

# Contamination of nitrate in groundwater and its potential human health: a case study of lower Mae Klong river basin, Thailand

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**Abstract** Nitrate contamination in groundwater is a worldwide problem especially in agricultural countries. Environmental factors, such as land-use pattern, type of aquifer, and soil-drainage capacity, affect the level of contamination. Exposure to high levels of nitrate in groundwater may contribute to adverse health effects among residents who use groundwater for consumption. This study aimed to determine the relationship between nitrate levels in groundwater with land-use pattern, type of aquifer, and soil-drainage capacity, in Photharam District, Ratchaburi Province, lower Mae Klong basin, Thailand. Health risk maps were created based on hazard quotient to quantify the potential health risk of the residents using US Environmental Protection Agency (U.S. EPA) health risk assessment model. The results showed the influence of land-use patterns, type of aquifer, and soil-drainage capacity on nitrate contamination. It was found that most of the residents in the studied area were not at risk; however, a groundwater nitrate monitoring system should be implemented.

**Keywords** Nitrate · Groundwater · Health risk assessment · Methemoglobinemia · Lower Mae Klong river basin · Ratchaburi Province · Thailand

## Introduction

Nitrate contamination in groundwater is a worldwide problem (Spalding and Exner 1993; Dunn et al. 2005; Liu et al. 2005; U.S.EPA 1993; Kyllmar et al. 2004). Nitrate is soluble in water, easily leaches through soil, and accumulates in groundwater (Nolan 2001). The natural level of nitrate in groundwater is  $\leq 2$  mg/L  $\text{NO}_3\text{-N}$  (Mueller and Helsel 1996) or 10 mg/L  $\text{NO}_3^-$ . Nitrate levels exceeding the background level may indicate contamination from sewerage, septic systems, industrial wastewater, and fertilizer (U.S.EPA 1993; Kyllmar et al. 2004; Liu et al. 2005; Burkart and Kolpin 1993; Mohamed et al. 2003; Keeney 1986; Eckhardt and Stackelberg 1995). Factors associated with the level of nitrate contamination include geological background, land-use pattern, agricultural practice, soil-drainage capacity, and type of aquifer (Nolan 2001; Hatfield and Follett 2008; Gardner and Vogel 2005; Meinardi et al. 1995; Fewtrell 2004; Dubrovsky and Hamilton 2010).

Exposure to high levels of nitrate from the consumption of contaminated water contributes to methemoglobinemia. The severity of this syndrome depends on exposure dose and individual susceptibility such as age, gender, genetics, or other health conditions. Infants aged below 6 months are at the highest risk of this syndrome due to low levels of a key methemoglobin reduction enzyme in the red blood cells (Shearer et al. 1972; Fan and Steinberg 1996; Avery 1999; Knobloch et al. 2000; Trivedi and VEDIYA 2012; U.S. EPA 2013). It has also been reported that older children exposed to  $>50$  mg/L  $\text{NO}_3^-$  nitrate in drinking water had a higher prevalence of

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methemoglobinemia than those exposed to drinking water containing nitrate  $<50$  mg/L  $\text{NO}_3^-$  (Sadeq et al. 2008). In addition, long-term exposure to nitrates might elevate the risk of non-Hodgkin's lymphoma (Weisenberg 1990; Payne 1993; Bruning-Fann and Kaneene 1993; van Maanen et al. 1994; Ward et al. 1996; Parslow et al. 1997; Barrett et al. 1998; Weyer et al. 2001; Wolfe and Patz 2002).

Nitrogen-based fertilizers are applied to enhance crop productivity. After nitrogen-based fertilizer treatment, soil microorganisms transform nitrogen into ammonium-nitrogen and nitrate-nitrogen, which plants use for growth (Frate 2007). Nitrogen in excess of plant uptake will leach through the soil, leading to groundwater contamination. Fertilizer consumption in Thailand has continued to increase since 2004 (Bureau of Agriculture Policy and Planning 2010). Some studies reported the level of nitrate contamination in groundwater to exceed the acceptable level of groundwater quality for drinking purpose (Kwanmeung et al. 2002; Rangsayatorn 2006; Tirado et al. 2008).

Photharam District, Ratchaburi Province, is located in the lower Mae Klong basin, in western Thailand. The area is primarily agricultural, and some residents still use groundwater for consumption. This study aimed to assess the relationship between land-use type, type of aquifer, and soil-drainage capacity with nitrate levels in groundwater. In addition, the potential health risks, in terms of the residents' hazard quotient (HQ) from exposure to nitrate in groundwater, were calculated using the health risk assessment model (U.S. EPA Region 6 2005); health risk maps were also created.

