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ORIGINAL ARTICLE

Long-Term Hydrologic Impact Assessment of Non-point Source Pollution Measured Through Land Use/Land Cover (LULC) Changes in a Tropical Complex Catchment

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Abstract

The contribution of non-point source pollution (NPS) to the contamination of surface water is an issue of growing concern. Non-point source (NPS) pollutants are of various types and altered by several site-specific factors making them difficult to control due to complex uncertainties involve in their behavior. Kelantan River basin, Malaysia is a tropical catchment receiving heavy monsoon rainfall coupled with intense land use/land cover (LULC) changes making the area consistently flood prone thereby deteriorating the surface water quality in the area. This study was conducted to determine the spatio-temporal variation of NPS pollutant loads among different LULC changes and to establish a NPS pollutant loads relationships among LULC conditions and sub-basins in each catchment. Four pollutants parameters such as total suspended solids (TSS), total phosphorus (TP), total nitrogen (TN) and ammonia nitrogen (AN) were chosen with their corresponding event mean concentration values (EMC). Soil map and LULC change maps corresponding to 1984, 2002 and 2013 were used for the calculation of runoff and NPS pollutant loads using numeric integration in a GIS environment. Analysis of Variance (ANOVA) was conducted for the comparison of NPS pollutant loads among the three LULC conditions used and the sub-basins in each catchment. The results showed that the spatio-temporal variation of pollutant loads in almost all the catchments increased with changes in LULC condition as one moves from 1984 to 2013, with 2013 LULC condition found as the dominant in almost all cases. NPS pollutant loads among different LULC changes also increased with changes in LULC condition from 1984 to 2013. While urbanization was found to be the dominant LULC change with the highest pollutant load in all the catchments. Results from ANOVA reveals that statistically most significant ($p < 0.05$) pollutant loads were obtained from 2013 LULC conditions, while statistically least significant ($p < 0.05$) pollutant loads were obtained under 1984 LULC condition. This reveals the clear effect of LULC changes on NPS pollution. The findings of this study may be useful to water resource planners in controlling water pollution for future planning.

Keywords Hydrologic impact · Remote sensing · Non-point source pollution · GIS · Malaysia

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1 Introduction

Water balance of a watershed is influenced by land use/land cover changes (LULC) (Fohrer et al. 2001). LULC change is the single most prominent element responsible for changes at both regional and global scale which results to several types of natural and biological changes (Bai, et al. 2008; Wang and Wang 2013; Li et al. 2016). A watershed becomes more hydrologically active when it develops with changing runoff components, stream flow and flood volume. The effect of LULC on storm runoff generation is very complicated. Several studies in the past have identified that LULC having a strong impact on water quality (Thanapakpawin et al. 2007; Zaimes et al. 2008; Shen et al. 2010) predominantly because of non-point source pollution (NPS) which are known to have a direct relationship with LULC change (Girmay et al. 2009). LULC changes adversely affect NPS pollutant loads by altering their sources and the way they are being transferred into water bodies.

As water is being drained from the land surface, it has the tendency to carry along with it residues of several types to other water bodies. Under the first flush phenomena, surface run off is a major source of non-point source pollution (NPS). The type of contaminant depends on the run off that is associated with the LULC and the Event Mean Concentrations (EMC) values of the pollutant load (Engel 2001; Novotny 2003). EMC quantifies the volume of pollutants conveyed per unit volume of runoff. For example, the major contaminants from run off that will pollute agricultural land use will be nutrients (mostly nitrogen and phosphorus) and sediments. Runoff from highly urbanized areas, on the other hand, may be polluted with rubber fragments, heavy metals, in addition to sodium and sulfate from road (Tong and Chen 2002). The problem of NPS pollution is an issue of great concern as it poses a great risk to water quality in developed countries (US EPA 2009).

Unlike point source pollution whose sources are known, NPS pollution is peculiar to complex mechanisms and techniques that are random and sporadic in occurrence. In addition to this, NPS pollution poses uncertainty with regards to discharge in channels and amounts, variability in both temporal pollution loads which results to difficulties in monitoring, simulation, treatment as well as control. To tackle the risk of NPS pollution, it is vital to have precise simulations and estimations of NPS (Shen et al. 2012).

Quantification of several kinds of NPS pollution is a key issue for successful land use planning as well as in alleviating the hazard (Engel et al. 2003; Zhang et al. 2011). Pollutant load estimation carried out through monitoring activities is a complex process that involves precise computation of both pollutant concentration (EMC) as well as runoff and accurate calculations that are mostly based on statistical methods. Therefore, it is important to set up initial monitoring activities of NPS pollutants for good load estimation (Meals et al. 2013). Comprehensive knowledge of the areas' topography and NPS sources is a prerequisite when characterizing pollutants. Identification and location of NPSs of pollution is desirable for pollutant loads and should be fully evaluated.

Several studies have been conducted in the past in Malaysia based on spot field evaluation on NPS pollution (e.g., Yusop et al. 2005; Chow and Yusop 2006; Eisakhani et al. 2009; Nazahiyah et al. 2007; Chow et al. 2011). It should, however, be noted that, the spatio-temporal variation of NPS pollution cannot be attained through spot field investigation and short-term monitoring, therefore, researches should inevitably be conducted using mathematical models (Li et al. 2016). The Kelantan River basin in the north-eastern part of Peninsular Malaysia was chosen as the study area due to its constant and frequent incidences of flooding which leads to build up of NPS pollution load in the area. The watershed is under illegal and unrestricted land cover conversion without giving attention to the environmental consequences which has altered the natural hydrologic system of the basin giving rise to several incidences of flood. This research has three main objectives; to determine the spatio-temporal variation of NPS pollutant loads among different catchments in Kelantan river basin, to determine the temporal variation of NPS pollutant loads among different LULC changes and to establish NPS pollutant loads relationship among different sub-basins in each catchment and LULC conditions.

2 Study Area

Kelantan River basin is positioned in the north-eastern part of Peninsular Malaysia between latitudes 4° 40′ and 6° 12′ north, and longitudes 101° 20′ and 102° 20′ east. The capital city of Kelantan is Kota Bharu which is situated at the Northern part of the state. Kelantan state occupies 4.40% of Malaysia's total area with a total of 15,099 km^2. The state has an estimated population of 1.539 million. The maximum length and breadth of the catchment are 150 and 140 km, respectively. The length of the main river is about 248 km long which drains an area of about 13,100 km^2, occupying more than 85% of the Kelantan state. The estimated quantity of the annual precipitation in the basin is about 2383 ± 120 mm, a large amount of which occurs during the north-east monsoon between mid-October and mid-January. The basin has an estimated runoff discharge of 500 $m^3\ s^{-1}$ (DID, 2000). The average annual temperature at Kota Bharu is 27.5 °C with mean relative humidity of 81%. The average flow of the Kelantan River measured at Guillemard Bridge is 557 $m^3\ s^{-1}$.

The Kelantan River divides into Galas and Lebir Rivers near Kuala Krai about 100 km from the river mouth. Galas River is formed by the junction of Nenggiri and Pergau Rivers. The origin of Nenggiri River is from the south-western part of the main mountain range. While the origin of Lebir River is from Tahan mountain range. The flow direction of Kelantan River system is northward where it passes along major towns like Kuala Krai, Tanah Merah, Pasir Mas and Kota Bharu, before finally discharging into the South China Sea. The majority of the catchment is steep mountainous area rising to a height of 2135 m, occupying 95% of the area while the rest is undulating land. The mountainous areas are covered primary with virgin forest while rubber and some paddy are cultivated in the lowlands. The eastern and western parts of the watershed, consists of mountains of various ranges, while the soil cover is granitic comprising a combination of fine to coarse sand and clay. The soil cover is approximately a meter deep on average but depths of more than 18 m may be encountered in localized areas. In the extreme of the southern half of the basin, the major soil type is fine sandy loam, which has its depth rarely exceeding a few meters. The other part consisting of almost one-third of the basin is covered by a variable soil cover that varies in depth, which is about 1 m to more than 9 m. The forested areas, mostly in the Lojing highlands, are experiencing serious logging activities which some people believe is the major cause of recent floods in the basin. The map of the study area is shown in Fig. 1.

3 Methodology

The flow chart adopted in this study is shown in Fig. 2. The framework is divided into four components. First, digital elevation model, LULC maps, remote sensing images and soil maps were prepared in a GIS environment using ArcMap 10.3. Second, ArcCN within the ArcMap environment was used to calculate curve numbers (CN) and runoff amount. The third component involves the calculation of pollutant load using numeric integration and, lastly the relationship among LULC condition based on NPS pollution loads were statistically analyzed.

In this paper, pollutant loads were estimated using numeric integration in a geographic information system (GIS) environment. Land use maps corresponding to 1984, 2002 and 2013 and SPOT 5 images were used to obtain the

