO started to expand to the north and south. The growth of the road network, improvement of road systems and public transportation in KLEMUR caused a reduction in traffic congestion in previously high traffic volume areas such as those located in Petaling Jaya, Klang and Shah Alam. This in turn led to a decrease in CO concentrations in highly populated areas (Hirota, 2010; Mahirah et al., 2015; Malaysian Highway Authority, 2010). Azhari et al. (2018) findings also show that the number of vehicles on the roads influences the rise of air pollutant concentrations thus indicating that traffic is the main contributor of the air pollution emissions. A decreasing trend in the levels of SO₂ was observed in the period 2000 to 2015. This is the result of the air quality improvement plans implemented in Malaysia which mainly focus on catalytic converters and reformulated fuels whereby vehicle emission regulations had introduced the 'Euro 1' standard in early 2000 and the 'Euro 4' standard in 2010 (Hirota, 2010).

3.1.3 The Major Sources of Air Pollutants

The results of the PCA loadings after the varimax rotation are shown in Table 3 respectively. From the results, the Kaiser-Meyer-Olkin's value is 0.683 and only strong factor loadings with values over 0.7 were selected for the PCA interpretation for each component.

[Table 3]

There are three factors produced after the varimax rotation. The first factor (F1) explains 52.68% of the total variance which shows strong positive factor loadings for NO (0.910), NO₂ (0.949), NO_x (0.975), SO₂ (0.722) and CO (0.926). The possible sources of air pollution in F1 are suspected to be influenced by motor vehicles emissions that emit NO, NO₂, NO_x and CO from incomplete combustion during traffic congestion (Grote et al., 2016; Zhang & Batterman, 2013). The concentration of SO₂ can be seen to be caused by industrial emissions and motor vehicles that utilize diesel fuel, such as heavy-duty vehicles (Kalghatgi, 2018; Şahin, 2019). The second factor (F2) explains 28.82% of the total variance and shows strong positive factor loadings for O₃ (0.916), temperature (0.733), wind speed (0.863) and strong negative factor loading for humidity (-0.953). This can be related to the ozone (O₃) formation mechanism (Wanjala et al., 2018). The rate of photochemical production to produce O₃ rise when there is a high temperature, high wind speed and low humidity (Jhun et al., 2015). While the last factor, the third factor (F3) explains 10.25% of the total variance and shows strong positive factor loading for PM₁₀ (0.877). The particulate matter will transport easily when the flow of wind speed increases which in turn leads to an increment in the quantity of UV light reaching the ground and influencing the rate of the photochemical process (Jasaitis et al., 2016; Li et al., 2013). From the PCA's results, the major sources of air pollution in KLEMUR were dominated by industrial activities and vehicular emissions from the congested roads near the

developed areas. Besides, meteorological factors also play an important role in influencing air pollutants such as O₃ and PM₁₀.

3.2 The Relationships of Land Use Categories with Air Pollutants

3.2.1 The Trend of Land Use Changes

KLEMUR land use areas have undergone a series of urbanization phases since the 1980s. As shown in Fig. 3, there are obvious changes between the land use scenario in KLEMUR for 2000 and 2015. The area proportions of KLEMUR in 2000 consisted of 2075 km² of built-up lands, 9721 km² of vegetation and 177 km² of water bodies. After more than a decade, the area proportions changed. In 2015 the area proportions consisted of 2554 km² of built-up lands, 9334 km² of vegetation and 94 km² of water bodies which indicated within the space of 16 years that there was an increase of 4% for built-up lands, a 3.3% decrease in vegetation and a 2.3% decrease for water bodies.

[Fig. 3]

A 5 km buffer was used to obtain land use variables from different spatial scales and to determine the transformation of land use characteristics around the continuous air quality monitoring stations located in KLEMUR. Land use patterns around the continuous air quality monitoring stations were measured by calculating the areas of the different land use categories within a 5 km radius buffer. The calculated land use area proportions for each category at each site in 2000 and 2015 are shown in Table 4.