## Materials and methods

### Study area

Photharam District, Ratchaburi Province, is located in the lower Mae Klong basin in western Thailand. It covers an area of 417.009 km<sup>2</sup>. The topography consists mainly of a flat plain, which is generally covered by agricultural land and communities, with a small area of forest in the west. It is divided into 19 subdistricts, 156 villages, 6 municipalities, and 14 subdistrict administrative organizations. The total population in 2007 was 830,275; most worked in the agricultural sector. The most important water resource is the Mae Klong river (Amphoe 2008). Groundwater supplies in this area are obtained mainly from unconsolidated deposits of flood plain, delta, and terraces. The sediments are more than 400 m thick, with at least three aquifers from 200 m deep to land surface. An intermediate and deep aquifer is separated by a thin confining clay layer, leading to hydraulic interconnection between layers. The average annual recharge was

estimated at 1.50 cm or approximately 10 % of annual rainfall. Groundwater discharge occurs by various means including plant transpiration, springs and diffusion, or discharge into streams. (Dutta et al. 1998).

### Sources of data

#### *Land-use data*

Land-use data were extracted from the Land Development Department land-use map (2001). The data were classified into three groups: paddy field, sugarcane, and mixed orchard, according to the major characteristic of the area (<http://photharam.ratchaburi.doae.go.th>).

#### *Aquifer data*

Aquifer data was derived from the Department of Groundwater Resources hydrogeology map (2001). Aquifers were classified into three types: Permian Carboniferous metasediment aquifer (PCMs), floodplain deposit aquifer (Qfd), and younger terrace deposit aquifer (Qyt). PCMs are metasediment aquifers composed of clastic sedimentary rock including quartz, feldspar, phyllite, and slate. Tuff and agglomerates can be found in rock fractures. These aquifers yield 5–10 m<sup>3</sup>/h. Qfd are alluvial floodplain aquifers composed of gravel, sand, and clay. These aquifers occur within the floodplain depths ranging between 20 and 60 m and yield from 20 to 50 m<sup>3</sup>/h. Qyt are composed of gravel, sand, silt, and clay next to the alluvial floodplain aquifer. The average yield is 20 m<sup>3</sup>/h with a depth range of 30 to 100 m.

#### *Soil-drainage capacity data*

Soil-drainage capacity was categorized into four groups—very well drained, well drained, moderately drained, and poorly drained, according to soil texture, as indicated in soil series data from the Land Development Department (2014).

#### *Groundwater nitrate concentration data*

Groundwater nitrate concentration data used in this study was derived from the study of Wongsanit (2009). The level of nitrate contamination in groundwater was classified into three categories—background level ( $<10$  mg/L  $\text{NO}_3^-$ ), acceptable level (range from 10 to 45 mg/L  $\text{NO}_3^-$ ), and exceed the acceptable level ( $>45$  mg/L  $\text{NO}_3^-$ ).

#### *Data analysis*

Statistical analysis was performed using SPSS statistical software (version 18). The Kruskal-Wallis test was used to determine relationships between groundwater nitrate

concentration and environmental factors (land-use pattern, type of aquifer, and soil-drainage capacity). Significance was set at 95 % (or  $\alpha=0.05$ ).

#### Quantitative health risk assessment

According to the inadequate amount of evidence in humans for the carcinogenicity of nitrate in drinking water, only noncarcinogenic health effects posed by long-term exposure to nitrate in drinking water were quantified in terms of hazard quotient (HQ) using the US EPA health risk assessment model (U.S. EPA Region 6 2005) as shown in Eq. (1).

$$HQ = CDI/RfD \quad (1)$$

where *CDI* is the sum of nitrate intake via drinking water as shown in Eq. (2) (U.S. EPA 1989; U.S. EPA 1991), and *RfD* is a nitrate reference dose which is 1.6 mg/kg/day (IRIS 2012).

$$CDI = C \times IR \times EF \times ED / (BW \times AT) \quad (2)$$

where

C	Nitrate concentration (mg/L)
IR	Intake rate (1 L/day for children and 2 L/day for adults)
EF	Exposure frequency (365 day/year)
ED	Exposure duration (6 years for children and 30 years for adults)
BW	Body weight (15 kg for children and 60 kg for adults)
AT	Averaging time (365 days/year×6 years for children and 365 days/year×30 years for adults)

An HQ value >1 indicated a significant noncarcinogenic risk level (U.S. EPA Region 6 2005).