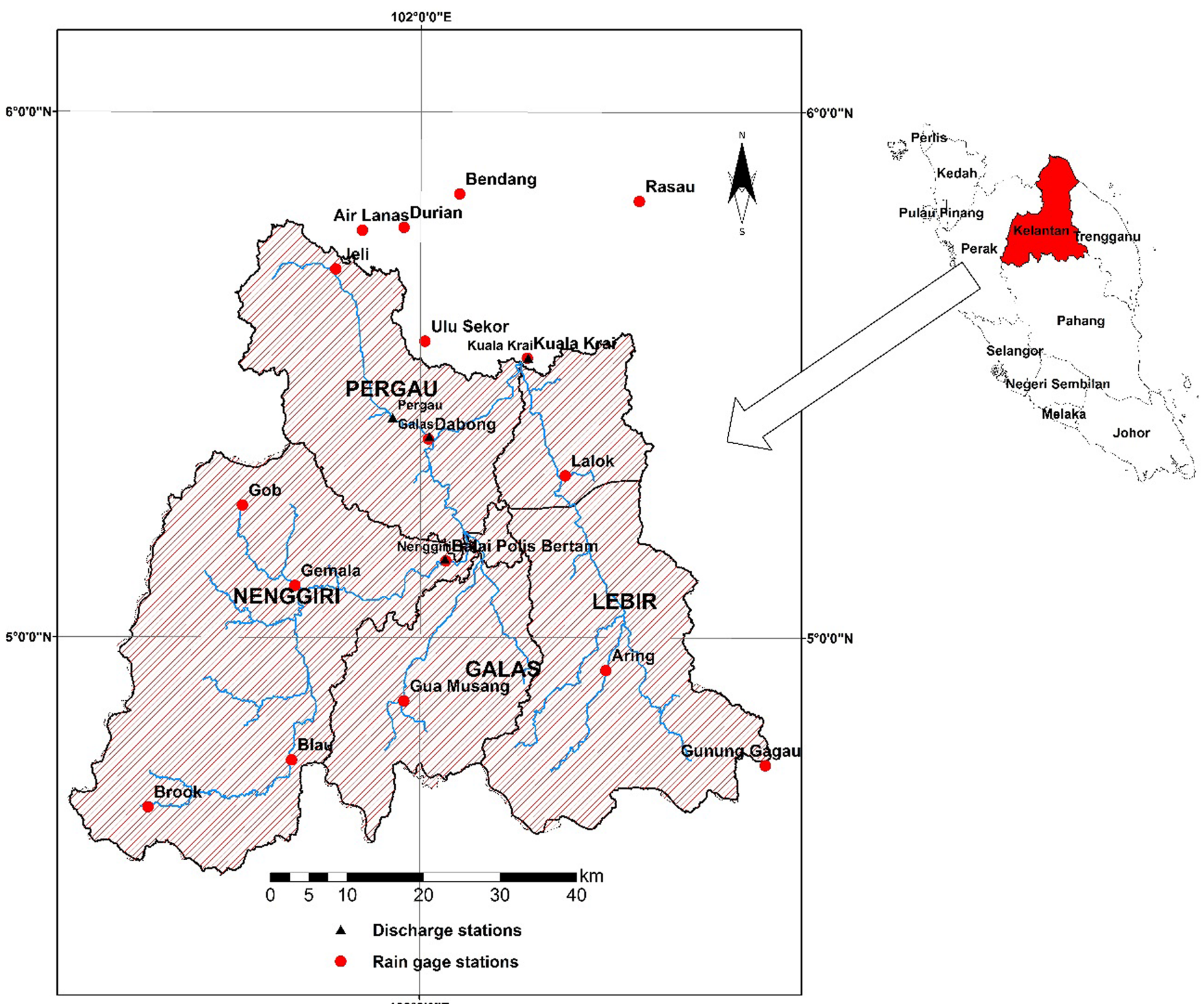

Fig. 1 Map of Kelantan river basin showing Galas, Pergau, Lebir, and Nenggiri catchments with rain gauge and discharge stations

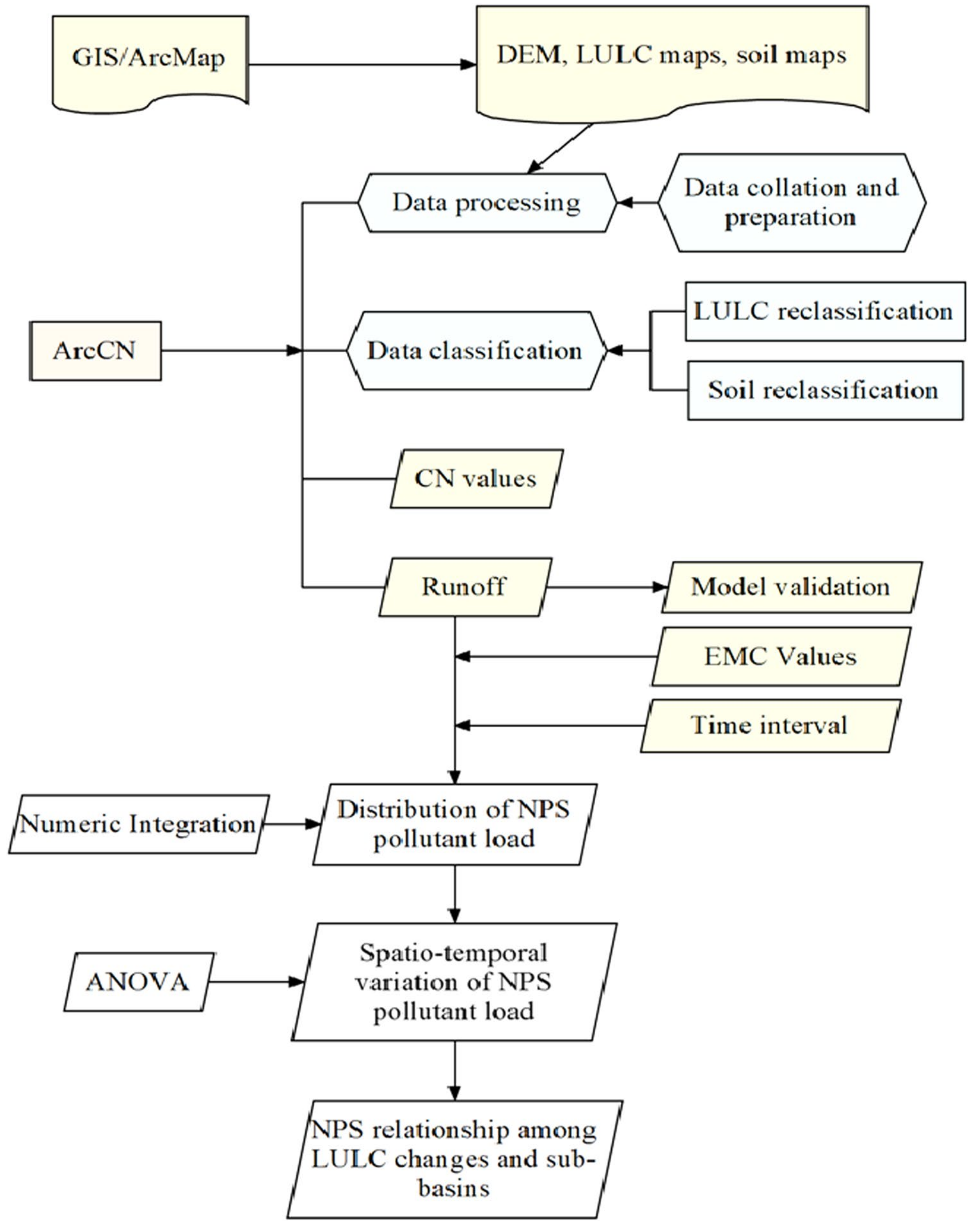

Fig. 2 Methodological framework adopted in this study

LULC change of the watershed. The digital elevation model (DEM) was used to extract the physiographic characteristics in the area, soil maps were used to derive the soil properties, long-term daily hydrological gauged data from 1984 to 2014 were obtained. The findings of this study will of serious help to water resource planners in controlling water pollution for future planning.

3.1 Data Sources and Preparation

Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER) Global Digital Elevation Model (GDEM) with 30 m spatial resolution was used as the Digital Elevation Model (DEM) source in this research for extracting the physiographic features of the basin. HEC-GeoHMS which is an extension of ArcMap software was used in delineating the basin. Four major sub-basins were extracted from the DEM, e.g., Galas, Nenggiri, Lebir, and Pergau. These basins were further divided into smaller sub-basins considering geomorphological similarities to increase the precision of runoff and NPS load estimation. LULC maps corresponding to the three LULC conditions (1984, 2002 and 2013) as well as soil maps are shown in Fig. S1, while the DEM for the four basins is shown in Fig. S2. Table S1 summarizes the description of datasets used in this study.

3.2 Land Use Classes, Rainfall and Soil Type

Three land use maps corresponding to years 1984, 2002 and 2013 and soil series maps of the basin were obtained from the Department of Agriculture (DOA), Malaysia. Land use classes in the LULC maps were categorized into forest, paddy, agriculture, grassland, urbanization, cleared land, mangrove swamp, secondary forest, rivers, ponds and lakes, and mining for ease of analysis. Daily rainfall and runoff data were obtained from Department of Irrigation and Drainage, Malaysia for the period of 1984–2014 and annual average rainfall were used for calibration and validation of ArcCN (Zhan and Huang 2004). Rainfall and discharge stations are shown in Fig. 1. These stations were chosen in this study based on their complete records and data availability.

3.3 Runoff Estimation using ArcCN

The Soil Conservation Service Curve Number (SCS-CN) method was used in this study. It was based on a water balance and two fundamental hypotheses (SCS 1956).

$$Q = \frac{(P + I_a)^2}{P - I_a + S}, \quad P \geq 0.2S \tag{1}$$

$$S = \frac{25400}{\mathrm{CN}} - 254, \tag{2}$$

where Q is the direct runoff (mm), P is the rainfall (mm), I_a is the initial abstraction (mm), S is the potential maximum retention after runoff begins (mm). CN values were determined by intersecting each of the LULC map with the soil map using ArcCN script in ArcMap from the land/soil intersect file (Zhan and Huang 2004). To determine runoff for the year 1984, available rainfall stations were used to determine average rainfall for the year 1984 LULC condition. For the 2002 LULC condition, averages calculated from 1984 to 2002 were used for computing runoff for that year whereas, for 2013 LULC condition runoff was determined using averages calculated from 1984 to 2013. These averages were further divided by 12 keeping in mind 12 large rainfall events corresponding to each LULC condition. Fig. S3 shows rainfall trends in Kelantan River. More detailed description of the ArcCN method can be obtained from Zhan and Huang (2004).

3.4 Pollutant Load Estimation

Pollutant loads are expressed as mass or weight of a pollutant that is transferred through a cross-sectional area of water body (rivers, streams) at a specified time. It is expressed in mass units (kilograms, tons), although the time interval is inherent when pollutant loads are formed, it should, however, be distinct from context. The NPS pollutant load studied in this research are; total suspended solids (TSS), total phosphorus (TP), total nitrogen (TN) and ammonia nitrogen (AN). The load of each pollutant was calculated using numeric integration (Meals et al. 2013) carried out in a GIS environment with the formula given below;

$$\mathrm{Load} = \sum_{i=1}^{n} c_i q_i t_i, \tag{3}$$

where c_i is the event mean concentration in the i^{th} sample, q_i is the corresponding runoff values, and t_i is the time interval represented by the i^{th} sample, calculated using Eq. 4 below;

$$\frac{1}{2}\left(t_{i+1} - t_{i-1}\right). \tag{4}$$

3.4.1 Event Mean Concentration (EMC)

Event mean concentration is the mean concentration of an urban pollutant measured during a storm runoff event. It can also be defined as the total mass of total constituents discharged expressed over the total runoff volume (Huber 1993; Adams and Papa 2001). It can be expressed in the equation below;

$$\mathrm{EMC} = C = \frac{M}{V} = \frac{\int Q(t)C(t)\mathrm{d}t}{\int Q(t)\mathrm{d}t}, \tag{5}$$

where M is total mass of pollutant during the entire runoff (kg), V is total volume of runoff (m^3), $C(t)$ is time varying pollutant concentration (mg L^{-1}), $Q(t)$ is time variable flow (L s^{-1}), and t is total duration of runoff (s). It should, however, be noted that EMC results from a flow-weighted average does not represent the time average of the concentration. Table S2 shows the EMC values used in this study which is adopted from DID (2012).