[Table 4]

Table 4 illustrates that the area proportions for different land use categories around these continuous air quality monitoring stations vary greatly. The highest area proportion of built-up lands in 2000 was recorded by Seremban station at 86.7%, followed by Shah Alam,

Petaling Jaya, Klang, Melaka, Bukit Rambai, Nilai in the range of 79.6% to 22.5% while the lowest area properties went to Tg Malim station at 1%. The area proportion of built-up lands changed across the years, however. In 2015, the area proportion of built-up lands showed an increasing pattern for all stations. The highest increment recorded was from Petaling Jaya station which recorded a 16.5% increment compared to other stations. According to Leh et al. (2014), the increase of built-up lands in Petaling Jaya was to fulfil the demand for residential areas and transportation which led to road network expansion in the nearby region. Economic drivers have essentially become the reason behind the rise in built-up lands consisting of residential and industrial construction, along with the expansion of transportation networks. Such changes have been the direct result of the growth of the urban population in KLEMUR (Hadi & Idrus, 2017). This concurs with a study by Wu et al. (2016) which revealed that land use changes caused by urban expansion were strongly correlated with the growth of the urban population and economic development. Furthermore, the rapid transition in land use changes in KLEMUR within the 16-year period was also due to the outcomes of New Economic Policy (NEP) in 1970 and the subsequent development policies and programs which led to widespread social, economic and environment change in KLEMUR (Hadi et al., 2011).

The area proportion for vegetation as a land use category in 2000 varied among the stations in KLEMUR. The area proportion of vegetation was between 12.6% to 96.4%, where Tg Malim station had the largest area of vegetation while Seremban had the lowest among all stations. The vegetation areas across stations over the 16-year period showed a decreasing trend, with the exceptions of Melaka and Bukit Rambai stations. This was due to their interest in multiple agricultural activities such as the production of palm oil (Kamalrudin & Ramli, 2014). The Petaling Jaya station showed a decrease of 16.2% in vegetation area. According to Xia et al. (2015), the loss of a massive quantity of vegetation areas and forests around the cities was due to a growing demand for built-up areas.

Water bodies occupy 41.4% of the 5 km buffer area at Melaka station. This is the highest proportion of water bodies compared to the other stations while the lowest proportion is at Shah Alam station with only 0.1%. All stations showed a decreasing trend within the 16-year period except for Seremban. Melaka and Bukit Rambai stations showed a decrease in water bodies by 8.2% and 7.0% respectively, these were among the highest decrement of water bodies areas due to coastal reclamation activities (Mohamed & Razman, 2018). Different area proportions of land use categories around these stations may affect the air quality in different ways. Most sites showed a rising trend in built-up lands within the 5 km buffer while water bodies and vegetation areas declined respectively. The expansion of built-up lands and conversion of vegetation and water bodies to develop the industrial and residential areas are mainly associated with the rapid urbanization that is occurring in the urban area (Di Sabatino et al., 2018).

3.2.2 The Correlation of Land Use Categories with Air Pollutants

In this study, the bivariate correlation analysis is the basis to determine the interlinked magnitude of land use categories with the air pollutants in KLEMUR over the 16-year study period. The correlation coefficient (Pearson's r) produced from the bivariate correlation analysis is shown in Table 5.

[Table 5]

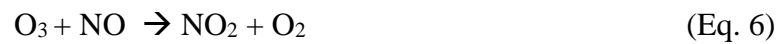
From the results in Table 5, a clear spatial scale effect was shown from the correlation between land use categories and air quality in KLEMUR. The air pollutants (NO, NO₂, NO_x and CO) concentrations are positively significant to the built-up lands within the 5 km buffer, whereby the correlation coefficients for the built-up lands are 0.37, 0.48, 0.42 and 0.52, respectively. The results also show the negatively significant correlation with the land use categories of vegetation and water bodies as well, except for the NO concentration which is

not significantly correlated with vegetation ($p = 0.11$) and the CO concentration which is not significantly correlated with water bodies ($p = 0.11$). These results indicate that the increase of built-up lands and the reduction of vegetation and water bodies has led to the increase of NO, NO₂, NO_x and CO which are contributed from vehicular emissions due the road network expansion in KLEMUR (Grote et al., 2016; Leh et al., 2014; Zhang & Batterman, 2013). These findings were consistent with a study by Cho and Choi (2014) and Azhari et al. (2018), where air pollutant levels, especially of NO₂ and CO, rose as the built-up lands increased and the green areas decreased, complying with the demand for urban growth.

The spatial scale effect shows different outcomes for SO₂, PM₁₀ and O₃ concentrations. The concentrations of SO₂ and PM₁₀ are associated with all land use categories within the 1 km buffer, whereas SO₂ concentrations show a significantly positive correlation with built-up lands ($r = 0.51$) and a significantly negative one with vegetation ($r = -0.45$) and water bodies ($r = -0.38$). The PM₁₀ concentration is negatively significant with water bodies only ($r = 0.38$). Whereas the O₃ concentration is associated with the land use categories within different buffer scales and has a significantly positive correlation with water bodies only ($r = 0.59$). These results indicate that land use categories have a direct impact on SO₂ concentrations while an indirect impact on PM₁₀ and O₃ concentrations in KLEMUR as the air pollutants were most affected by the built-up lands that consist of industrial and residential areas (Zahari et al., 2016). Furthermore, the air pollutants were found to decrease as there was a rise in green areas and water bodies promoting the dispersion of air pollutants (Cho & Choi, 2014; She et al., 2017; Xu et al., 2016; Zhu et al., 2019).