#### Health risk mapping

The HQ value of each sampling point was calculated and used to simulate the health risk level over the entire study area. ArcGIS Desktop version 9.1 (with Geostatistical Analyst extension) was used to generate a predictive risk area from HQs in the form of a risk map using ordinary kriging interpolation method, which provided the least error HQ prediction. The risk area was classified into four classes: class 1,  $HQ \leq 1$ ; class 2,  $HQ \geq 1-2$ ; class 3,  $HQ \geq 2-3$ ; and class 4,  $HQ > 3$ .

## Results and discussion

### Nitrate concentration in groundwater

Groundwater nitrate concentrations of 73 sampling wells (Fig. 1) derived from the study of Wongsanit (2009) were used

in this study. Nitrate concentrations ranged from 1 to 110 mg/L  $NO_3^-$  with the mean of 8.17 mg/L  $NO_3^-$ , and the median of 1.1 mg/L  $NO_3^-$ . Four samples exceeded the acceptable level of groundwater quality for drinking purpose (Ministry of Natural Resources and Environment 2008). The results showed most of the samples (80.8 %) had nitrate contamination levels <10 mg/L  $NO_3^-$ , 13.7 % had nitrate levels between 10 and 45 mg/L  $NO_3^-$ , and four samples (5.5 %) had >45 mg/L  $NO_3^-$  (Fig. 2).

The distribution of nitrate contamination levels in different land-use patterns is shown in Fig. 3. Among four groundwater samples containing nitrate more than the acceptable level, three were in sugarcane planting areas and one sample was in a mixed orchard area. The median nitrate concentration in the mixed orchard, paddy field, and sugarcane planting areas was 1.1, 5.8, and 14.5 mg/L  $NO_3^-$ , respectively. The Kruskal-Wallis test showed nitrate contamination levels among land-use patterns were statistically significantly different ( $p < 0.05$ ) (Table 1).

The distributions of nitrate contamination levels in the three aquifer types are shown in Fig. 4. The median nitrate concentrations in PCms, Qfd, and Qyt were 10.15, 1.1, and 1.05 mg/L  $NO_3^-$ , respectively. The Kruskal-Wallis test showed nitrate contamination levels among aquifer types were statistically significantly different ( $p < 0.05$ ) (Table 1).

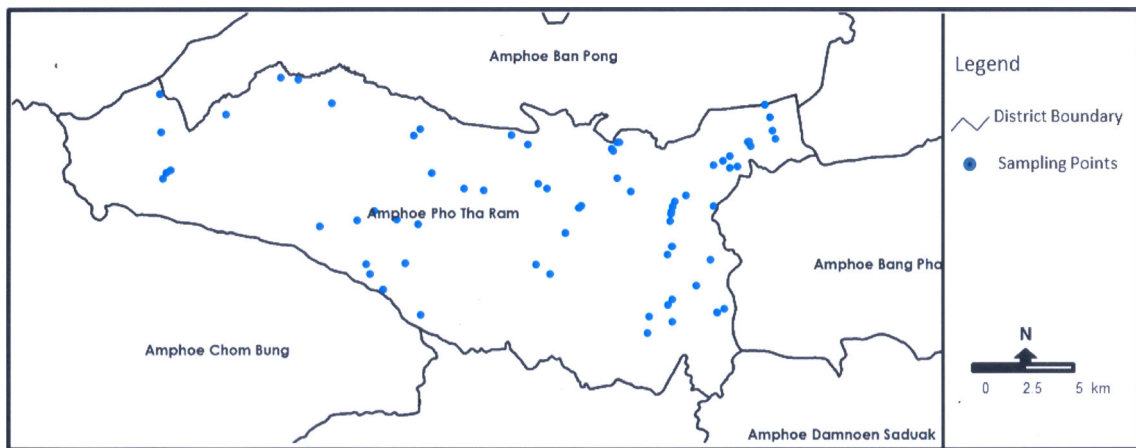
The distributions of nitrate contamination levels in the area with different soil-drainage capacities are shown in Fig. 5. The median nitrate concentration in very well drained, well drained, moderately drained, and poorly drained soils were 8.5, 1.1, 10.25, and 1.1 mg/L  $NO_3^-$ , respectively. The Kruskal-Wallis test showed that nitrate levels differed significantly by soil-drainage capacity ( $p < 0.05$ ) (Table 1).

### Human health risk of nitrate in groundwater

Human health risk was calculated based on the US EPA model and the parameters previously described. The results showed that the HQ values ranged between 0.04 and 4.58 for children and 0.02 and 2.29 for adults. The mean±SD of HQ values for children was  $0.34 \pm 0.72$  and for adults,  $0.17 \pm 0.36$ . The highest HQ values were 4.58 and 2.29 for children and adults, respectively. There were five wells with HQ values >1 for children and four wells with HQ values >1 for adults.