3.5 Model Validation and Statistical analysis

Runoff values calculated from SCS-CN method were validated prior to pollutants load estimation. This procedure was carried out by comparing measured runoff values from 1984, 2002 and 2013; predicted LULC conditions using correlation coefficient (R) and Nash–Sutcliffe Efficiency (NSE) (Nash and Sutcliffe 1970). The procedures are presented in Eqs. (6) and (7) below.

$$R = \frac{\sum_{i=1}^{N}\left(Q_{\mathrm{Obs}} - \overline{Q}_{\mathrm{Obs}}\right)\left(Q_{\mathrm{sim}} - \overline{Q}_{\mathrm{sim}}\right)}{\sqrt{\sum_{i=1}^{N}\left(Q_{\mathrm{Obs}} - \overline{Q}_{\mathrm{Obs}}\right)^2\left(Q_{\mathrm{sim}} - \overline{Q}_{\mathrm{sim}}\right)^2}} \tag{6}$$

$$E = 1 - \frac{\sum_{i=1}^{N}(Q_{i\mathrm{Obs}} - Q_{i\mathrm{Sim}})^2}{\sum_{i=1}^{N}(Q_{i\mathrm{Obs}} - \overline{Q}_{i\mathrm{Obs}})^2}, \tag{7}$$

where, Q_{Sim} is the simulated discharge at time $t = i$, Q_{Obs} is the observed discharge at time $t = i$, $\overline{Q}_{sim}$ is the average simulated discharge $\overline{Q}_{Obs}$ is the average observed discharge; N is the number of observations.

Measured average annual rainfall data were used to calculate average annual runoff of each of the stations used in the study area for the year 1984 which represents 1984 LULC condition. For the 2002 LULC condition, the average annual runoff data from 1984 to 2002 was used, whereas for the 2013 LULC condition the average annual runoff data from 1984 to 2013 was calculated from rainfall during that period. These averages were further divided by 12 keeping in mind that there are 12 large runoff annual events corresponding to each LULC condition.

Statistical Analytical System (SAS) version 9.4 was used to carry out Analysis Of Variance (ANOVA) for the comparison of NPS pollutant loads among the three LULC conditions in this study for the entire watershed. Mean separation was carried out using least significance difference (LSD) and significant means were grouped using Tukey's range test. This comparison was aimed at exploring statistical similarities or differences existing among sub-basins in each catchment and different LULC conditions.

4 Results and Discussion

4.1 Results of Model Validation and Statistical Analysis

The results of the validation are expressed graphically and are shown in Fig. 3. Values of R ranged from 0.7198 to 0.9018 indicating strong positive correlation between the measured and estimated model values in all the catchments. Changes in R may be attributed to changes in LULC condition and its quantity which was seen visible in all the years

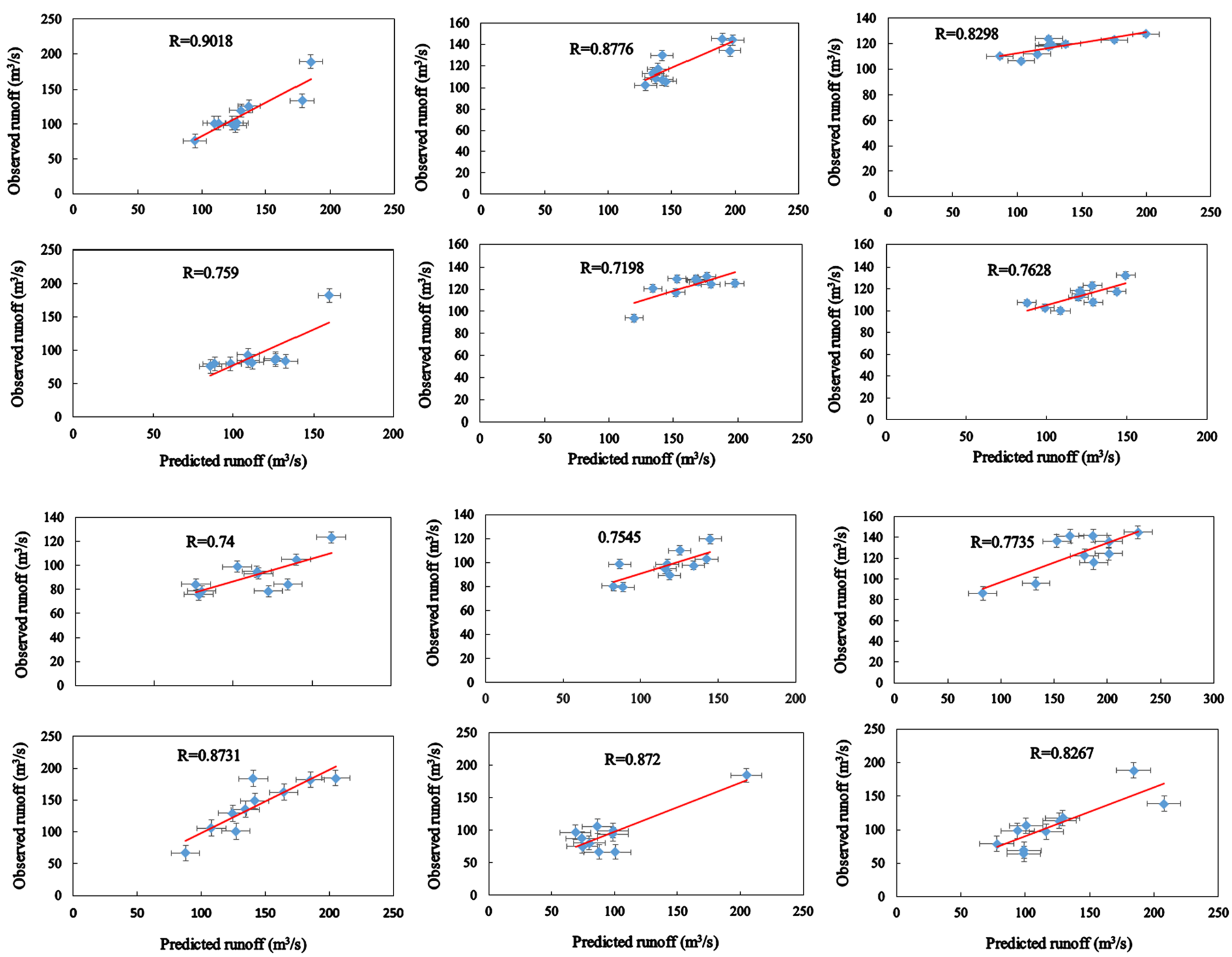

Fig. 3 Relationship between observed and predicted runoff values **a** Galas 1984 **b** Galas 2002 **c** Galas 2013 **d** Pergau 1984 **e** Pergau 2002 **f** Pergau 2013 **g** Lebir 1984 **h** Lebir 2002 **i** Lebir 2013 **j** Nenggiri 1984 **k** Nenggiri 2002 **l** Nenggiri 2013

as well as in the catchments (Ozdemir and Elbaşı 2015). NSE values in this study ranged from 0.45 to 0.65 indicating moderate to higher model performance in all the catchments across different LULC conditions. Generally, NSE statistic ranged from $-\infty$ and 1.0, where 1 is considered as the optimum. Values between 0.00 and 1.00 are considered satisfactory, while values ≤ 0.00 implied that the measured hydrological values produced better results than the simulated values, thus, indicating unacceptable model performance (Moriasi et al. 2007; Ozdemir and Elbaşı 2015).

4.2 Land Use Analyses of Past and Present LULC Changes

LULC changes were carefully analyzed in the major basins of Kelantan river basin from 1984 to 2014. The area (both in km^2 and %) occupied by each LULC change with its corresponding change according to 1984, 2002 and 2013 and is shown in Fig. 7. Deforestation was observed as the dominant LULC change across the watershed while agricultural activities were observed to increase non-uniformly across the catchments due to increase in urbanization in the area.

In Galas basin (Fig. S4), an estimated deforestation of about 51% was observed, while in Pergau (Fig. S4), it was found to be around 19.32% during 1984–2002. Unlike deforestation, grassland had increased by 15.77% in Galas and 7.11% in Pergau whereas secondary forest has undergone little or no changes in both locations. From 2002 to 2013 smaller area of the forested lands were lost to deforestation for both Galas basin (3.40%) and Pergau basin (7.11%). The major LULC changes observed during this period, were decreased in grassland from 15.85 to 0.46% in Galas and from 12.33 to 0.53% in Pergau. On the other hand, an increase in secondary forest can be seen (from 0.35 to 8.68%) in Galas and from 0.00 to 11.19% in Pergau.

The past and present LULC changes in Lebir and Nenggiri catchments are presented in Fig. 7c, d. In these basins deforestation was also observed during 1984–2002. In Lebir catchment, 24.91% deforestation was observed, whereas in Nenggiri it accounts for 17.69% of the total LULC change from 1984 to 2002. Agricultural activities and grassland both increased (15.77 and 10.92%, respectively) in Lebir and 2.49% and 14.84%, respectively, in Nenggiri. Deforestation occurred slightly in Lebir (2.91%) whereas afforestation was recorded in Nenggiri (6.98%) from 2002 to 2013. The major LULC change was decreased in grassland for both Lebir and Nenggiri while urbanization and cleared land all witnessed slight increase during the same period under study.

In all the catchments deforestation was found to be the foremost LULC change observed during 1984 to 2014. Deforestation can be attributed mainly due to intense logging and agricultural activities in the area that is believed to be a source of concurrent runoff activities as reported in previous studies in the same location by (Wan 1996; Jamaliah 2007; Adnan and Atkinson 2011). This increase in runoff has glaring effect on NPS pollution in the area.

4.3 Effect of Runoff on NPS Pollution in Kelantan River Basin

In all the catchments, runoff values increased with changes in LULC condition which also lead to build up in NPS pollution. Urbanization on the other hand was found to have increased as the number of years' increase is one of the major sources of surface runoff as well as NPS pollution due to increase in impervious surfaces that lead to increase in total pollutant load in the watershed. This increase contributed in altering the runoff dynamics of the basin as well as pollutant transport and delivery although not as rapid as deforestation. Previous surfaces that include all water bodies which have the tendency not to cause runoff were categorized as rivers, ponds and lakes. They were observed across the catchments predominantly under 2002 and 2013 LULC changes probably making the basins to have lower runoff and pollutant load deposition values under these conditions when compared with 1984 LULC condition where the catchment is dominated by forest.

