



Detecting and predicting the impact of land use changes on groundwater quality, a case study in Northern Kelantan, Malaysia



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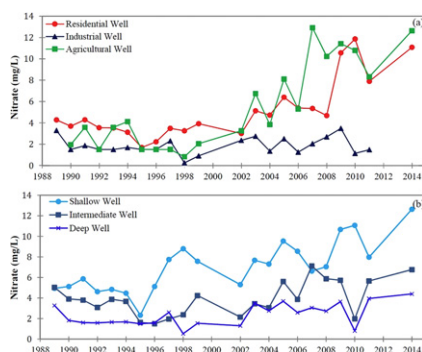
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HIGHLIGHTS

- This study aims to quantify impact of deforestation on groundwater quality.
- Groundwater quality variations detected in Kelantan from 1989 to 2014
- Nitrate increased 8.1% annually in agricultural wells.
- The significant increase in nitrate was only observed in a shallow aquifer.
- ARIMA model predict nitrate will rise 2.64% annually until 2030.

GRAPHICAL ABSTRACT



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ABSTRACT

The conversions of forests and grass land to urban and farmland has exerted significant changes on terrestrial ecosystems. However, quantifying how these changes can affect the quality of water resources is still a challenge for hydrologists. Nitrate concentrations can be applied as an indicator to trace the link between land use changes and groundwater quality due to their solubility and easy transport from their source to the groundwater. In this study, 25 year records (from 1989 to 2014) of nitrate concentrations are applied to show the impact of land use changes on the quality of groundwater in Northern Kelantan, Malaysia, where large scale deforestation in recent decades has occurred. The results from the integration of time series analysis and geospatial modelling revealed that nitrate ($\text{NO}_3\text{-N}$) concentrations significantly increased with approximately 8.1% and 3.89% annually in agricultural and residential wells, respectively, over 25 years. In 1989 only 1% of the total area had a nitrate value greater than 10 mg/L; and this value increased sharply to 48% by 2014. The significant increase in nitrate was only observed in a shallow aquifer with a 3.74% annual nitrate increase. Based on the result of the Autoregressive Integrated Moving Average (ARIMA) model the nitrate contamination is expected to continue to rise by about 2.64% and 3.9% annually until 2030 in agricultural and residential areas. The present study develops techniques for detecting and predicting the impact of land use changes on environmental parameters as an essential step in land and water resource management strategy development.

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1. Introduction

Massive urbanization and industrialization, development of agricultural lands, deforestation, mining and road building as examples of human activities have resulted in global climate change, regional ecological deterioration, and landscape degradation (Deng and Shangguan, 2016; Pérez-Fernández et al., 2016). Land degradation may have a direct impact on soil degradation, which destroys ecosystem functions and effects the hydrological, biological and geochemical cycles (Keesstra et al., 2012; Pérez-Fernández et al., 2016). Land use and land cover patterns are the cumulative results of numerous natural and socio-economic factors in relation to human activities in time and space. Generally, the world's natural resources are utilized in an unsustainable manner due to unprecedented population growth, and industrial and agricultural development (Keeler and Polasky, 2014).

According to the United Nations Food and Agricultural Organization (FAO), an estimated 5.2 million hectares of forests were lost from 2000 to 2010 (FAO, 2010), with shows the highest rate of deforestation in Asia (FAO, 2012). Around 15% of tropical rainforests are in Southeast Asia, which play a significant role in environmental biodiversity, as well as stabilizing the world's climate and protecting the environment against soil erosion, floods, and droughts. The FAO (2010) reported a yearly tropical rainforest loss in Southeast Asia of 2.4 Mha in the 1990s, 0.4 Mha from 2000 to 2005, and 1.0 Mha for the period 2005–2010. The consequences of land use changes not only affect climate change, wildlife habitats, soil carbon storage and soil erosion, but also have a negative effect on water quality by increasing sediments, nutrients, and agricultural chemicals (Zhang et al., 2015).

In 2016, the United Nations adopted the Sustainable Development Goals (SDGs) in which the protection of water resources is an important topic. To be able to do this it is of importance to understand the complex interactions between land use, the soil system and ecosystem services (Keesstra et al., 2016). A good understanding of linkages between surface and sub-surface processes in the nutrient and hydrological cycle will be essential to be able to develop sustainable land and water management strategies.

The conversion of natural forests and grassland to urban and farmland, together with population growth and intensive agricultural activities, is producing a potential source of contaminants from fertilizers, sewage disposal, and landfill, which often influence the hydrological system and change the runoff and water quality (Fletcher et al., 2013). Valle et al. (2014) mentioned that human association to some land uses as a source of groundwater quality changes, directly or indirectly. In recent decades, increasing nitrogen and nitrate concentrations in surface and groundwater have been of great concern due to the drastic agricultural and urban development (Gardner and Vogel, 2005), which revealed a certain relation between the nitrate concentration in the water resources and agricultural activities (Yu et al., 2014).

Previous studies mostly characterized the status of nitrate concentration in the groundwater (García-Díaz, 2011; Kurunc et al., 2016), influence on public health (Manassaram et al., 2007; Powlson et al., 2008), source of nitrate (Wakida and Lerner, 2005; Lawniczak et al., 2016), and transport and vulnerability in aquifers based on the short-term fluctuations (Kurunc et al., 2011; Sheikhy Narany et al., 2013). However, it is difficult to quantify and detect the link between land use changes and human activities, and groundwater quality due to the variability of nitrate concentrations across the landscape (Zhang et al., 2013). Chen et al. (2010) revealed that nitrate concentration in groundwater in areas used for rotating irrigated cultures was higher than areas occupied by forests in Huantai County in the North China Plain. In other research by Schilling and Jacobson (2010) in Neal Smith National Wildlife Refuge, Iowa, USA, nitrate concentration in groundwater decreased by converting cropland to perennial land cover (prairie) within a decade. In similar research by Valle et al. (2014) the impact of land use conflicts on quality of groundwater in the Sordo River basin (northern Portugal), which represented high concentration of nitrate in moderate and major

conflict areas, as result of the nitrification of N-fertilizers. Therefore, the application of historical groundwater nitrate data can be applied to detect the impact of urbanization and agricultural development on water quality (Gardner and Vogel, 2005).