From the correlation analysis, the buffer radius with the highest correlation coefficient is considered as the optimum radius scale. Table 5 shows the values of the correlation coefficients with the optimum radius scale in bold. There are great differences in the optimum radiuses concerning different air pollutants that were affected by land use categories in KLEMUR and these findings were consistent with a study by Xu et al. (2016). Since the

emissions of SO₂ and PM₁₀ are mainly attributed from the point sources, the optimum radiuses are relatively smaller between different land use categories and air pollutants (SO₂ and PM₁₀) (Pereira et al., 2007; Popescu et al., 2011). While the optimum radiuses for NO, NO₂, NO_x and CO are relatively larger because the emissions mostly originate from non-point sources such as traffic emissions (Grote et al., 2016; Zhang & Batterman, 2013). The optimum radius between built-up lands and O₃ is relatively small compared to the optimum radiuses between vegetation and water bodies. However, it is moderate when compared to the other air pollutants. This is likely caused by emissions of O₃ from its precursors like NO_x and CO which mostly originate from vehicle emissions and high combustion industrial activities as well as VOCs from natural and anthropogenic sources (Jhun et al., 2015). The concentration of NO₂ with little UV radiation (424 nm) has the ability to produce O³P atoms and prompts O₃ formation (Eq. 4 and Eq. 5). NO is capable of reducing the O₃ concentration due to titration processes and the regeneration of NO₂ in the environment. The Eq. 4, Eq. 5 and Eq. 6 reactions result in a null cycle when no other chemical species are involved. The formation of NO₂ can also be accelerated by other oxidants such as RO_x (RO_x = OH + OH₂ + RO₂) from VOCs in the atmosphere (Eq. 7 and Eq. 8) (Wang et al., 2017).



The relationship between land use categories and the air pollutants in KLEMUR shows there is a significant impact on most of the air pollutants emissions depending on variations of land use activities. Built-up lands in KLEMUR have a positive correlation with all the air

pollutant emissions focused on in the study, except for PM₁₀ and O₃. Vegetation has a negative correlation while water bodies show both kinds of correlation towards the air pollutants. This study finding is consistent with previous studies that determined the impacts of urban land expansion on air pollution and where their data findings demonstrated that urban land expansion had a significant relationship and influenced air pollution (Cárdenas Rodríguez et al., 2016; Huang & Du, 2018).

4 Conclusion

This paper has examined the relationship between urban land use changes and urban air quality in KLEMUR, the largest conurbation area in Malaysia, using spatio-temporal perspectives. Overall, the average concentrations of air pollutants in KLEMUR over the 16-year study period are below the Malaysian Ambient Air Quality Standards (MAAQS). Only the PM₁₀ concentrations exceeded the value suggested by the World Health Organization (WHO) guidelines for ambient PM₁₀ concentrations (50 µg/m³). The results showed that land use categories in KLEMUR have a significant impact ($p < 0.10$) on NO, NO₂, NO_x, CO and SO₂ since the correlation analysis showed the air pollutants were positively associated with built-up land ($r = 0.37, 0.48, 0.42, 0.52$ and 0.51), negatively associated with vegetation ($r = -0.32, -0.42, -0.36, -0.49$ and -0.45) and also negatively associated with water bodies ($r = -0.41, -0.39, -0.41, -0.33$ and -0.38). The correlation results also indicated that land use categories have an insignificant impact on the PM₁₀ and O₃ since they are not significantly correlated with built-up land nor vegetation ($p > 0.10$). The land use trends within the 16-year period showed an increase in built-up lands (4.0%) and simultaneous reduction in vegetation (3.3%) and water bodies (2.3%). The highest percentage of land use change in the 5 km buffer from the continuous air quality monitoring stations was recorded at Petaling Jaya station where built-up lands in that area increased by 16.5% and the vegetation areas decreased by 16.2%. Whereas the highest percentage of land use change in water bodies was recorded at Bandaraya Melaka

station with 8.2% of destruction. The transition in land use was driven by economic development and population growth which in turn led to road network expansion, coastal reclamation activities and industrialization.