For pollutant loads like TN, AN and TP that are associated with runoff from agricultural activities and are commonly considered as non-urban pollutants, it will be anticipated that transformation of non-urban land use types to urban land use type will lead to decrease of these pollutants in this watershed (Bhaduri et al. 2000). However, this is not the case in this study. The results showed that the transformation of non-urban land use types from 1984 to 2002 had caused an increase of about 0.54 and 0.43% in Pergau and Lebir respectively. This has led to an increase in TP by about 36% and TN by about 0.17% in Lebir and increase in TP by about 24% and TN by about 24% in Pergau. The rapid increase in agricultural activities from 7.20 to 25.72% in Pergau and that in Lebir from 8.29 to 24.06% is attributed to cause this increase of TP, TN, AN and TP during this period. Even though pollutant loads in urbanized land uses are produced at a slower rate than in non-urbanized land use types, one of the major consequences of urbanization is increase in runoff, which contribute enormously to NPS pollution build up. Another factor that may have influenced runoff, which directly affect the NPS load in the watershed, is the presence of high sloping areas. High values of pollutant loads recorded across the watershed may be due to undulating areas. Since the bigger the slope, the more the runoff as well the more the NPS pollution.

4.4 Spatio-Temporal Variation of NPS Pollutant Loads

The results of the spatio-temporal variation of NPS pollutant loads are presented in Fig. 4, 5, 6 and 7.

4.4.1 TSS

TSS pollutants are derived from litters from agricultural and forested land uses, oil and grease from urbanized land use during storm runoff. Finer particles present high potential to block drainage system. These pollutants, therefore, need to be eliminated from time to time, if possible after each rain storm before the next to avoid NPS pollution build up (DID 2012). Since TSS loads were derived during storm events by involving multifaceted process such as build up as well as washing away from impervious surfaces, transfer in the sewers, sedimentation and re-suspension of sediments among others, etc. (Rossi et al 2005). Therefore, EMC values used for estimating TSS loads did not give accurate estimate, but rather an approximation of the loads in the area. Figure 4 shows the spatio-temporal variation of TSS NPS pollutant loads from 1984 to 2013 in Kelantan river basin. TSS was increased with changing LULC condition from 1984 to 2013 across the watershed. The total TSS loads in Galas (Fig. 4) increased from 106,959 kg year^{-1} in 1984 to 134,498 in 2013, while in Pergau it increased from 18,933 kg year^{-1} to 289,181 kg year^{-1} during the same period. In both Lebir and Nenggiri, highest TSS pollutant loads were obtained in sub-basins W160 and W200, respectively, probably due to their locations at the outlet, where high runoff may have favored the accumulation of TSS pollutant loads. By comparison,

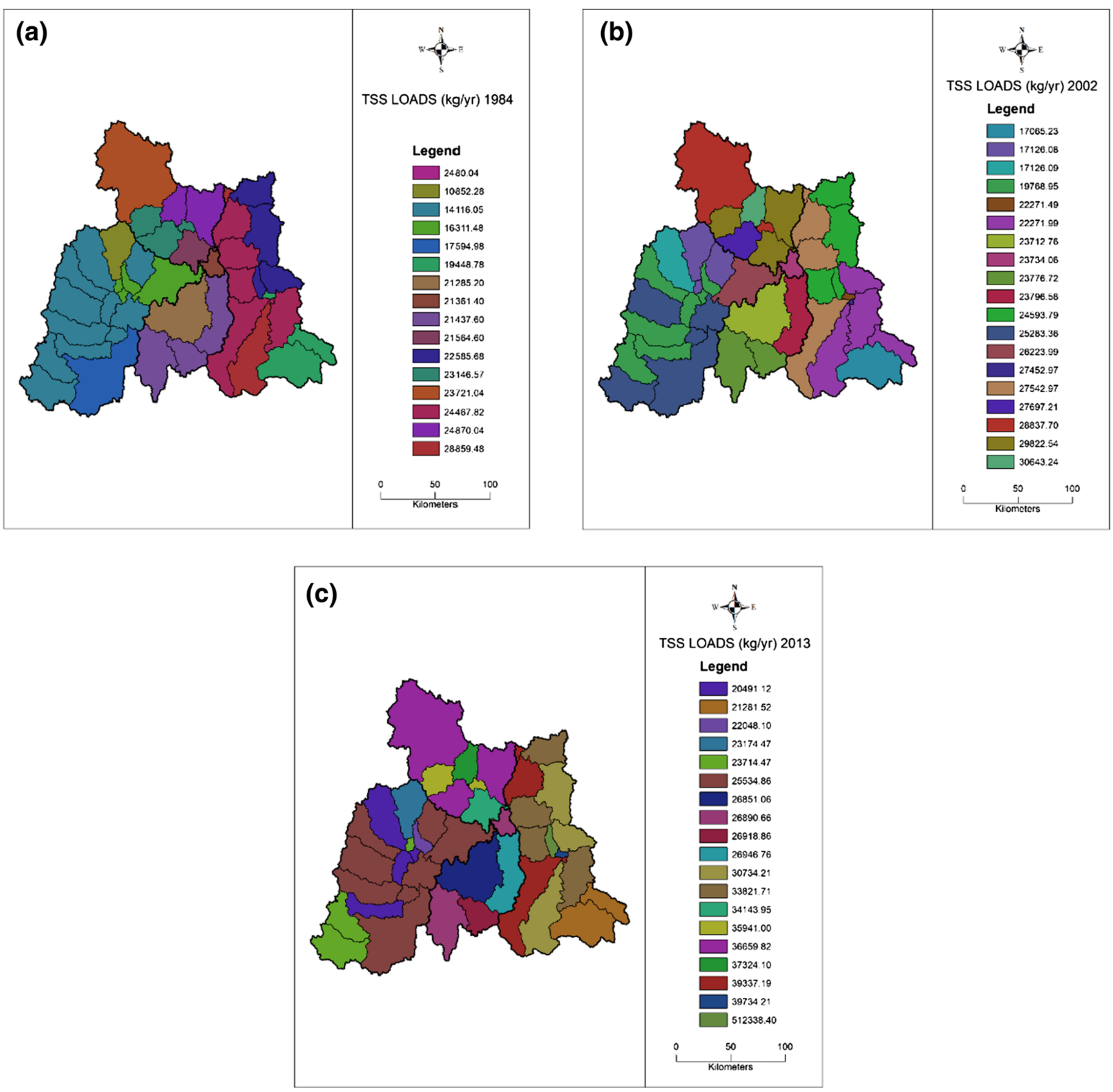

Fig. 4 TSS loads (kg year^{-1}) in Galas, Pergau, Lebir and Nenggiri **a** 1984 **b** 2002 **c** 2013

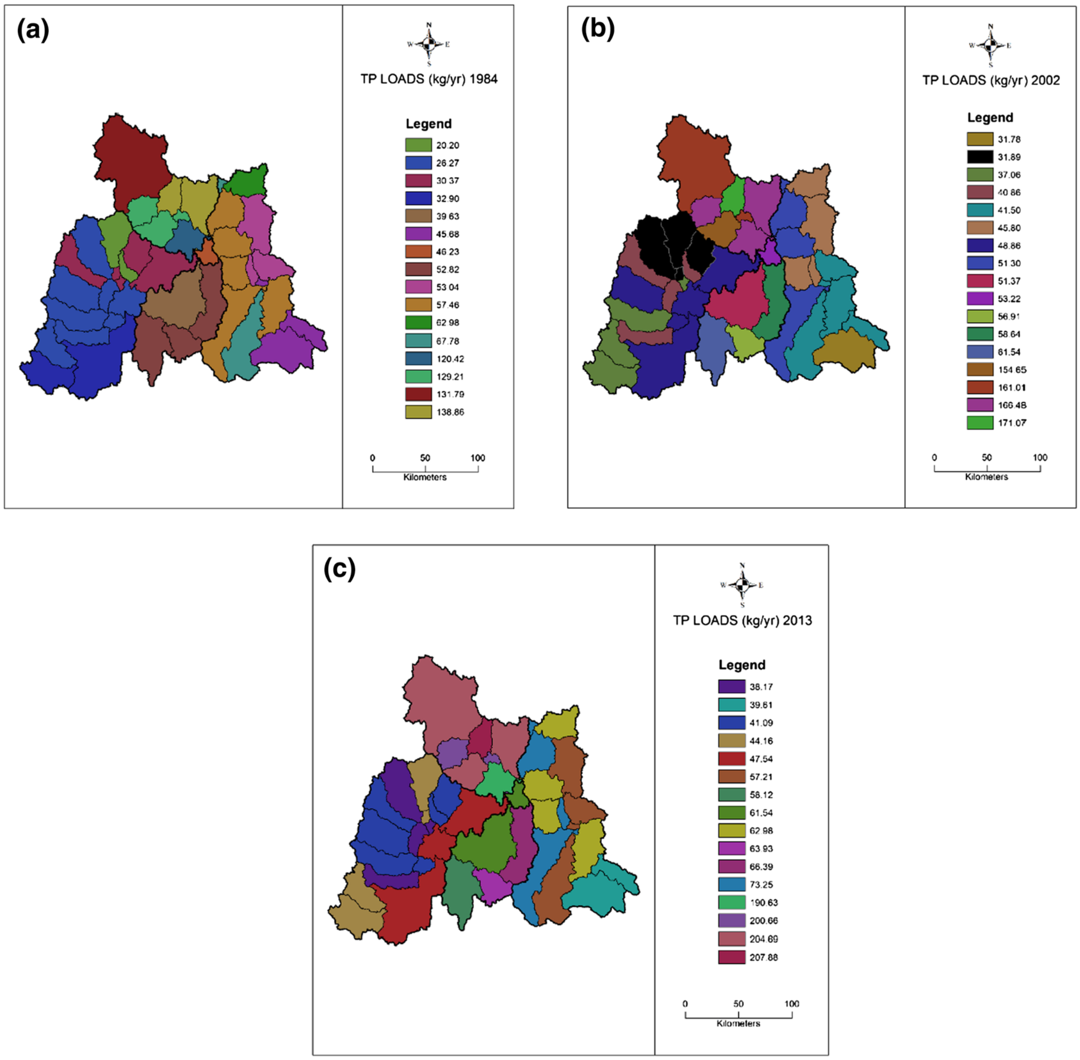

Fig. 5 TP loads (kg year^{-1}) in Galas, Pergau, Lebir and Nenggiri **a** 1984 **b** 2002 **c** 2013

highest total TSS loads change from 1984 to 2013 were obtained in Lebir with 75%, followed by 62% from Nenggiri, 52% in Pergau and 26% in Galas.