Malaysia, where rapid deforestation and urbanization (around 7%) have been reported from the 1970s to 1990s (Hassan, 2004), were used to reveal the impact of land use changes on groundwater quality. According to the current literature it would be expected that the extensive and rapid land use changes to crops, paddy, palm oil and rubber cultivation, would significantly increase the nitrate concentration of the shallow aquifers (Gardner and Vogel, 2005; Keeler and Polasky, 2014).

Historical environmental observations play an important role for detecting environmental changes over time. Time series analysis is another type of quantitative prediction, which is the record of consecutive observations (Temme et al., 2004) to develop the models describing the underlying relationships (Zhang, 2003). Therefore, the pattern of recorded data should be identified to characterize the nature of the phenomena and to predict future values. The underlying relationships between the past observations of the same variable could be applied to develop a model to extrapolate the time series in the future. Although hydrological systems have highly stochastic nature, suitable models have been developed to describe the complex phenomena. Accurate prediction of the concentration of contamination in ground water is a vital tool to assess the success of land and water resource management. The autoregressive integrated moving average (ARIMA) model is one of the most popular time series prediction models, and is helpful in predicting the future values based on past observations (Zhang et al., 2013).

The main aim of the present study was to detect, predict, and visualize the temporal and spatial variation of nitrate in groundwater due to long term land use changes in Northern Kelantan by integrating time series analyses and geospatial modelling, which give a clear view of the influence of human activities on the environment.

2. Materials and methods

2.1. Study area

The study was carried out in Northern Kelantan in the north-eastern side of Kelantan State, the largest state in Peninsular Malaysia, which lies between latitudes 5° 53' N and 6° 14' N and longitudes 102° 7' E and 102° 28' E (Fig. 1a). The study area covers an area of around 880 km²; mostly consisting of alluvial deltas and coastal plains (Sefie et al., 2015). Since Malaysia is in an equatorial zone, the climate is controlled by the northeast (NE) and southwest (SW) monsoon's regimes. The annual rainfall changes from 1750 mm in the monsoon season from November to January and 0 mm in the dry season from March to May. The average temperature is around 26 °C throughout the year in the study area.

Northern Kelantan is covered by Quaternary alluvium of marine and fluvial origin, which has a large potential for groundwater sources. Mesozoic granitic and sedimentary or metasedimentary bedrock underlay the alluvium layer, which mainly consists of shale, sandstone, phyllite and slate. The thickness of the quaternary alluvium varies from a few meters near the mountains to >150 m near the coast (Nur-Hayati, 2011) (Fig. 1e).

Based on lithological studies, the study area consists of three aquifer layers; shallow aquifer, intermediate aquifer, and deep aquifer. The shallow aquifer is mostly unconfined and semi-confined with varying from 2 to 3 m up to 17.5 m. The intermediate and deep aquifers lie at a depth of around 20 m to 50 m, and >50 m, respectively (Nur-Hayati, 2011). Generally, the shallow aquifers fed by precipitation, feeding rivers. Based on the study of Noor (1979), the shallow and intermediate aquifers are hydraulically interconnected due to the existence of the silty layer between them.

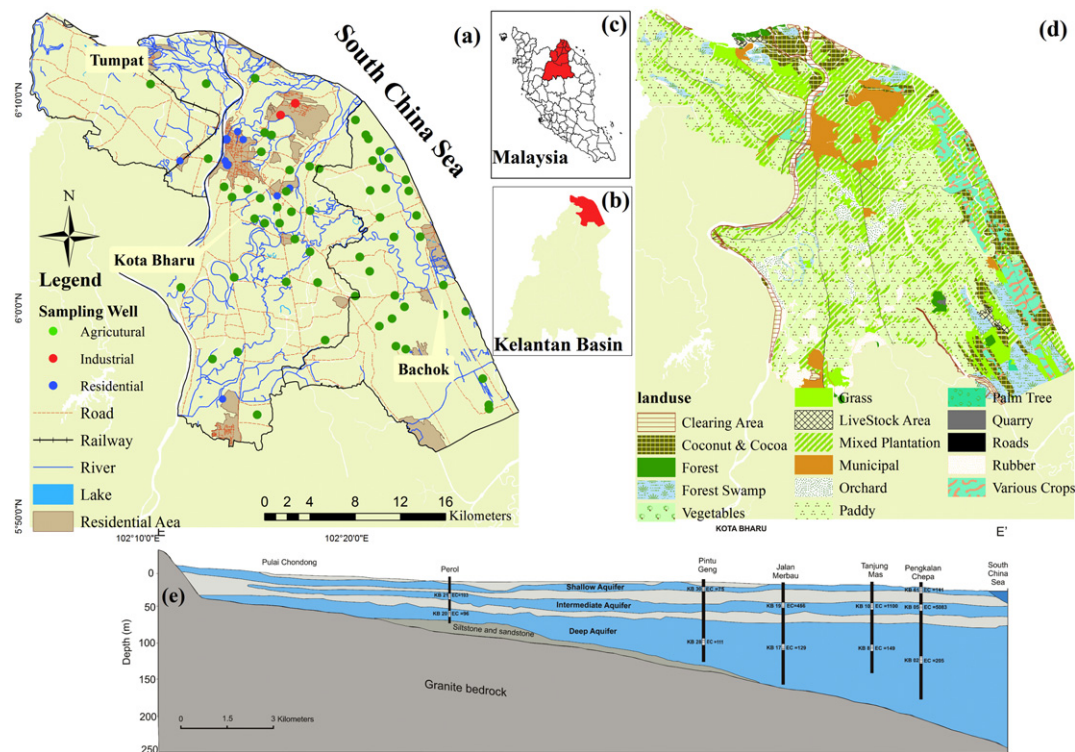


Fig. 1. Location of the Northern Kelantan (a), in Kelantan State (b) and Peninsular Malaysia (c), Land use activities map 2011 (d) and Geological cross section (e).