The overall trend for air pollutant concentrations in KLEMUR showed a rising trend, except for the SO₂ concentration which declined gradually, as impacted by the implementation of an emissions reduction plan aimed at improving motor vehicle technology and reformulating fuels to emit less sulphur. The reduction of SO₂ in KLEMUR within the 16 years gave positive feedback to policymakers and led to the continuous implementation of the current policies, improving them so that other air pollutants concentrations could be reduced while opting for sustainable urban land use planning. The PCA results demonstrated that 52.68% of the total variance in KLEMUR was influenced by emissions from motor vehicles and industrial activity since the strong factor loadings (> 0.70) consisted of NO, NO₂, NO_x, CO and SO₂. In addition, the results from the buffer analysis showed the optimum radiuses for NO, NO₂, NO_x, and CO are relatively larger (buffer: 5 km) because the emissions mostly originated from non-point sources such as traffic emissions. While the optimum radiuses for SO₂, PM₁₀ and O₃ are relatively smaller (buffer: 1-3 km) because the emissions mostly originated from point sources such as industrial areas. Different land use categories have impacted on different air pollutants variously, whether on the magnitude of correlation or spatial scales effect of correlation. The land use changes in KLEMUR are evidence of the critical urban transition driven by the demands to meet the city's needs. These changes have caused major stress and worsened the quality of the urban air.

The process to improve air quality will require further time and effort as air pollution problems cannot be solved completely by relying on monitoring, emission controls or technological advancements alone. These study findings suggest land use changes and patterns do affect air quality, thus strategies for land use planning for urbanization must take into consideration requirements to reduce the intensity of air pollutions while maintaining good air