4.4.2 TP

The sources of TP are both from agricultural activities and urbanization, which usually binds to soil particles. Figure 5 shows TP NPS pollutant load with its corresponding spatio-temporal variation from 1984 to 2013. The spatio-temporal variation of TP is distributed in accordance to changes in LULC condition. Total TP loads in both Galas and Pergau (Fig. 5) were increased by about 24% from 1984 to 2002 and by about 23% from 2002 to 2013. In Lebir, total TP loads ranged from 679 to 926 kg year^{-1} from 1984 to 2013, whereas in Nenggiri it ranged from 468 to 718 kg year^{-1}. Although the highest TP was not obtained from 1984 LULC condition as obtained in other catchments, but rather under 2002 LULC condition. This may be attributed to higher pervious surfaces obtained under 2002 LULC condition as compared to that of 1984 LULC condition (Fig. S4). In all the catchments, except for Galas, highest total TP loads were recorded in sub-basins closest to the outlets. It could be affirmed that runoff at the outlet resulting from all the sub-basins leads to the accumulation of high TP loads in these catchments.

4.4.3 TN

The term TN encompasses AN, nitrate nitrogen, nitrite-nitrogen as well as organically bonded nitrogen. Agriculture and sewage have been identified as some of the most important sources of TN load in water. Tables 1, 2, 3 and 4 and Fig. 6 shows the trend of TN load in Kelantan river basin.

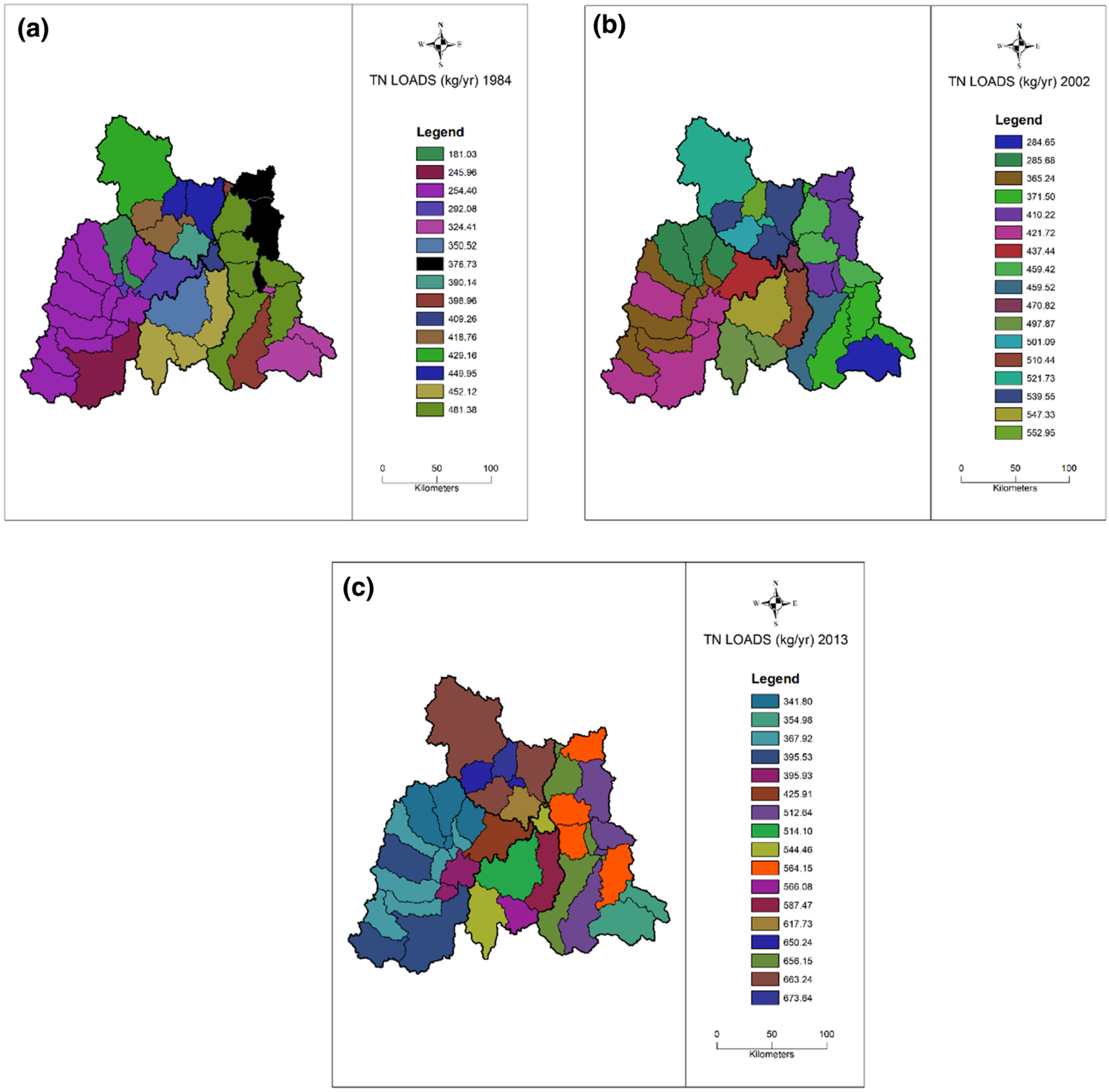

Fig. 6 TN loads (kg year^{-1}) in Galas, Pergau, Lebir and Nenggiri **a** 1984 **b** 2002 **c** 2013

Total TN load in Galas ranged from 2116 to 2757 kg year^{-1} and 3425–5232 kg year^{-1} from 1984 to 2013 (Fig. 6). Highest TN in Galas under 1984 LULC condition was found in sub-basin W80, W90 and W100 (452 kg year^{-1}), whereas under 2002 and 2013 LULC condition, W80 dominated with 510 and 587 kg year^{-1}, respectively. This dominance of sub-basin W80 over the others may be because of rapid deforestation in that area (from 1984 to 2002 and from 2002 to 2013) that may have resulted in increase in runoff, which in turns leads to TN loads accumulation. TN in Lebir increased by about 36% from 1984 to 2013 corresponding to increase in agriculture by about 67% (Fig. 6). Whereas, in Nenggiri, TN load was estimated at about 48% corresponding to over 300% increase in agricultural activities in the area (Fig. 6). Although, the percentage increase in agriculture in Nenggiri was much higher than Lebir, higher total TN loads are recorded in Lebir due to higher runoff activities when compared to Nenggiri.

4.4.4 AN

AN load was increased with changes in LULC condition where higher loads were observed during 1984–2013 (Fig. 7). In Galas and Pergau (Fig. 7c), 2013 LULC condition recorded the highest load of 89.89 and 174 kg year^{-1}, respectively. Although Galas has the higher percentage of agriculture (which is the major source of NPS in the area) compared to Pergau, higher runoff activities were recorded in Pergau due to the presence of more forested areas in Galas. In Lebir (Fig. 7), about 2% increase in AN was

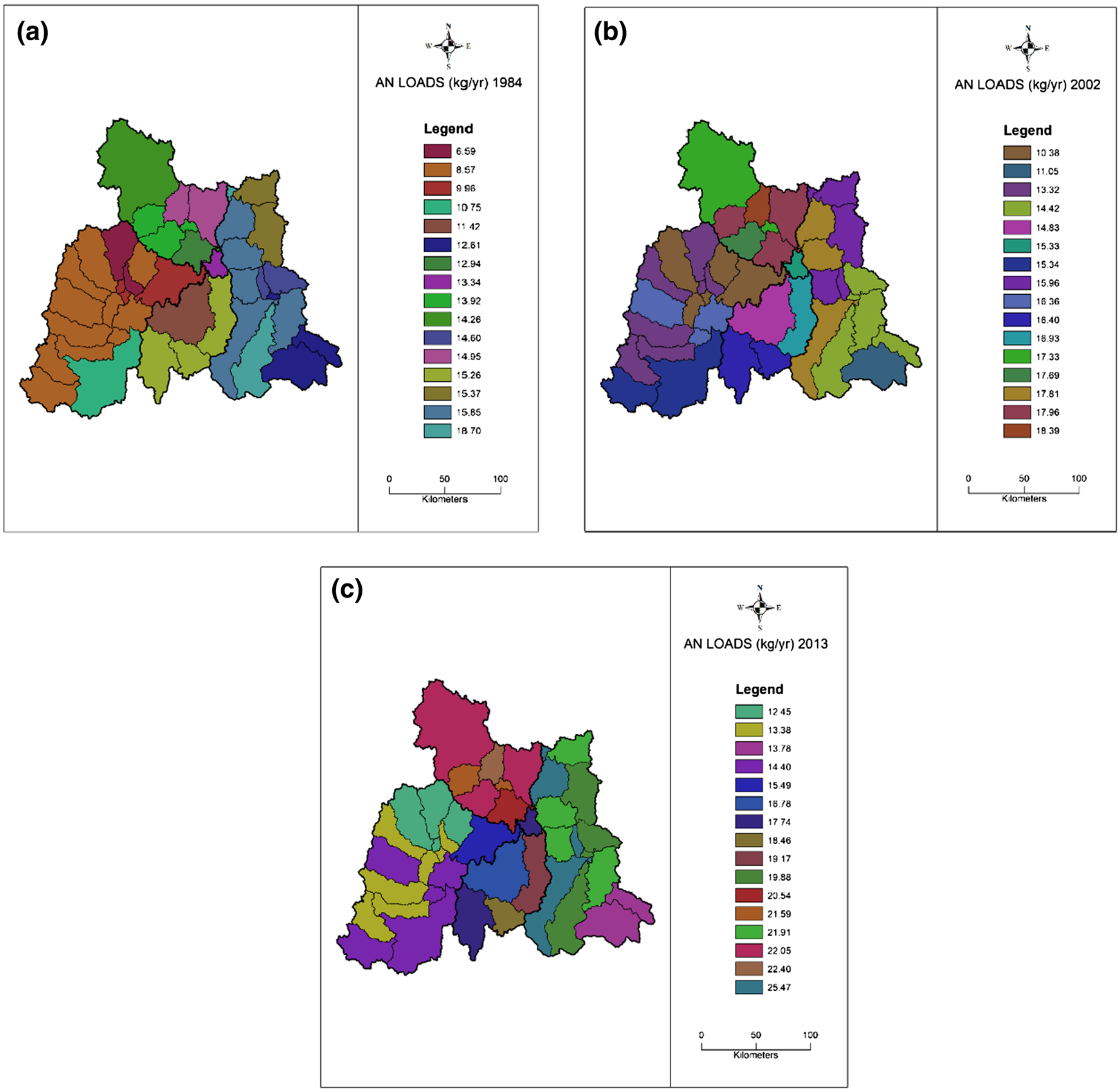

Fig. 7 AN loads (kg year^{-1}) in Galas, Pergau, Lebir and Nenggiri **a** 1984 **b** 2002 **c** 2013

Table 1 NPS pollutant load (kg year^{-1}) in Galas according to land use changes from 1984 to 2013

Land use type	TSS			TP			TN			AN		
	1984	2002	2013	1984	2002	2013	1984	2002	2013	1984	2002	2013
Forest	2819	3197	3595	14.10	15.97	17.97	93.98	107	120	4.71	5.32	5.99
Paddy	15,240	16,917	19,156	42.28	47.95	53.91	376	426	479	4.71	5.32	5.99
Agriculture	15,240	16,917	19,156	42.28	47.95	53.91	376	426	479	4.71	5.32	5.99
Grassland	15,240	16,917	19,156	42.28	47.95	53.91	376	426	479	4.71	5.32	5.99
Urbanization	43,180	47,931	54,276	61.10	69.28	77.91	518	639	719	46.99	53.29	59.94
Cleared land	15,240	16,917	19,156	42.28	47.95	53.91	376	426	479	4.71	5.32	5.99
Total	106,959	118,796	134,495	244.	277	311	2116	2450	2755	70.54	79.89	89.89

observed between 1984 and 2002 and 37% was observed from 2002 to 2013. Whereas, in Nenggiri (Fig. 7), huge difference was recorded from both 1984–2002 and 2002–2013 LULC (52 and 56%, respectively). This large difference may be attributed to inconsistency in the runoff volume in the area.