Groundwater is fully developed for drinking water use since 1935 Kelantan State. Due to the high recharge rate and easy access to the groundwater, the shallow aquifer is very productive in terms of drinking water extraction. In the study area, approximately 70% of potable water is derived from groundwater sources. Also, groundwater is a primary source for farmers to cultivate rice and grow tobacco. Groundwater abstraction was around 36,370 m³/d in 1981, which increased sharply to 115,600 m³/d in 1993. In 2011, the groundwater abstraction was approximately 134,000 m³/d, and is estimated to increase with 2.5% per year from now on, due to intensive agricultural activities and population growth in the residential areas, such as, Kota Bharu, Tumpat, Wakaf, Pengkalan, Chepa, and Bachok.

2.2. Land use

Around 71.8% (around 10,784 km²) of Kelantan State is covered with forest reserves, which are mainly located in the upstream region (Jusoff and Setiawan, 2003). From 1970 to 1990s land use change from forests to farm lands was progressive with 7% growth, but the growth slowed in the 1990s to 1.4%. A total of 481 km² of forest reserve in this state had been encroached upon by 1993 (Jusoff and Setiawan, 2003). Although the economy has improved significantly since the agricultural development at the expense of the forest; deforestation can have a negative influence on environmental stability, wildlife and the ecosystem and increase soil erosion and flood risk in the study area. The current land use in the study area comprises mainly of paddy cultivation, mixed agriculture, palm trees, built up areas, forest and grassland (Fig. 1d). However, the percentage of each land use type in the study area shows changes from 1989 to 2014.

There were 1,174,000 people living in Kelantan State in 1990, with around 80% of the population concentrated in Northern Kelantan (Kumari and Kavanagh, 1990). The built-up areas, mixed agriculture lands, and paddy cultivation covered approximately 38 km², 181 km², and 275 km² (4.5%, 21.1%, and 32.1% of total area), respectively, in the

1980s. In addition, around 214 km² (25% of total area) is covered by forest and grassland. The population of Kelantan State population increased to 1,718,000 people in 2015, which mostly occupies the districts of Kota Bharu, Tumpat, and Bachok. The built-up areas, paddy cultivation lands, mixed agricultural areas, and palm tree lands increased to 48 km² (5.57%), 290 km² (34.0%), 203 km² (23.7%), and 134 km² (15.63%), respectively in 2011 (Nur-Hayati, 2011) (Fig. 1d), while, the forest and grassland areas decreased to 73 km² (8.5% of total area) due to deforestation.

2.3. Groundwater quality sampling

The regional groundwater samples were collected from 1989 to 2011 by the Minerals and Geoscience Department Malaysia (JMG), and from 2009 to 2014 by the National Hydraulic Research Institute of Malaysia (NAHRIM) in Northern Kelantan. During these years, >1500 groundwater samples were collected from 101 sampling wells. However, the groundwater data set has some gaps in 2000, 2001, 2012, and 2013. A total of 101 wells were divided into three main aquifers, shallow aquifer (64 wells), intermediate aquifer (14 wells) and deep aquifer (23 wells), which were selected from the residential (53 wells), agricultural (40 wells) and industrial areas (7 wells). The frequency of sampling in the groundwater wells was typically monthly, however missing sampling was observed during some months. With regard to the long term data and different sources, the annual mean value of long term data is applied in this study, which had the same time series trend and annual variation pattern, as in previous researches (Nur-Hayati, 2011; Sefie et al., 2015). Moreover, since the data are taken through time, missing observation are very common and can negatively affect time series analysis or even invalid the prediction. Three different methods include removing missing data, applying the mean of the adjacent observations, and Maximum Likelihood Estimation (MLE) are applied to substitute the missing values, which showed MLE is an accurate model for dealing with missing values in prediction modelling.

2.4. Time series analysis

The underlying relation between previous observed values could be investigated using stochastic models, which are often known as time series models and have significant capacity to predict future values (Asadollahfardi et al., 2011). Time series analyses have been demonstrated as being reliable and capable predicting models for various environmental, hydrological and water resource management studies. Three separate components, i) a trend or long-term component, ii) cyclical or oscillating trend, and iii) irregular or random component (García-Díaz, 2011) can influence the observation's sequence, which should be detected and deduced in the time series data.

2.4.1. Trend analysis

It is expected that the groundwater quality in Northern Kelantan, has been affected by land use changes and intensive human activities over the last decades. The Mann-Kendall (Mann, 1945; Kendall, 1975) test is the most common trend test in hydro-meteorological studies, which estimates trends using the Theil and Sen's median slope estimator for specific time periods by the percentage changes over the mean (Yue et al., 2002). Mann-Kendall is a non-parametric method and free from statistical distribution. According to the test, H_0 (null hypothesis) assumes that observations do not show any trend or serial correlation over time, and H_1 (alternative hypothesis) assumes that there is an increasing or decreasing monotonic trend in time series observations. A rejected null hypothesis reveals that "there is a trend in the time series nitrate concentrations in the groundwater". Moreover, Sen's slope estimator (Sen, 1968) could be applied to show if the time series observations show an increasing trend (positive value of Sen's slope) or a decreasing trend (negative value of Sen's slope).

2.4.2. Time series predicting model

There are several different approaches to fit an appropriate time series model to the observations. Several hydro meteorological studies applied ARIMA modelling to predict future values based on previous observations (Yurekli and Kurunc, 2006; García-Díaz, 2011;

Narayanan et al., 2013). In this study, the ARIMA model is applied to predict the nitrate concentration in the groundwater for the period 2015–2030 based on the nitrate data collected from 1989 to 2014 in Northern Kelantan. Box and Jenkins (1976) introduced a general prediction model based on an integration of autoregressive (AR), integrated (I), and moving average (MA), which is expressed as ARIMA (p, d, q).

Stationary is a prerequisite for ARIMA modelling. A differencing option should be performed when the observed value shows trend and heteroscedasticity. I (d) is part of the ARIMA model, which determines whether the time series observations are modelled directly or required differences between sequential observations. In practice, d is usually 0, 1 or 2 and rarely >2. A stochastic linear ARIMA model consists of four stages, exploration, estimation, diagnostic checking, and prediction (Box and Jenkins, 1976; Rodríguez-Iturbe et al., 1985).