quality in urban areas. The quantitative relationship between land use and air quality is critical in providing policymakers with alternative ways to improve air quality. Thus, it is recommended to generate an inventory of the emission sources for data sharing in order to establish joint operations in controlling the pollution within the cities in KLEMUR. Improvements in land use policies also need to be cast in more open multi-level partnerships with shared common core values. This will enable there to be a livable extended mega urban region which is ordered by a more ethical consideration of wealth sharing, improved decision making for future development, and where sustainable development is prioritized while a high quality environment is maintained. These strategies should include the apportionment of significant areas of designated green spaces, the conservation of forested areas and water bodies areas as well as the implementation of strict guidelines aimed at reducing the production of air pollutants during urban development and daily urban activity processes. Therefore, the needs to amend current policies related to land use change is critical in ensuring that future development activities have a minimum impact on the environment, ensure a better quality of life for the increasing demands of a growing population and also improve urban air quality without constraining economic growth.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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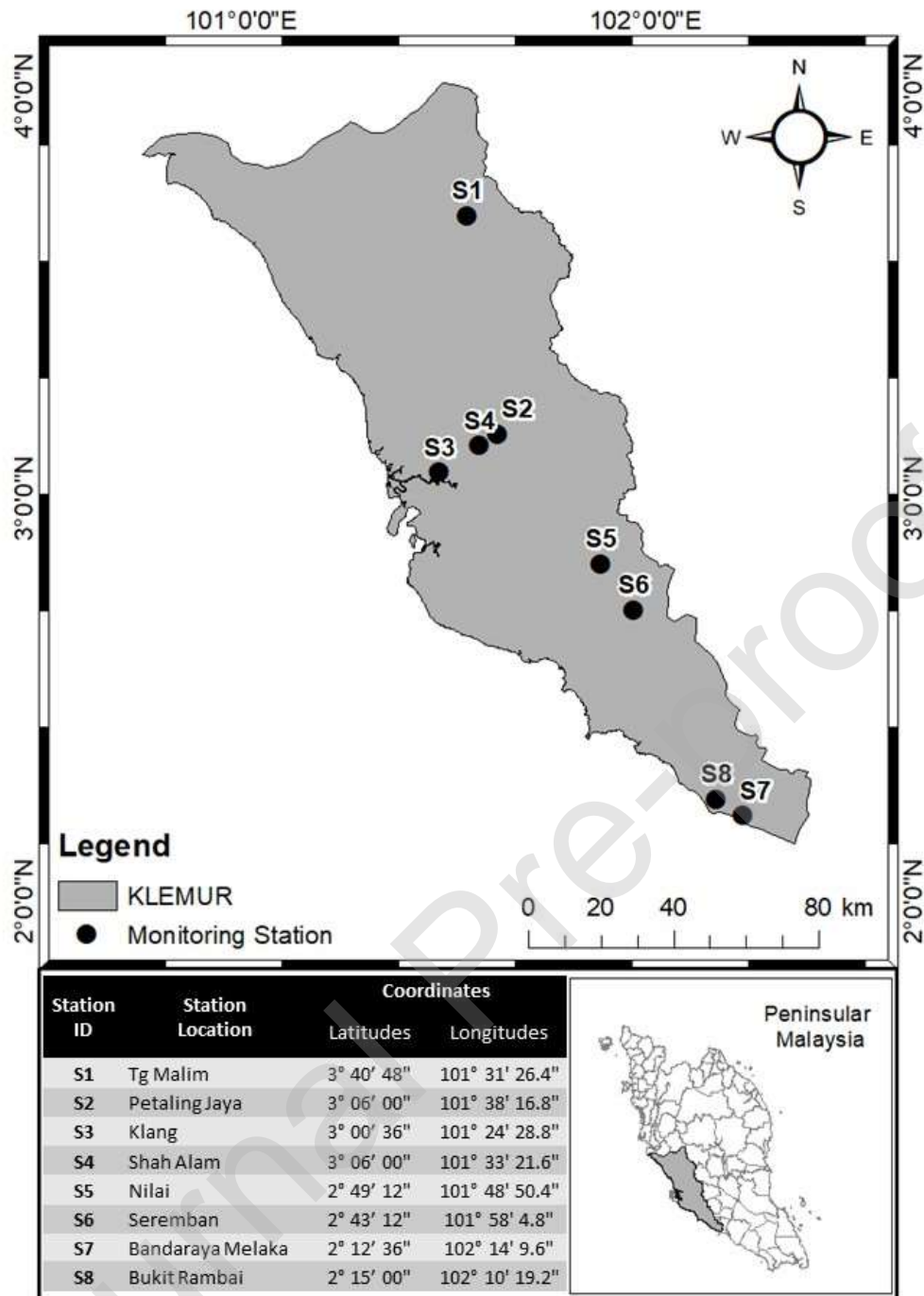
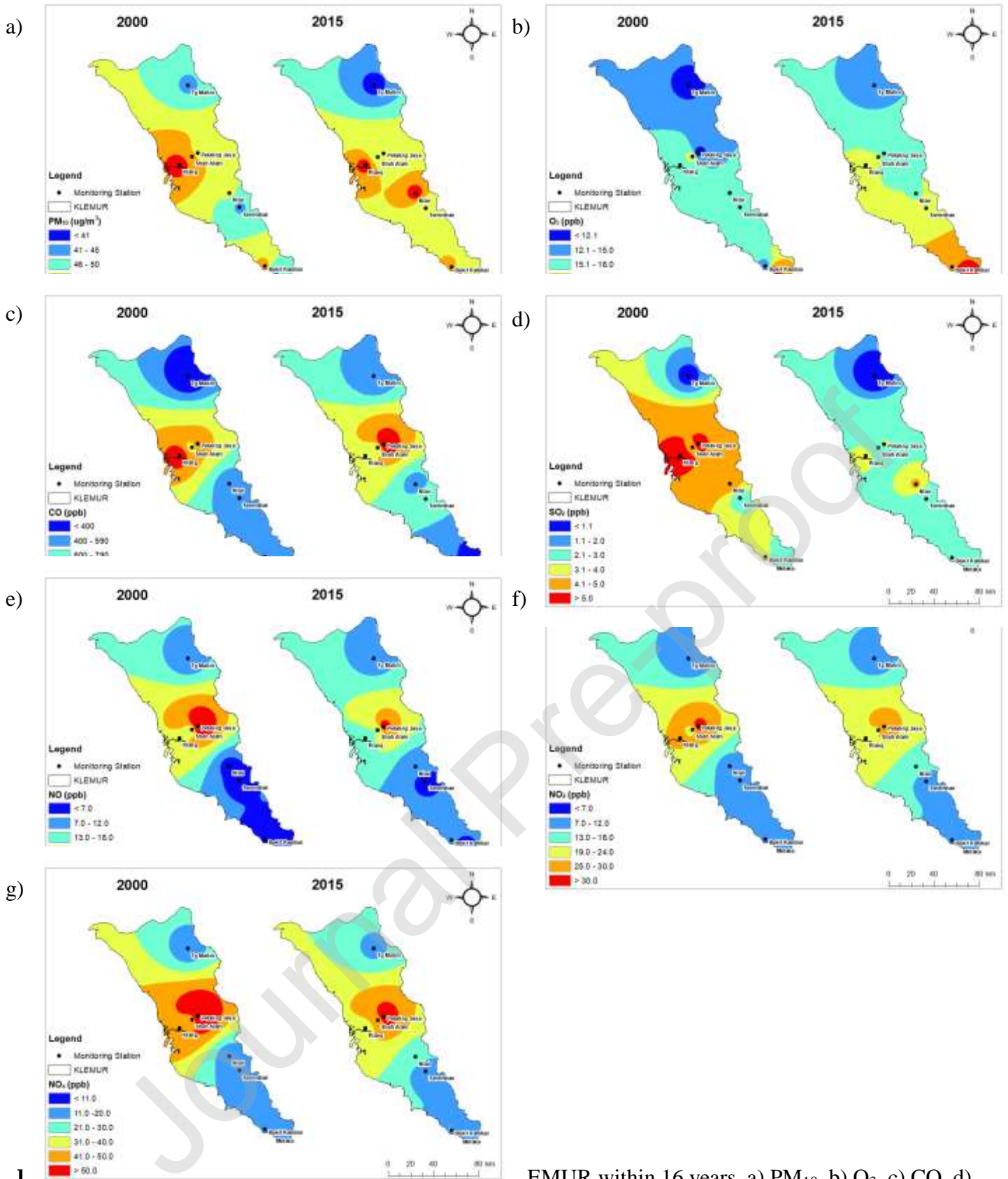