Table 2 NPS pollutant loads (kg year^{-1}) in Pergau according to land use changes from 1984 to 2013

Land use type	TSS			TP			TN			AN		
	1984	2002	2013	1984	2002	2013	1984	2002	2013	1984	2002	2013
Forest	4014	4968	6131	20.07	24.85	30.67	134	166	204	6.70	8.30	10.24
Paddy	20,071	24,840	30,655	60.21	74.50	91.97	535	662	817	6.70	8.30	10.24
Agriculture	20,071	24,840	30,655	60.21	74.50	91.97	535	662	817	6.70	8.30	10.24
Grassland	20,071	24,840	30,655	60.21	74.50	91.97	535	662	817	6.70	8.30	10.24
Urbanization	52,677	70,379	86,856	86.96	107.65	132.83	803	994	1226	66.91	83.85	102
Cleared land	24,262	24,840	30,655	60.21	74.50	91.97	535	662	817	6.70	8.30	10.24
Mangrove swamp	4014	4968	6131	20.07	24.85	30.67	134	166	204	6.70	8.30	10.24
Secondary forest	4014	4968	6131	20.07	24.85	30.67	134	166	204	6.70	8.30	10.24
Mining	40,142	49,679	61,310	–	–	–	80.28	99.37	123	–	–	–
Total	189,336	234,322	289,179	388	480	593	3425	4239	5229	114	142	174

Table 3 NPS pollutant loads (kg year^{-1}) in Lebir according to land use changes from 1984 to 2013

Land use type	TSS			TP			TN			AN		
	1984	2002	2013	1984	2002	2013	1984	2002	2013	1984	2002	2013
Forest	8580	8855	12,084	42.88	44.28	60.42	298	295	403	14.29	14.63	20.11
Agriculture	42,901	44,273	60,422	186	133	181	1191	1181	1611	14.29	14.63	20.11
Grassland	42,901	44,273	60,422	186	133	181	1191	1181	1611	14.29	14.63	20.11
Urbanization	121,552	125,440	171,195	186	192	262	1768	1771	2417	144	146	201
Cleared land	42,901	44,273	533,423	186	133	181	1191	1181	1611	14.29	14.63	20.11
Secondary forest	8580	8855	12084	42.88	44.28	60.42	298	295	403	14.29	14.63	20.11
Mining	85,801	88,546	1,208,434	–	–	–	179	177	242	–	–	–
Total	353,216	364,515	2,058,064	830	680	926	6116	6081	8298	215	219	301

Table 4 NPS pollutant loads (kg year^{-1}) in Nenggiri according to land use changes from 1984 to 2013

Land use type	TSS			TP			TN			AN		
	1984	2002	2013	1984	2002	2013	1984	2002	2013	1984	2002	2013
Forest	6017	9095	9793	30.50	44.75	46.86	197	303	308	9.85	14.78	15.38
Agriculture	30,247	45,476	48,964	91.60	134	141	789	1213	1232	9.85	14.78	15.38
Grassland	1714	3071	2880	6.44	7.25	8.64	45.72	81.89	76.80	0.57	1.02	0.96
Urbanization	85,700	128,846	138,739	132	193	203	1433	1819	1848	99	148	154
Cleared land	30,247	30,583	48,964	91.60	134	141	740	1213	1232	9.85	14.78	15.38
Secondary forest	6017	9095	9793	30.50	44.75	46.86	197	303	308	9.85	14.78	15.38
Mining	60,493	90,953	97,933	–	–	–	137	182	185	–	–	–
Total	220,435	317,119	357,066	383	558	587	3539	5115	5190	139	208	216

4.5 NPS Pollutant Load Variation Among Different LULC Changes

The results of NPS Loads according to LULC changes are presented in Tables 1, 2, 3 and 4. LULC types in Kelantan river basin were classified into forest, paddy, agriculture, grassland, urbanization, cleared land, mangrove swamp, secondary forest, rivers, ponds and lakes and mining for ease of analysis as well as for increase in precision of the results. NPS pollutant loads among different LULC changes also increases with changes in LULC condition. In Galas (Table 1), estimated TSS load under 1984 LULC condition outweigh that of 2002 LULC condition with total values of 106,959.40 and 118,796.80 kg year^{-1}, respectively. In addition, TSS loads under 2013 LULC condition was recorded to supersede that of 2002 LULC condition. In Pergau total estimated TSS ranged from 189,335.50 to 289,180.7 kg year^{-1} from 1984 to 2013. In all the catchments, urbanization was recorded to give the highest supply of TSS under all LULC conditions. In Lebir and Nenggiri (Tables 3 and 4), urbanization recorded the highest TSS loads of 121,551.70 and 85,700.35 kg year^{-1} under 1984 LULC condition, while forest recorded the lowest in both catchments (8580.12 and 6016.74 kg year^{-1}). Higher values of TSS recorded in urbanized areas may be due to large runoff events caused by impervious surfaces. The relative changes of NPS in Kelantan river basin are not only governed by the type of LULC and nature of the pollutant load but also by the amount of annual rainfall high enough to cause runoff. This is why changes in pollutants loads from 1984 to 2002 and from 2002 to 2013 were not as notable as the LULC changes. For instance, from 1984 to 2002 where massive deforestation of 45% was observed in Galas, these changes were not observed to reflect in the amount of TSS (about 11%) recorded in that year. This may be because only the average rainfall in 1984 was used in computing runoff under 1984 LULC condition. (Fig. S3). Whereas, for 2002 and 2013 LULC conditions, the average values during 1984–2002 and 1984–2013 were used, respectively.

The temporal distribution of TP loads with regards to LULC changes was also found to be regular with changes in LULC condition (Tables 1, 2, 3 and 4). Urbanization was recorded as the LULC change with the highest TP load in all the catchments. Even though agriculture is expected to have a significant contribution of TP loads due to addition from fertilizers and animal manure, higher runoff activities are more likely to occur in urbanized land compared to agricultural land. Thus, making TP loads higher in all catchments in this study. TP loads in Galas (Table 1) ranged from 829 to 926 kg year^{-1} from 1984 to 2013 with urbanization recording the highest load and forest recording the lowest load. Higher TP loads were recorded in Lebir more than all the catchments with estimates of 829, 679 and 926 kg year^{-1} for 1984, 2002 and 2013 LULC conditions, respectively.

TN and AN (Tables 1, 2, 3 and 4) have increased with changes in LULC condition as seen in other pollutants. Similarly, TP urbanization was found to be the LULC change with the highest contribution to both TN and AN load. Even though, agriculture has been reported as one of the major contributors of TN as well as AN load in most NPS studies. The reason for this is that, higher runoff activities were recorded from 1984 to 2013 LULC condition in urbanized areas and, therefore, more NPS loads (TN and AN) are likely to be transferred under these land uses compared to others. From 1984 to 2002 an increase in TN load of 15% was recoded in Galas and 24% in Pergau, while from 2002 to 2013 an increase of 13% and 23% for Galas and Pergau, respectively. Total TN values in Lebir ranged from 6114.03 to 8297.87 kg year^{-1} from 1984 to 2013 and from 3539.47 to 5190.74 kg year^{-1} in Nenggiri. Total AN load for the 2013 LULC which recorded the highest compared to other LULC conditions in all the catchments are 89.89 kg year^{-1} for Galas, 174 kg year^{-1} for Pergau, 302 kg year^{-1} for Lebir and 216 kg year^{-1} for Nenggiri.

4.6 NPS Pollutant Load Relationships Among Different LULC Conditions

4.6.1 Galas

Results of the NPS pollutant loads relationships among sub-basins and LULC conditions in Galas and Pergau are shown in Table 5. The comparison was done between sub-basins W60, W70, W80, W90 and W100 and 1984, 2002 and 2013 LULC conditions. In Galas, sub-basin W80 (26947^{a}) under 2013 LULC condition was ranked statistically most significant ($p<0.05$) when compared to other sub-basins and LULC conditions with regards to TSS. While sub-basin W70 (21285^{k}) under 1984 LULC condition was classified as least significant using the same comparison. Other sub-basins such as W80 (21438^{i}), W90 (21,437^{i}) and W100 (21438^{i}) all under 1984 LULC conditions were grouped as statistically the same by the Tukey's range test. The results obtained from TSS, TP, TN and AN all indicated that sub-basin W80 under 2013 LULC as the statistically most significant ($p<0.05$) when compared to other sub-basins and other LULC conditions. The statistically most significant results obtained from TP, TN and AN are 66.39^{a}, 587.58^{a} and 19.06^{a}, respectively. Mean separation using LSD carried out on TP grouped some sub-basins under different LULC conditions are statistically the same, for example, W80 (52.88^{e}), W90 (52.92^{e}), W100 (52.88^{e}) all under 1984 LULC conditions and W70 (53.30^{e}) under 2002 LULC condition. Whereas under AN statistical grouping was done up to 2 and 3 orders, for example, sub-basins W60 (17.55^{ab}) and W90 (17.82^{ab}) all under 2013 LULC condition were ranked as the most significant and at the second most significant. While sub-basin W90 (15.28^{cd}) under 1984 LULC condition and sub-basin W60 (15.38^{cd}) under 2002 LULC condition were ranked with two same orders even though from different LULC conditions. This makes them statistically similar to other pollutant loads with the same letter in that group. Another noticeable example is where sub-basins W90 (16.30^{bcd}) and W100 (16.45^{bcd}) all under 2002 LULC condition were ranked up to three orders. The statistically most significant pollutant loads obtained from 2013 LULC conditions and that of statistically least significant obtained under 1984 LULC condition is a clear indication of the effect LULC changes on NPS pollution.