2.4.2.1. Model exploration. The main idea of model exploration is that the time series generated by ARIMA model should have some theoretical autocorrelation properties (Zhang, 2003), which may identify by matching the empirical autocorrelation patterns with the theoretical patterns using the autocorrelation function (ACF) and the partial autocorrelation function (PACF) (Box and Jenkins, 1976). The general behavior of a univariate model is studied in two parts. First, the observed data are checked for stationarity and normality. A stationary time series has a quasi-normal distribution with zero mean and constant variance (Hassan, 2014). Proper differencing and transformation methods need to be performed in the non-stationarity time series to remove the trend and stabilize the variance before fitting the ARIMA model. Since water quality data does not have fixed mean, when the observed data shows trend and heteroscedasticity, application of a log or a square root transformation is suggested to reduce it to a stationary series (Narayanan et al., 2013).

Second, the behavior of ACF and PACF is used to show the stationarity and seasonality of series. The ACF is a useful tool to show if earlier values in the series have some relation to later values. PACF is the amount of correlation between variable and a lag of itself that is not

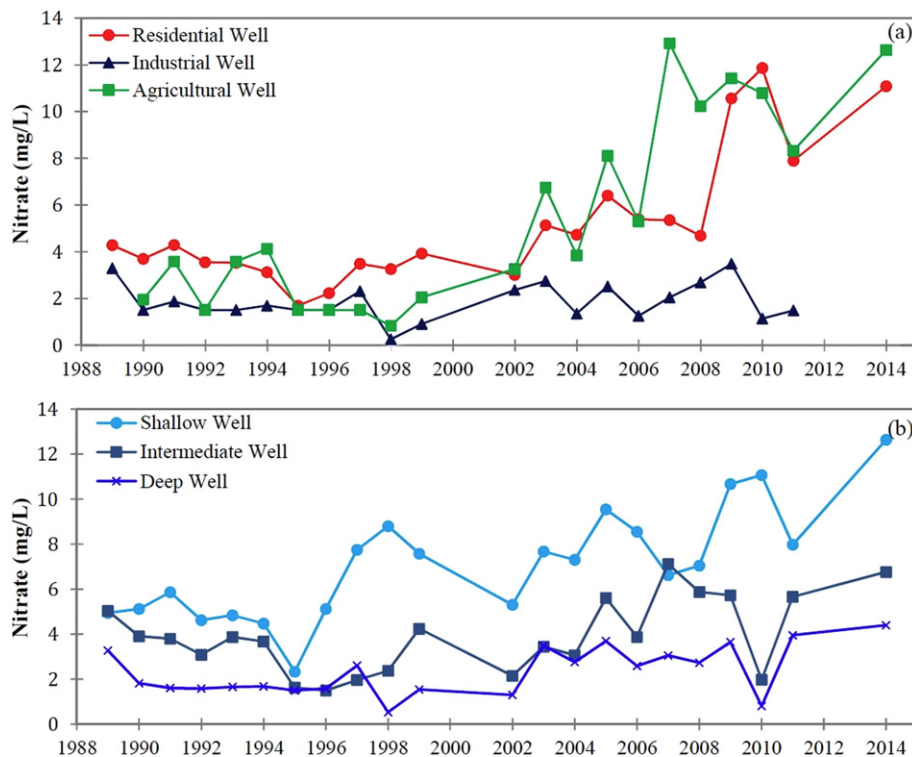


Fig. 2. Nitrate contamination variations based on land use activities (a) and aquifer depth (b) from 1989 to 2014 in the study area.

Table 1

Mann-Kendall's test and Sen's slope for nitrate contamination from 1989 to 2014 in Northern Kelantan.

	Residential wells	Agricultural wells	Industrial wells
Kendall's tau	0.489	0.571	0.013
p-Value (two-tailed)	0.001	0.000	0.955
Sen's slope:	0.256	0.509	0.000
Trend	Increasing	Increasing	No-trend

	Shallow wells	Intermediate wells	Deep wells
Kendall's tau	0.558	0.238	0.281
p-Value (two-tailed)	0.000	0.130	0.071
Sen's slope:	0.300	0.100	0.090
Trend	Increasing	No-trend	No-trend

explained by correlation at all low order lags. A tentative model is selected by matching ACF and PACF of stationary series.

2.4.2.2. Parameters estimation. The effective estimates of the parameters should be obtained after the tentative model is specified. The stationarity and inevitability for autoregressive and moving average models should be contented (Box and Jenkins, 1976). The significance of the parameters should be tested, which is associated with the parameter value are standard errors of estimate and related t-value.

2.4.2.3. Diagnostic checking. In the next step, after identification a tentative model and estimation of model's parameters, the model adequacy should be checked, which is basically done to check if the model assumption about the error, are satisfied (Samsudin et al., 2011). If the model is not adequate, a new tentative model should be identified, which is again followed by the steps of parameters estimation and model verification.

To evaluate the accuracy of the fitted model, root mean squared error (RMSE), mean absolute percentage error (MAPE), and correlation

coefficient between observed and predicted series is computed. Akaike Information Criterion (AIC) helps to identify best model, in absence of clear statistics results. A model with minimum AIC being considered. The AIC is expressed as:

$$AIC(p, q) = N \ln(\sigma_e^2) + 2(p + q)$$

where, p and q are the lags of the ARIMA (p, d, q).

For a reliable forecasting model, the residuals must satisfy the requirements of a white noise process such as uncorrelated and normally distributed around a zero mean. Testing for independence against serial dependence is a fundamental problem in time series analysis (Yurekli and Kurunc, 2006). To determine whether nitrate time series data are independent, the residual auto correlation (RACF) function of the series should be obtained. If the RACF's is significantly different from zero, it may reveal the inefficiency of the current model. Moreover, in order to test the null hypothesis that a current set of autocorrelations is white noise Ljung-Box statistic test (Q) is used to check the adequacy of the model. The test hypothesis can be checked for the model adequacy by choosing a level of significance and compared with χ^2 distribution with respective degree of freedom at 5% significant level. A p-value > 0.05 implies that the model is correctly specified to describe the correlation information in time series (Ljung and Box, 1978):

$$Q(r) = n(n+2) \sum_{k=1}^m (n-k)^{-1} r_k^2$$

Homoscedasticity is another important characteristic of residual which shows no change in variance of the residual. Breusch and Pagan (1979) introduced a useful test to determine heteroscedasticity of residuals. The residuals from the model fit to data should be divided into two