Fig. 1 The location of selected continuous air quality monitoring stations in the KLEMUR located at the west coast of Peninsular Malaysia.



1 JEMUR within 16 years. a) PM₁₀, b) O₃, c) CO, d) SO₂, e) NO, f) NO₂ and g) NO_x

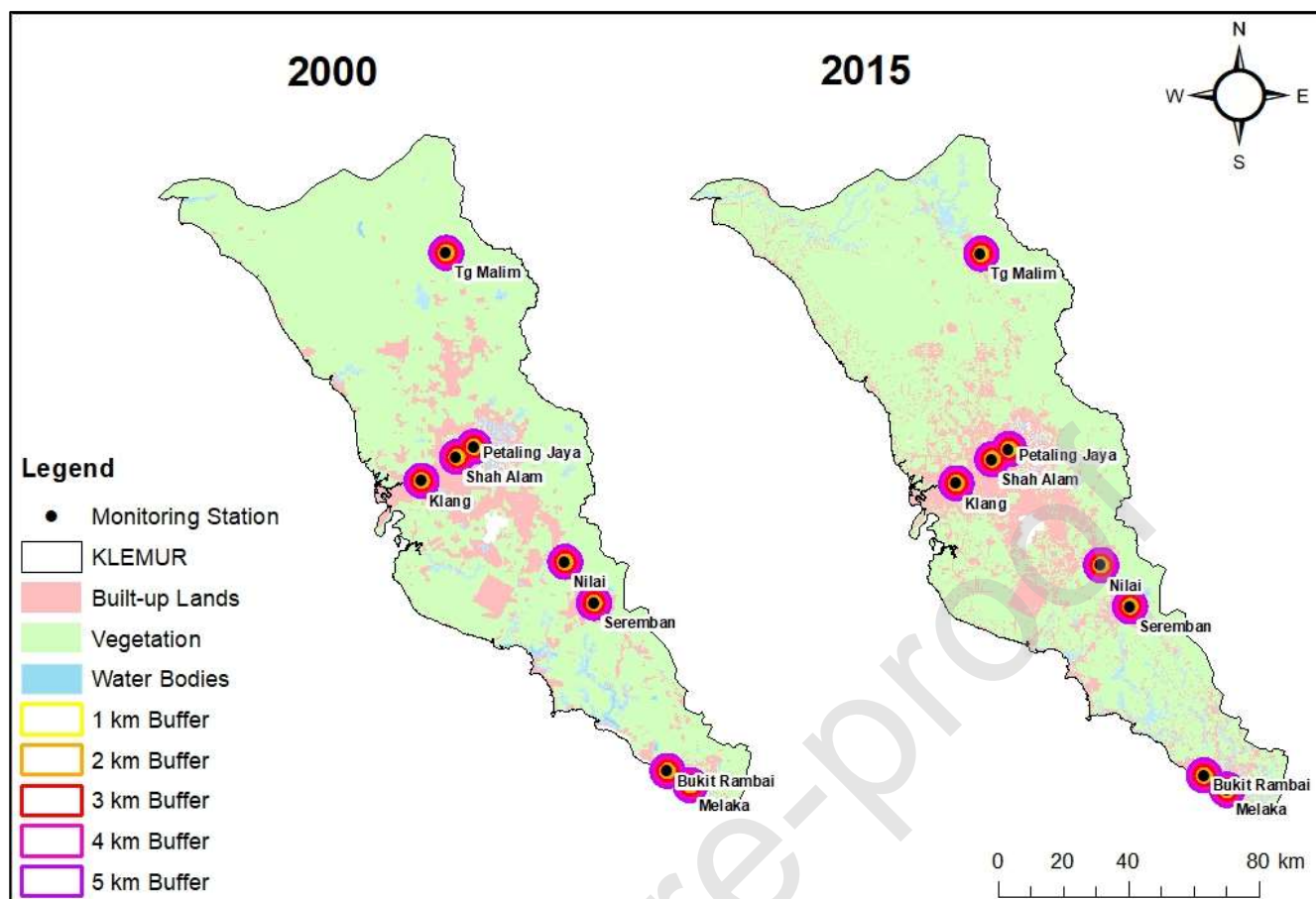


Fig. 3 The map shows land use activities in KLEMUR for year 2000 and 2015.

Table 1 The measurement of the variables that were recorded from the selected continuous air quality monitoring stations in KLEMUR.

Variables	Sensor type	Brand	Method	Detection Limit	Accuracy
O ₃	Teledyne API Model 400/400E	Teledyne Technologies Inc., USA	using Beer-Lambert law	0.4 ppb	0.5%
CO	Teledyne API Model 300/300E	Teledyne Technologies Inc., USA	using infrared energy absorption	40 ppb	0.5%
SO ₂	Teledyne API Model 100A/E	Teledyne Technologies Inc., USA	using the UV fluorescence method	0.4 ppb	0.5%
NO, NO ₂ and NO _x	Teledyne API Model 200A/200E	Teledyne Technologies Inc., USA	using the chemiluminescence method	0.4 ppb	NO _x = 0.5%

	Maximum	165.00	150.00	129.00	174.00	154.00	134.00	151.00	124.00	(1 h)
	Mean	17.66	15.44	17.52	21.59	17.86	19.35	21.68	18.62	
	Standard deviation	18.63	17.68	16.79	23.57	16.91	17.69	15.86	14.93	
NO (ppb)	Minimum	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	-
	Maximum	108.00	374.00	329.00	231.00	149.00	104.00	143.00	150.00	
	Mean	8.21	39.70	20.72	18.52	9.01	5.24	4.59	8.61	
	Standard deviation	8.75	35.46	24.27	22.52	11.86	6.60	7.53	9.47	
NO ₂ (ppb)	Minimum	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	170
	Maximum	61.00	216.00	127.00	142.00	80.00	55.00	54.00	86.00	(1 h)
	Mean	6.99	30.82	21.67	19.98	12.84	7.80	9.15	9.70	
	Standard deviation	4.52	13.86	11.60	11.67	8.19	5.44	6.66	5.34	
NO _x (ppb)	Minimum	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	-
	Maximum	139.00	424.00	377.00	274.00	174.00	123.00	166.00	176.00	
	Mean	13.94	70.16	41.15	37.48	20.92	12.29	12.49	17.41	
	Standard deviation	10.42	41.04	31.15	29.05	16.70	9.26	11.45	12.73	
SO ₂ (ppb)	Minimum	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	130
	Maximum	25.00	96.00	179.00	139.00	116.00	78.00	60.00	102.00	(1 h)
	Mean	1.52	5.27	5.58	4.38	4.65	3.08	2.75	3.81	
	Standard deviation	1.03	4.71	5.52	4.67	5.46	2.94	2.24	3.44	
CO (ppb)	Minimum	10.00	10.00	10.00	10.00	10.00	10.00	10.00	10.00	30000
	Maximum	5980.00	10410.00	15510.00	9790.00	4460.00	8160.00	10370.00	11450.00	(1 h)
	Mean	394.40	1429.0	1102.0	871.00	568.70	536.90	533.00	589.80	
	Standard deviation	276.10	825.69	863.68	623.67	329.58	384.08	404.06	369.52	

Table 3 The PCA loadings after the varimax rotation. The bold values in the table indicate the strong factor loading (> 0.7).

Variables	F1	F2	F3
PM ₁₀	0.364	-0.144	0.877
O ₃	-0.264	0.916	-0.180
NO	0.910	-0.266	0.043
NO ₂	0.949	-0.015	0.131
NO _x	0.975	-0.170	0.071
SO ₂	0.722	0.358	0.412
CO	0.926	-0.214	0.213
Temperature	0.143	0.733	-0.603
Humidity	-0.061	-0.953	-0.001
Wind Speed	-0.401	0.863	-0.008
Eigenvalue	5.27	2.88	1.03
Variability (%)	52.68	28.82	10.25
Cumulative (%)	52.68	81.50	91.74

Table 4 The areas proportion of different land used categories around each of the continuous air quality monitoring stations within 5 km buffer in KLEMUR.