4.6.2 Pergau

ANOVA was conducted for the comparison of NPS pollutant loads among the three LULC conditions (1984, 2002 and 2013) and eight sub-basins (W300, W310, W320,

Table 5 NPS pollutant loads relationships among sub-basins and LULC conditions in Galas and Pergau

	Year	Sub-basin				
		W60	W70	W80	W90	W100
Galas						
TSS (kg year^{-1})	1984	21361^{j}	21285^{k}	21438^{i}	21,437^{i}	21438^{i}
	2002	23734^{g}	23713^{h}	23797^{e}	23777^{f}	23777^{f}
	2013	26891^{c}	26851^{d}	26947^{a}	26891^{c}	26919^{b}
TP (kg year^{-1})	1984	46.17^{f}	39.50^{g}	52.88^{e}	52.92^{e}	52.88^{e}
	2002	53.30^{e}	51.66^{e}	58.53^{d}	56.83^{d}	56.93^{d}
	2013	61.58^{c}	58.19^{d}	66.39^{a}	61.57^{c}	63.95^{b}
TN (kg year^{-1})	1984	409.12^{j}	350.43^{k}	452.21^{i}	452.19^{i}	451.99^{i}
	2002	470.73^{g}	457.21^{h}	510.39^{e}	497.92^{f}	497.89^{f}
	2013	544.62^{c}	514.04^{d}	587.58^{a}	544.14^{c}	566.07^{b}
AN (kg year^{-1})	1984	13.40^{e}	14.78^{de}	15.43^{cd}	15.28^{cd}	15.01^{cde}
	2002	15.38^{cd}	14.78^{d}	16.64^{bc}	16.30^{bcd}	16.45^{bcd}
	2013	17.55^{ab}	16.64^{bc}	19.06^{a}	17.82^{ab}	18.67^{a}

	Year	Sub-basin							
		W300	W310	W320	W330	W340	W350	W360	W390
Pergau									
TSS (kg year^{-1})	1984	24,871^{j}	24,871^{i}	23,147^{k}	24,871^{i}	23,147^{k}	23,147^{k}	21,565^{l}	24,871^{i}
	2002	28,838^{g}	29,823^{f}	29,823^{f}	30,644^{e}	28,838^{g}	27,697^{h}	29,823^{f}	28,838^{g}
	2013	36,660^{b}	36,660^{b}	35,942^{c}	37,235^{a}	35,942^{c}	36,660^{b}	34,144^{d}	35,942^{c}
TP (kg year^{-1})	1984	132.08^{j}	139.42^{i}	129.55^{k}	139.67^{i}	129.56^{k}	129.62^{k}	120.78^{l}	139.50^{i}
	2002	161.82^{g}	167.16^{f}	166.98^{f}	171.48^{e}	161.63^{g}	154.78^{h}	167.22^{f}	161.05^{g}
	2013	205.56^{b}	205.53^{b}	201.15^{c}	208.69^{a}	200.81^{c}	204.94^{b}	191.09^{d}	201.47^{c}
TN (kg year^{-1})	1984	429.67^{j}	450.54^{i}	419.01^{k}	450.57^{i}	419.06^{k}	419.44^{k}	390.70^{l}	450.44^{i}
	2002	522.35^{g}	539.86^{f}	540.26^{f}	553.21^{e}	522.48^{g}	501.46^{h}	539.63^{f}	522.35^{g}
	2013	663.93^{b}	663.43^{b}	650.76^{c}	674.42^{a}	650.32^{c}	663.32^{b}	618.23^{d}	650.64^{c}
AN (kg year^{-1})	1984	14.58^{de}	15.30^{d}	14.87^{de}	15.34^{d}	14.11^{de}	14.13^{de}	13.43^{e}	15.10^{de}
	2002	17.77^{c}	18.64^{c}	18.63^{c}	18.85^{c}	18.07^{c}	18.21^{c}	18.91^{c}	18.16^{c}
	2013	22.31^{ab}	22.40^{ab}	22.19^{ab}	23.16^{a}	22.01^{ab}	22.38^{ab}	20.70^{b}	22.03^{ab}

Least significant means with the same letter are not significantly different at $p < 0.05$

W330, W340, W350, W360 and W390) in Pergau and are shown in Table 5. For all the pollutant loads in this catchment, sub-basin W330 under 2013 LULC condition and sub-basin W360 were ranked as the statistically most significant and least significant ($p < 0.05$) when compared with other sub-basins and other LULC conditions. Pollutants loads that were ranked statistically most significant are 37235^{a} for TSS, 208.69^{a} for TP, 674.42^{a} for TN and 23.16^{a} for AN. While pollutant loads that were grouped as statistically least significant ($p < 0.05$) for TSS, TP, TN and AN are 21565^{l}, 120.78^{l}, 390.70^{l} and 13.43^{e}, respectively. For both TSS, TP and AN, the mean separation carried out using LSD grouped the pollutant loads at just one order. While for AN, grouping was done for up to two orders, for example, most of the sub-basins under 1984 and 2013 LULC conditions were ranked for up to two orders except for W310 (15.30^{d}), W330 (15.34^{d}) and W360 (13.43^{e}) under 1984 LULC condition. While under 2013 LULC condition W330 (23.16^{a}) and W360 (20.70^{b}) were grouped with one order while the rest were grouped with two orders. The statistically most significant ($p < 0.05$) pollutant loads obtained under 2013 LULC condition and statistically least significant ($p < 0.05$) pollutant loads obtained under 1984 LULC condition clearly indicated the influence of LULC change on NPS pollution. The 1984 LULC condition that is characterized with low runoff due to high percentage of forested areas will result to build up of low NPS pollutant loads. While the reverse is the case for the 2013 LULC condition which has undergone deforestation mostly agricultural activities, illegal logging and urbanization will result to high NPS pollution due to high rate of runoff.

4.6.3 Lebir

Results of NPS pollutant loads relationships among LULC conditions and sub-basins in Lebir are shown in Table 6. The comparison involved three LULC conditions (1984, 2002 and 2013) and fifteen sub-basins. In all the pollutant loads, 2013 LULC conditions was found to be statistically the most significant ($p<0.05$) when compared to other sub-basins and other LULC conditions. Although the sub-basins involved are not the same for all the pollutants, this could be due to LULC changes from one sub-basin to another that may have altered runoff which in turn influences pollutant load. Under TSS sub-basin W220 (512339^{a}) was statistically the most significant ($p<0.05$), while for both TP, TN and AN sub-basins W160, W180, W220, W250 and W260 were grouped as statistically most significant ($p<0.05$) compared to other sub-basins and LULC conditions. In the case of statistically least significance ($p<0.05$), sub-basin W240 under 1984 LULC conditions was recorded for both TSS (19450^{l}), TP (46.31^{h}), TN (324.91^{k}) and AN (13.03^{qr}). Ranking of means with regards to statistical significance was done using one order for both TSS and TN where means with the same letters were grouped as statistically the same. While for TP ranking of means was done for up two orders, for example; sub-basin W190 (53.53^{ef}) under 1984 LULC condition and for three orders; sub-basin W160 (51.90^{efg}) under 2013 LULC condition. A more complex ranking was obtained from AN pollutant where ranking was done for up to four orders; sub-basin W160 (19.13^{efgh}) and up to five orders; sub-basin W180 (16.48^{ijklm}) all under 1984 LULC condition. This complex ranking is due to statistical similarities involving the pollutant loads where any two or more means carrying the same letters are grouped as statistically the same. From the result, it could be observed that, 2013 LULC condition was the statistically most significant ($p<0.05$) compared to 2002 and 1984 LULC condition. Thus, indicating the influence of LULC changes on NPS pollution in the watershed.

4.6.4 Nenggiri

NPS pollutant loads relationships were compared among three LULC conditions and different sub-basins in Nenggiri and the results are presented in Table 7. The results indicated for TSS pollutants sub-basin W200 (26225^{a}) under 2002 LULC condition was ranked as the statistically most significant ($p<0.05$) compared to other sub-basins and LULC conditions. While for TP, sub-basins W200 (49.30^{a}), W280 (49.20^{a}), W290 (49.42^{a}) and W320 (49.65^{a}) all under 2002 LULC condition were grouped as the statistically most significant ($p<0.05$) sub-basins when compared to other sub-basins and LULC conditions. For TN, sub-basins W200 (438.05^{a}) and W220

Table 6 NPS pollutant loads relationships among LULC conditions and sub-basins in Lebir

	Year	Sub-basin														
		W160	W170	W180	W190	W200	W210	W220	W230	W240	W250	W260	W270	W280	W290	W300
TSS	1984	28,860^{e}	22,586^{i}	24,468^{h}	22,586^{i}	24,468^{h}	24,468^{h}	22,586^{i}	22,586^{i}	19,450^{l}	24,468^{h}	24,468^{h}	28,860^{e}	19,449^{l}	24,469^{h}	19,449^{l}
(kg year^{-1})	2002	27,543^{f}	24,594^{g}	27,544^{f}	24,594^{g}	27,543^{f}	24,594^{g}	24,594^{g}	22,273^{j}	22,273^{j}	27,543^{f}	27,544^{f}	22,273^{j}	22,273^{j}	22,273^{j}	17,066^{m}
	2013	39,338^{b}	33,822^{c}	39,338^{b}	30,735^{d}	33,822^{c}	33,822^{c}	5,12,339^{a}	30,735^{d}	30,735^{d}	39,338^{b}	39,338^{b}	30,735^{d}	21,282^{k}	33,822^{c}	2128^{k}
TP (kg year^{-1})	1984	68.04^{b}	53.40^{efg}	57.73^{d}	53.53^{ef}	58.26^{d}	57.97^{d}	53.76^{e}	53.36^{efg}	46.31^{h}	57.60^{d}	58.00^{d}	68.08^{b}	46.50^{h}	58.00^{d}	46.36^{h}
	2002	51.90^{efg}	46.43^{h}	51.54^{g}	46.46^{h}	51.55^{g}	46.12^{h}	46.16^{h}	41.96^{ij}	42.29^{j}	51.91^{efg}	51.82^{fg}	41.76^{ij}	41.90^{ij}	42.13^{ij}	32.17^{k}
	2013	73.76^{a}	63.58^{c}	73.76^{a}	57.74^{d}	63.45^{c}	63.29^{c}	73.73^{a}	57.87^{d}	57.91^{d}	73.47^{a}	73.84^{a}	57.63^{d}	40.31^{j}	63.27^{c}	40.34^{j}
TN (kg year^{-1})	1984	399.41^{g}	377.20^{h}	481.71^{d}	377.19^{h}	481.87^{d}	481.65^{d}	376.96^{h}	481.91^{d}	324.91^{k}	481.68^{d}	324.95^{k}	399.39^{g}	325.26^{k}	481.84^{d}	324.94^{k}
	2002	371.88^{i}	410.57^{f}	459.91^{e}	410.91^{f}	459.75^{e}	410.65^{f}	410.77^{f}	460.05^{e}	372.20^{i}	459.71^{e}	459.91^{e}	372.30^{i}	372.07^{f}	371.92^{i}	285.28^{l}
	2013	656.63^{a}	564.36^{b}	656.51^{a}	512.88^{c}	565.03^{b}	564.70^{b}	656.71^{a}	512.85^{c}	512.96^{c}	656.48^{a}	656.76^{a}	513.16^{c}	355.65^{j}	564.47^{b}	355.44^{j}
AN (kg year^{-1})	1984	19.13^{efgh}	15.87^{mno}	16.48^{ijklm}	16.11^{mno}	16.37^{jklmn}	16.17^{mno}	15.36^{mno}	15.04^{mnop}	13.03^{qr}	16.53^{ijklm}	16.28^{klmno}	18.91^{efgh}	13.17^{qpr}	16.48^{ijklm}	13.03^{qr}
	2002	16.58^{ijklm}	16.16^{mno}	18.21^{hijkl}	16.51^{ijklm}	18.31^{ghij}	16.26^{klmno}	16.23^{lmno}	15.17^{mnop}	15.05^{mnop}	18.44^{fghi}	18.23^{ghijk}	14.91^{mnop}	15.08^{mnop}	14.85^{mnopq}	11.68^{r}
	2013	25.76^{a}	22.51^{b}	26.22^{a}	20.22^{defg}	22.47^{b}	22.09^{bcd}	25.89^{a}	20.60^{bcde}	20.38^{cdef}	25.90^{a}	26.11^{a}	20.46^{cde}	14.44^{nopq}	22.34^{bc}	14.34^{opq}