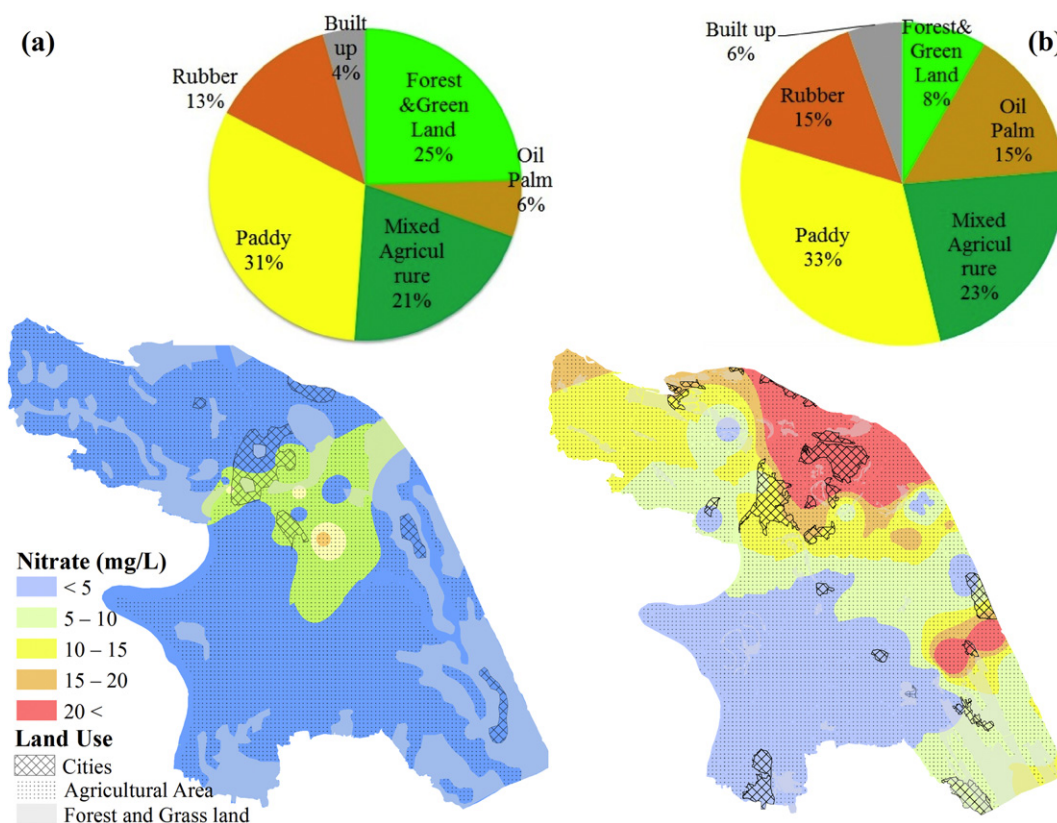


Fig. 3. Spatial distribution of nitrate contaminations in 1989 (a) and 2014 (b) in Northern Kelantan.

groups and residual sum of squares of the first and second group (ESS_F) and (ESS_S) should be obtained:

$$F_{cal} = \frac{ESS_H / (n_H - k_p)}{ESS_L / (n_L - k_p)} \approx F_{table}[(n_H - k_p), (n_L - k_p)]$$

where, k_p is the degree of freedom. If $F_{cal} < F_{table}$ critical value, the residuals are assumed to be homoscedastic.

The several statistical tests could be used to check normality assumption in the residuals. In this study Jarque-Bera (JB) test (Jarque and Bera, 1980) and Anderson-Darling test (Anderson and Darling, 1952) were used for the diagnostic checking of residuals for normality.

$$JB = \frac{N - n_p}{6} \times \left[S^2 + \frac{(K - 3)^2}{4} \right] \sim \chi^2_{(2)}$$

where, S is the skewness, K the kurtosis, n_p the number of parameters and N the sample size.

JB has an asymptotic chi-square distribution with 2° of freedom to verify if the skewness and kurtosis of the time series different from the expected normal distribution.

2.5. Geospatial technique

The spatial structure of the observed nitrate concentration could be predicted using the geospatial technique. Although, the geospatial

technique including deterministic and geostatistical techniques, were first applied in geosciences (Matheron, 1963, 1971), recently, the techniques have been widely applied by researchers in other fields of study. Deterministic methods are classified as global (uses the whole dataset for predicting each point) and local method (uses data in a neighborhood around the interpolation point) (Varouchakis and Hristopoulos, 2013), which can be either an exact or inexact interpolator (Webster and Oliver, 2007). Deterministic interpolation methods, such as Inverse Distance Weighting (IDW), estimate the value of an attribute at unsampled locations based on the weights assigned to each sample value depending on the distance between the sampled location and the unsampled location.

3. Results and discussion

3.1. Impact of land use changes on groundwater quality

In Northern Kelantan, the forest and grass land area covered nearly 25% of the total area in 1988. However, it decreased to 8.6% (73 km²) of the total area by 2011. In contrast to forest, which had a negative growth (around -4.5%) over the last 25 years, paddy fields, oil palm trees, mixed-agriculture lands, and built up areas had an annual growth of approximately 0.25%, 4.24%, 0.37%, and 0.95%, respectively, between 1988 and 2011. The Mann-Kendall test was applied to quantitatively reveal the effect of land use changes. The results from 25 years (1989–2014) of groundwater monitoring in Northern Kelantan shows a