Station ID	Station Location	Area Proportion (2000)			Area Proportion (2015)			Rate of change (%)		
		Built-up lands	Vegetation	Water bodies	Built-up lands	Vegetation	Water bodies			
S1	Tg Malim	1.0%	96.4%	2.6%	12.5%	85.8%	1.7%	↑11.5%	↓-10.6%	↓-0.9%
S2	Petaling Jaya	67.2%	32.3%	0.6%	83.7%	16.0%	0.3%	↑16.5%	↓-16.2%	↓-0.3%
S3	Klang	64.6%	30.5%	4.9%	76.8%	19.9%	3.4%	↑12.2%	↓-10.7%	↓-1.5%
S4	Shah Alam	79.6%	20.3%	0.1%	95.7%	4.3%	0.0%	↑16.1%	↓-16.0%	↓-0.1%
S5	Nilai	22.5%	76.6%	0.9%	26.6%	73.4%	0.0%	↑4.0%	↓-3.1%	↓-0.9%
S6	Seremban	86.7%	12.6%	0.7%	87.8%	10.5%	1.7%	↑1.1%	↓-2.1%	↑1.0%
S7	Melaka	44.4%	14.2%	41.4%	46.8%	20.0%	33.2%	↑2.4%	↑5.8%	↓-8.2%
S8	Bukit Rambai	22.9%	51.0%	26.1%	29.9%	51.0%	19.1%	↑7.0%	=0.0%	↓-7.0%

Table 5 Result of bivariate correlation analysis between land use categories and air pollutants in KLEMUR. The bold values represent the highest correlation coefficient between the same land use category and a certain air pollutant (r = correlation coefficient, * $p < 0.10$, ** $p < 0.05$, *** $p < 0.01$).

Land Use Category	Buffer (km)	PM ₁₀		O ₃		NO		NO ₂		NO _x		CO		SO ₂	
		r	p	r	p	r	p	r	p	r	p	r	p	r	p
Built-up Lands	1	0.19	0.24	0.21	0.22	0.2	0.4	0.3	0.45	0.51	0.02				
	2	0.19	0.24	0.27	0.16	0.1	0.3	0.2	0.41	0.46	0.04				
	3	0.18	0.25	0.28	0.14	0.2	0.3	0.2	0.42	0.44	0.04				
	4	0.12	0.32	0.24	0.19	0.2	0.4	0.3	0.45	0.43	0.04				
	5	0.11	0.34	0.14	0.30	0.37	0.08	0.48	0.03	0.42	0.05	0.52	0.02	0.46	0.04
Vegetation	1	0.10	0.35	0.24	0.19	0.2	0.4	0.3	0.44	0.45	0.04				
	2	0.08	0.38	0.25	0.18	0.2	0.3	0.3	0.46	0.42	0.05				
	3	0.06	0.41	0.31	0.12	0.2	0.3	0.3	0.44	0.39	0.07				
	4	0.05	0.43	0.33	0.11	0.2	0.3	0.3	0.44	0.36	0.09				
	5	0.07	0.39	0.29	0.14	0.32	0.11	0.42	0.05	0.49	0.03	0.38	0.07		
Water Bodies	1	0.38	0.07	0.44	0.05	0.3	0.3	0.3	0.28	0.15	0.38	0.07			

2	- 0.3 7*	0. 08	0.50 **	0. 03	- 0.3 2	0. 11	- 0.2 9	0. 14	- 0.3 1	0. 12	- 0.26 17	0. 17	- 0.36 *	0. 09
3	- 0.3 2	0. 12	0.56 **	0. 01	- 0.3 3	0. 10	- 0.2 7	0. 15	- 0.3 1	0. 12	- 0.23 19	0. 19	- 0.31 12	0. 12
4	- 0.1 7	0. 26	0.59 **	0. 01	- 0.3 9*	0. 07	- 0.3 5*	0. 09	- 0.3 8*	0. 07	- 0.30 13	0. 13	- 0.32 11	0. 11
5	- 0.0 6	0. 41	0.57 **	0. 01	- 0.4 1*	0. 06	- 0.3 9*	0. 07	- 0.4 1*	0. 06	- 0.33 11	0. 11	- 0.31 12	0. 12