Least significant means with the same letter are not significantly different at $p<0.05$

Table 7 NPS pollutant loads relationships among LULC conditions and sub-basins in Nenggiri

	Year	Sub-basin								
		W180	W190	W200	W210	W220	W230	W240	W250	W260
TSS (kg year^{-1})	1984	10,853^{l}	14,117^{k}	16,313^{j}	14,117^{k}	14,117^{k}	16,313^{j}	16,313^{j}	16,313^{j}	14,117^{k}
	2002	17,127^{i}	17,127^{i}	26,225^{a}	17,127^{i}	19,770^{g}	19,770 ^{g}	17,127^{i}	19,770^{g}	19,770^{g}
	2013	23,715^{d}	20,492^{f}	25,536^{b}	25,535^{b}	25,535^{b}	22,049^{e}	23,715^{d}	20,492^{f}	20,491^{f}
TP (kg year^{-1})	1984	20.20^{i}	26.78^{h}	31.18^{fg}	31.07^{g}	31.20^{efg}	20.97^{i}	30.91^{g}	31.10^{g}	27.21^{h}
	2002	32.61^{efg}	32.59^{efg}	49.30^{a}	32.49^{efg}	41.82^{c}	41.64^{c}	32.77^{efg}	37.57^{d}	41.37^{c}
	2013	44.95^{b}	38.85^{d}	48.28^{a}	42.02^{c}	41.74^{c}	41.60^{c}	44.53^{b}	38.90^{d}	38.75^{d}
TN (kg year^{-1})	1984	181.82^{m}	255.18^{k}	292.82^{i}	255.37	254.93 ^{k}	181.61^{m}	293.07^{i}	292.81^{i}	255.23^{k}
	2002	285.95^{j}	286.11^{j}	438.05^{a}	286.01^{j}	438.05^{a}	366.42^{fg}	286.12^{j}	365.85^{g}	365.63^{g}
	2013	342.42^{h}	342.04^{h}	426.70^{b}	342.37^{h}	368.51^{ef}	368.90^{e}	342.34^{h}	368.84^{e}	368.61^{e}
AN (kg year^{-1})	1984	7.02^{n}	9.30^{jklm}	10.40^{jkl}	9.14^{klmn}	9.18^{jklmn}	7.28^{nm}	10.88^{ijkl}	10.59^{jkl}	9.52^{jklm}
	2002	14.10^{cdefg}	10.66^{jkl}	11.12^{hijkl}	11.27^{hijk}	14.20^{cdefg}	13.87^{efg}	10.68^{lkj}	13.74^{gf}	11.00^{hijkl}
	2013	13.25^{fgh}	13.19^{fgh}	16.09^{abcde}	13.16^{ghf}	14.09^{cdefg}	14.15^{cdefg}	13.01^{ghi}	14.22^{cdefg}	13.91^{efg}

	Year	Sub-basin							
		W270	W280	W290	W300	W310	W320	W330	W340
TSS (kg year^{-1})	1984	14,117^{k}	14,117^{k}	14,117^{k}	14,117^{k}	14,117^{k}	17,596^{h}	14,117^{k}	14,117^{k}
	2002	25,284^{c}	25284^{c}	25,284^{c}	19,769^{g}	19,770^{g}	25,284^{c}	19,770^{g}	25,284^{c}
	2013	25,535^{b}	25,536^{b}	25,536^{b}	25,535^{b}	20,492^{f}	25,536^{b}	23,715^{d}	23,715^{d}
TP (kg year^{-1})	1984	27.16^{h}	27.21^{h}	27.03^{h}	27.07^{h}	26.90^{h}	33.48^{e}	26.79^{h}	33.40^{ef}
	2002	49.79^{a}	49.20^{a}	49.42^{a}	37.56^{d}	41.69^{c}	49.65^{a}	37.85^{d}	37.84^{d}
	2013	48.34^{a}	41.39^{c}	41.60^{c}	41.67^{c}	38.40^{d}	47.94^{a}	45.09^{b}	44.97^{b}
TN (kg year^{-1})	1984	255.33^{k}	254.74^{k}	255.18^{k}	254.90^{k}	255.10^{k}	246.95 ^{l}	254.72^{k}	254.82^{k}
	2002	422.14^{c}	422.23^{c}	422.37^{c}	365.71^{g}	366.05^{g}	422.09^{c}	366.04^{g}	422.43^{c}
	2013	396.48^{d}	395.85^{d}	396.21^{d}	368.72^{e}	368.25^{ef}	396.32^{d}	368.71^{e}	396.06^{d}
AN (kg year^{-1})	1984	9.60^{jkl}	9.22^{jklmn}	9.57^{jkl}	9.27^{jklmn}	9.13^{klmn}	11.45^{hij}	8.98^{lmn}	9.11^{klmn}
	2002	16.92^{ab}	17.21^{a}	16.99^{ab}	13.74^{gf}	14.03^{cdefg}	16.21^{abcd}	13.96^{defg}	16.28^{abc}
	2013	14.85^{bcdefg}	15.17^{abcdefg}	15.23^{abcdefg}	13.79^{gf}	14.19^{cdefg}	15.29^{abcdef}	14.02^{cdefg}	14.98^{abcdefg}

Least significant means with the same letter are not significantly different at $p < 0.05$

(438.05^{a}) all under 2002 LULC condition were ranked as the statistically most significant ($p < 0.05$) compared to other sub-basins and LULC conditions. For AN loads, sub-basins W280 (17.21^{a}) was found to be the statistically most significant ($p < 0.05$) compared to other sub-basins and LULC conditions. Although, other sub-basins such as, W270 (16.92^{ab}) and W290 (16.99^{ab}) under 2002 LULC condition and sub-basins W280 (15.17^{abcdefg}) and W290 (15.23^{abcdefg}) under 2013 LULC condition are grouped as statistically similar to W280, because they carry the same letter in their ranking. The ranking is therefore not limited to W280 alone, but to all sub-basins with similar letters used for ranking in all the sub-basins and across the LULC conditions. For all the pollutant loads in Nenggiri, sub-basin W180 under 1984 LULC condition was grouped as the statistically least significant ($p < 0.05$) when compared with other sub-basins and LULC conditions. Mean values of pollutant loads for sub-basin W180 are as follows; TSS (10853 ^{l}), TP (20.20^{i}), TN (181.82^{m}) and AN (7.02^{n}). Mean separation carried out in this catchment ranked some pollutants up to just one order such as what is obtained in TSS and TN. While a more complex way of ranking was observe for AN where means were grouped up to 7 orders, for example; W280 (15.17^{abcdefg}) and W290 (15.23^{abcdefg}) all under 2013 LULC condition. This complex way of ranking reveals the statistical similarities existing among the means carrying the same letters. It can be inferred from the result that NPS pollution in Nenggiri is influenced by long-term LULC changes. Although, 2002 LULC condition and not 2013 LULC condition was reported to observe the statistically most significant ($p < 0.05$) sub-basin unlike in other catchments. High percentage of forested areas were observed under 2013 LULC condition compared to 2002 LULC condition. Therefore, 2002 LULC condition will likely to favor large runoff which in turns carries along with it NPS pollutants as compare to 2013 LULC condition.

5 Conclusion

With the aid of GIS tool, numeric integration was carried out to estimate spatio-temporal changes on NPS in Kelantan river basin from 1984 to 2013. The following conclusions were drawn. All the pollutant loads estimated in this study were observed to be affected by LULC changes, runoff and EMC values. The extensive LULC change mostly involves deforestation for logging and agricultural purposes. In all the catchments, pollutant loads mostly increase with changes in LULC condition as one moves from 1984 to 2013. NPS pollutant loads among different LULC changes also increase with changes in LULC condition from 1984 to 2013. Urbanization was found to be the dominant LULC change with the highest pollutant load in all the catchments. Higher values of pollutant loads recorded in urbanized areas may be due to large runoff events caused by huge impervious surfaces which when combined with EMC values give figures that are far above those of other LULC changes given rise to high-level TSS in urbanized places.

Analysis of variance (ANOVA) was conducted for the comparison of NPS pollutant loads among the three LULC conditions used and the sub-basins in each catchment in this study for the entire watershed. Mean separation was carried out using least significance difference (LSD) and significant means were grouped using Tukey's range test. The results revealed that 2013 LULC condition was the statistically most significant ($p < 0.05$) LULC condition in Galas, Pergau and Lebir, while in Nenggiri it was found to be the 2002 LULC condition. In all the catchments 1984 LULC condition was found to be the statistically least significant ($p < 0.05$) LULC condition compared to other LULC conditions. The findings of this study will of serious help to water resource planners in controlling water pollution for future planning.

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Compliance with Ethical Standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest. Authors further declare that there is no financial or personal relationship with a third party whose interests could be positively or negatively influenced by this article's content.

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