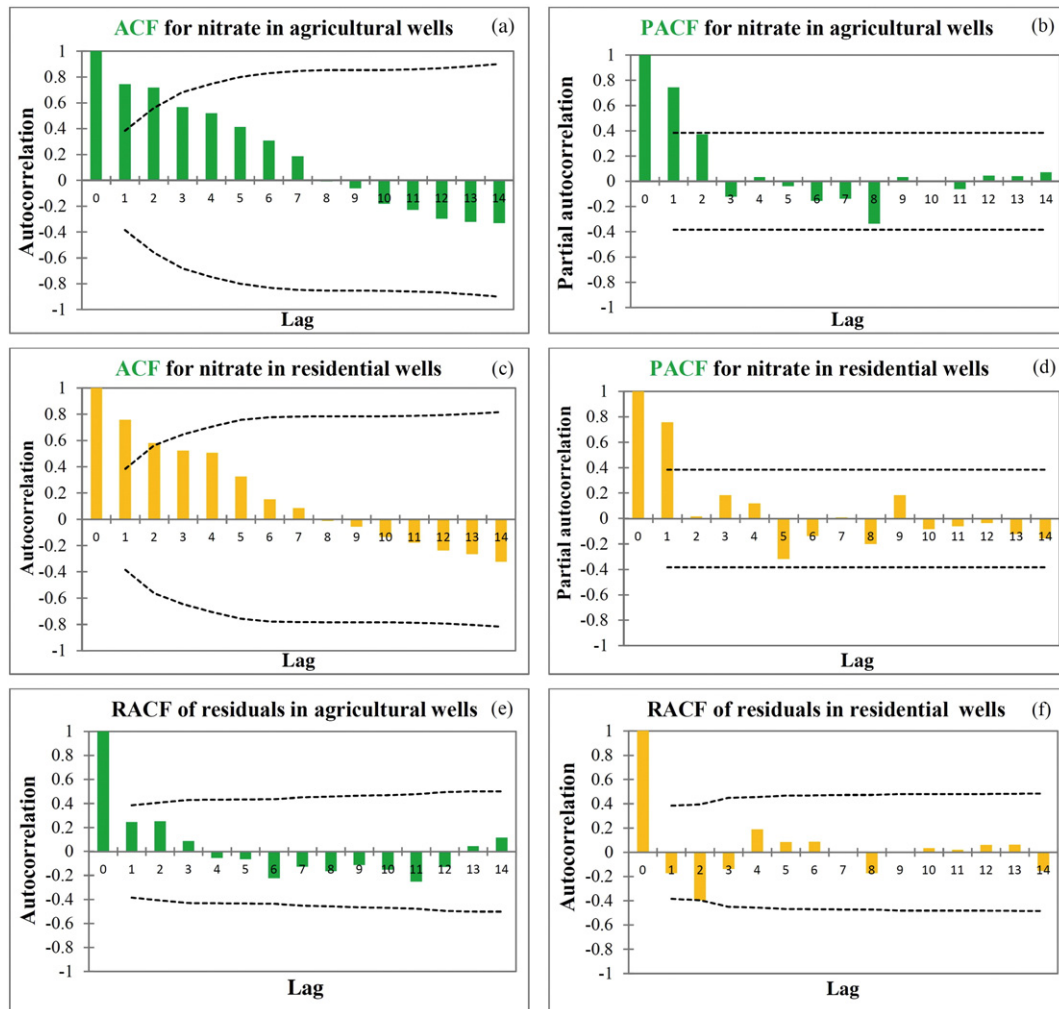


Fig. 4. Autocorrelation function (ACF) and partial autocorrelation function (PACF) of nitrate contamination in agricultural (a and b) and residential wells (c and d), and autocorrelation function (ACF) of residuals using differencing method (e and f).

coherent pattern in nitrate concentration. Based on Fig. 2 and Table 1, there are some statistically significant positive trends in nitrate concentration in the agricultural and urban sampling wells from 1989 to 2014. The pattern of long-term nitrate variations in the sampling wells when focusing on land use change; the data clearly reveal a significant trend of nitrate concentration in the agricultural wells and residential wells from 1988 to 2014 (Table 1). The positive values of Sen's Slope indicate an increasing trend in nitrate concentrations in both agricultural and residential wells. However, there is no significant trend in the time series data for nitrate concentration in industrial wells, which is also shown by the zero slope of the Sen's Slope (Table 1). This may be due to the depth of industrial wells in the study area, which are mostly (except one sampling well) classified as deep well (depth > 50 m) and abstracted water from deep aquifer.

Previous studies in the Kelantan State indicated that from 1989 to 1997 around 732 km² of permanent forest area had been converted into non-forestry purposes, including agriculture, residential, and mining with a deforestation of about 2.27% (Jusoff and Setiawan, 2003). Based on the ENEX Report (1977), groundwater is the main source of domestic and agricultural water supply for farmers in the Kelantan basin.

According to Keesstra et al. (2012), the application of inappropriate fertilizer under agricultural activities and disposal of waste waters can lead to an accumulation of nitrate, nitrite, and phosphate in soil, sediment, and water. In Northern Kelantan Basin, besides the extensive use of nitrogen fertilizers, the excessive rainfall in the region also contributes to nitrate leaching below the root zone and ultimately to the groundwater.

In the time window of 1989 to 2002 (13 years) the results indicate fluctuations in the nitrate concentration in the groundwater. However, there is no statistically significant trend in nitrate concentration during this period. Based on Fig. 2a, industrial wells present a lower concentration of nitrate with a minimum 0.25 mg/L in 1998. In contrast, residential wells reveal a higher concentration of nitrate with a maximum of

4.22 mg/L in 1991 in the study area. The findings indicate that the source of nitrate is not only from agricultural land, but is also associated with the residential areas, such as rural and urban lands.

There is strong positive trend for the time window starting from 2002 to 2014, despite some fluctuations, which mostly occurred in 2004 and 2006 and the gaps in data between years 2012 and 2013. Nitrate fluctuations mostly observed in groundwater belongs to agricultural wells and shallow wells, which may show the impact of heavy rainfall and runoff from farms on leaching of nitrate to groundwater.

Agricultural and residential wells reveal maximum concentrations of around 12.6 mg/L and 11.1 mg/L, respectively, in 2014. In total, the nitrate concentration in the groundwater showed an annual growth of 8.1% and 3.9% from 1989 to 2014 in the agricultural and residential wells, respectively. However, industrial wells present a very smooth pattern during these years.

Leaching is a direct loss of nitrate from a point or non-point source to below the root zone and finally to the deeper groundwater, which depends on the level and type of fertilization, soil properties, type of crops, and method of cultivation (Kurunc et al., 2011; Lawniczak et al., 2016). Therefore, it is expected that land use activities have a crucial impact on shallow aquifers. Unconfined aquifers are of primary concern due to groundwater pollution, especially where the overlying layer is relatively permeable and thin. Naturally, nitrate concentrations decrease with increasing depth to the groundwater. Shamshuddin et al. (2013) mentioned that the shallow aquifer is the main source of water for domestic and agricultural usage in the Kelantan River Basin, which is exposed to a higher risk of nitrate due to human activities on the surface.

The results of this study show a significant trend of nitrate concentration in the shallow aquifer (Fig. 2b, Table 1) from 1989 to 2014. The positive value of Sen's slope represents the increasing trend of nitrate concentration in the shallow aquifer, with a minimum value of 2.3 mg/L in 1995 and a maximum value of 12.6 mg/L in 2014. The increasing trend of nitrate concentration in the agricultural and

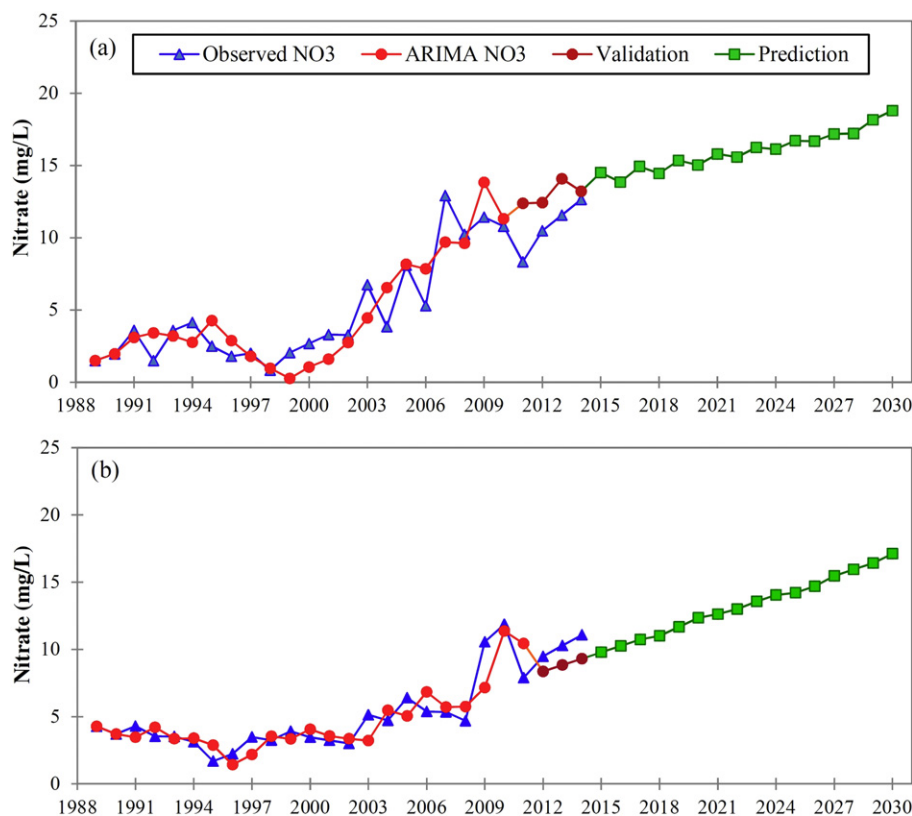


Fig. 5. Prediction of nitrate contamination from 2015 to 2030 in agricultural wells (a) and residential wells (b) in Northern Kelantan.

Table 2
Model statistics for nitrate contamination in agricultural and residential wells.

	Model	AIC	RMSE	MAPE	R ²
Agricultural wells	1, 2, 2	90.6	1.72	36.5	0.88
	2, 2, 2	92.3	1.68	37.7	0.85
	1, 2, 3	84.3	1.85	48.6	0.81
	2, 2, 3	93.5	1.52	39.1	0.87
Residential wells	1, 2, 2	86.6	1.30	18.9	0.83
	2, 2, 2	81.7	1.03	16.0	0.86
	1, 2, 3	68.7	1.06	18.4	0.82
	2, 2, 3	80.3	1.11	19.9	0.76

residential areas (Fig. 2a) and shallow wells (Fig. 2b) clearly indicate the significant role of manmade sources, such as the application of nitrogen fertilizers, livestock areas, discharge from septic tanks, and leaking sewers, in the degradation of groundwater quality, over 25 years in Northern Kelantan. In contrast to the shallow wells, the intermediate and deep wells do not show any significant variations in nitrate concentrations from 1989 to 2014. Although the intermediate aquifer shows a higher concentration of nitrate compared to the deep aquifer, the nitrate concentrations do not have meaningful trends over 25 years of observations.

3.2. Spatial pattern of land use changes on nitrate contamination

The spatial distributions of nitrate contamination in groundwater were prepared to compare the changes in nitrate concentration in groundwater between 1989 and 2014. The maps clearly show the changes in nitrate contamination in the study area between 1989 and 2014 (Fig. 3). Based on, Fig. 3a, in 1989, approximately 87.04% of the total area (744 km²) shows a nitrate concentration in groundwater of <5 mg/L. In total, around 99% of the study area (848 km²) had a nitrate concentration of <10 mg/L in 1989, which is the upper limit for nitrate concentration in drinking water by the WHO (2011) and USEPA (2011), and 7 mg/L which is upper limit for water required treatment in Malaysia (DOE, 2006). The historic land use map was overlaid to identify the impact of land use changes on groundwater quality variations (Fig. 3). However, even in the areas with agricultural activities and urbanization, the average of nitrate contamination was around 4.3 mg/L in year 1989. In contrast to 1989, the spatial distribution of nitrate contamination shows a significant increase in year 2014. Based on the Fig. 3b, approximately 62% (530 km²) show a nitrate concentration lower than the permissible limit for drinking water (10 mg/L). In the remainder of the area (around 316 km²) nitrate contaminations above 10 mg/L were found. The area around the Kota Bharu and towards the South China Sea show nitrate concentrations of >20 mg/L, which could threaten human health and the ecosystem in these areas. The highest measured levels of average nitrate concentration in 2014 up to 12.9 mg/L, which is approximately three times higher than the nitrate concentrations in 1989.

Although physical expansion of the forest and grassland obviously decreased from 1989 to 2014 in the study area (Fig. 3), it should be noted that nitrate moves relatively slowly in soil and groundwater (approximately between 10 and 50 years to deep water table (Alley, 1993)). Therefore, it is expected that the impact of the current land use changes on groundwater quality and nitrate contamination will become evident only in the coming years.

3.3. Prediction modelling of nitrate contamination

A stochastic linear ARIMA model is applied for predicting the nitrate contamination in agricultural and residential wells from 2015 to 2030 using input data from 1989 to 2014. In the first stage of time series analysis, the nitrate data are explored for normality and stationarity using trend analysis and the autocorrelation function. The differencing technique is applied to eliminate linear trends in time series data. In the present study, plots of ACF and PACF of nitrate data in agricultural and residential wells are presented in Fig. 4. The confidence limits are indicated by the continuous line below and above the x-axis in the plots (Fig. 4), which shows that the two spikes (at lag 1 and 2) that are above the confidence limits in the nitrate data belong to both agricultural and residential wells. The ARIMA (p, 2, q) model is recommended for eliminating the trend in the nitrate data using the differencing technique. There is no spike exceeding the confidence level after the application of the differencing method, which indicates that the nitrate series difference is stationary.

In the next step, the ARIMA model should be fitted after application of the differencing method. The classical Box and Jenkins (1976) approach is applied to identify the appropriate structure including AR (p) and MA (q). The ACF and PACF plots are used to decide tentative ARIMA models for nitrate contamination in agricultural and residential wells. Based on Fig. 5, the plot of ACF and PACF shows two spikes and one spike, respectively, in both wells, which can suggest $p = 1$ and $q = 2$ for nitrate contamination. Therefore, ARIMA (1, 2, 2) is proved to be the best fitted model for predicting nitrate contamination. However, eight tentative ARIMA models were chosen with different values of p, d, q, which were within the reasonable range (Table 2).

Out of the four selected models for time series nitrate concentration in agricultural and residential wells, the best models were selected based on the lowest root mean squared error (RMSE), mean absolute percentage error (MAPE), mean absolute error (MAE), Akaike Information Criterion (AIC), and the highest coefficients of determination (R²). The model that shows the lowest value for these parameters was chosen for validation. Although ARIMA (1, 2, 3) has lower AIC and ARIMA (2, 2, 3) has lower RMSE, in the case of nitrate contamination in agricultural wells ARIMA (1, 2, 2) has a higher R² and lower MAPE. In the residential wells ARIMA (2, 2, 2) shows lower values of RMSE, MAPE, and higher values of R², in contrast to the other suggested models. Therefore ARIMA (1, 2, 2) and (2, 2, 2) were selected as the fitted models for predicting nitrate contamination in agricultural and residential wells, respectively (Table 2).

Last step, the model's residuals are examined, which should show consistent behavior if the fitted model is appropriate. Two approaches, namely RACF and Ljung-Box test are applied for the independence assumption of the residuals for the best models. The RACF results (Fig. 4e and f) indicated that residuals were not significantly different from a white noise series at the 5% significant level. Moreover, the Ljung-Box test shows that the p -value was greater than α (0.05) for the residuals, which indicates that the residuals do not show dependency in both selected model in agricultural and residential wells (Table 3). The results related to the normality of residuals using Jarque-Bera test shows in agricultural wells p -value 0.786 > 0.05 and in residential wells p -value 0.753 > 0.05, which indicates the normal distributions of residuals. The results of JB test is confirmed using Anderson-Darling test, where p -value 0.711 and 0.682 > 0.05 in agricultural and residential wells, respectively (Table 3).

Table 3
Model validation parameters.

Sampling well	Selected model	Dependency Ljung-Box	Heteroscedasticity		Normality test	
			Breusch-Pagan test	White test	Anderson-Darling	Jarque-Bera
Agricultural wells	1, 2, 2	0.886	0.065	0.116	0.711	0.786
Residential wells	2, 2, 2	0.585	0.057	0.109	0.682	0.753

The Breusch–Pagan test and White test are used to check the homoscedasticity of the residuals. The results show no heteroscedasticity in the residuals of each time series dataset for both wells. Since the selected models are well-fitted, the residuals obtained have the property of white noise (independent over time and normal distribution) with no number of significant spikes of the residuals. The results of predicting the nitrate contamination in agricultural and residential wells using ARIMA (1, 2, 2) and (2, 2, 2) models are presented in Fig. 5.

The model outcomes suggest that the nitrate contamination will increase linearly from 2015 to 2030 in both wells in Northern Kelantan. Based on Fig. 5a, the nitrate contamination would increase from 13.6 mg/L in 2014 to approximately 18.8 mg/L in 2030 in the agricultural wells. As per Sen's slope estimate the annual growth rate of nitrate contamination from 1989 to 2014 was 8.1%, which would be decreased to 2.64% from 2015 to 2030. In residential wells, the nitrate contamination also would increase from 11.1 mg/L in 2014 to 17.1 mg/L in 2030 (Fig. 5b). The annual growth rate of nitrate contamination was 3.89 from 1989 to 2014, which was predicted to be stable (with 3.9% annual growth) from 2014 to 2030.

4. Conclusion

Deforestation and agricultural expansion are assumed to have significant impact on groundwater quality. The present study in Northern Kelantan, Malaysia, clearly shows and quantifies the significant role of land use changes and human activities on the degradation of groundwater quality over a quarter of a century. Based on the Mann–Kendal's test, the nitrate concentrations show a significant positive trend in both the agricultural and residential wells, while, the forests and grass lands show an annual decrease of rate about 4.5% from 1989 to 2014. Twenty-five years of data for the groundwater quality clearly reveal the negative impact of human activities by increasing nutrient input into the environment due to, sewage spills, and chemical fertilizers. It is obvious from the study that the shallow aquifer is much more vulnerable than the deeper aquifers to nitrate contamination. The nitrate concentration shows an annual increase of around 3.74% in the shallow aquifers; however, it did not show significant variations in the intermediate and deep aquifers from 1989 to 2015.

The spatial patterns of nitrate contamination reveal that only 1% of the total area had nitrate values > 10 mg/L in 1989. At the end of 2014, this percentage increased to nearly 48% of the total area. The modelling approach used in this study predicts an increasing annual trend of around 2.3% and 3.9% in agricultural and residential wells respectively.

Degradation of groundwater quality is a growing problem in the absence of any land and water management. The continuing deforestation in the study area can serve as an example for many areas around the world, and it is important to know the effects of these land use changes on the quality of groundwater resources. The prediction of future nitrate values is useful for developing sustainable land and groundwater resource management strategies for the future.

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