### Health risk map

In this study, ordinary kriging, multiquadric, thin-plate spline, inverse distance weighting, inverse multiquadric, and simple kriging were performed to create the prediction models based on the HQ value of each well. The mean absolute percentage error (MAPE) was calculated to validate the models. The results showed ordinary kriging was the most appropriate model



**Fig. 1** Sampling locations

since it provided the smallest MAPE compared with the other models. Ordinary kriging was used to create a health risk map using spherical semivariogram parameters. The range, nugget, and partial sill were 13,825, 0.034, and 0.277 m, respectively. The predictive ability of the model was assessed by cross-validation. The MAPE of the final model was 5.63.

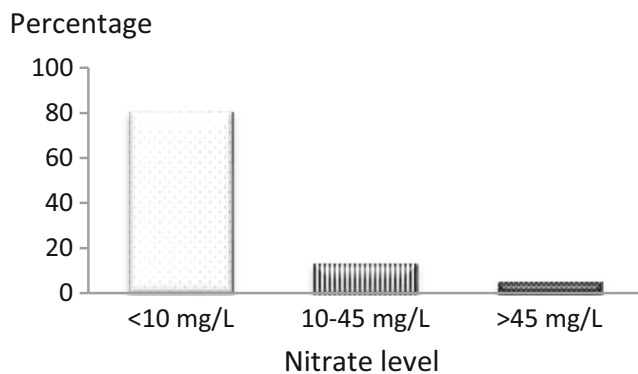
The health risk map of the children showed that 92.81 % of the study area was class 1; 5.99 %, class 2; 0.96 %, class 3; and 0.24 %, class 4 (Fig. 6). For the adults, most of the study area (98.80 %) was class 1, 1.16 % was class 2, and 0.04 % was class 3 (Fig. 7).

**Discussion**

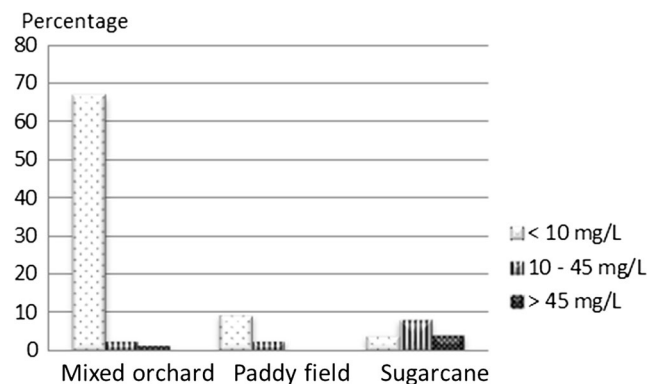
Groundwater in some locations of Potharam District, Ratchaburi Province, had nitrate concentrations higher than the acceptable level of groundwater quality for drinking purpose, the highest concentration was 110 mg/L NO<sub>3</sub><sup>-</sup>. This agreed with several studies that found high nitrate levels in groundwater, especially in intensive agricultural areas. The highest concentration of nitrate in groundwater in the study area was lower than that found in Nakhon Pathom and

Kanchanaburi province, but higher than Chiang Mai and Suphan Buri Province (Phupaibul et al. 2004; Tirado 2007). Compared with other studies, the highest groundwater nitrate concentration in the study area was lower than those found in Spain and in northeastern Romania (Guimera 1998; Căilean et al. 2009). However, it was higher than those found in Taiwan (96.06 mg/L NO<sub>3</sub><sup>-</sup>), the Philippines (109.2 mg/L NO<sub>3</sub><sup>-</sup>), northeast China (42.15 mg/L NO<sub>3</sub><sup>-</sup>), and Malaysia (42.49 mg/L NO<sub>3</sub><sup>-</sup>) (Lui et al. 2011; Tirado 2007; Su et al. 2013; Jamaludin et al. 2013). The presence of high levels of nitrate in groundwater worldwide indicates that groundwater quality is being deteriorated, and the population’s health might be affected from exposure to nitrate in groundwater.

Our results also demonstrated the influence of land-use pattern, type of aquifer, and soil-drainage capacity on nitrate concentrations in groundwater. Sugarcane planting areas had the highest median nitrate concentration, followed by paddy field and mixed orchard; the Kruskal-Wallis test showed a significant difference in nitrate concentration by land-use pattern (*p*<0.05). The significant differences in groundwater nitrate levels between land-use patterns are likely to have resulted from different rates of fertilizer application in sugarcane



**Fig. 2** Percentage of groundwater samples in the study area by nitrate contamination level



**Fig. 3** Percentage of nitrate contamination levels for different land-use patterns

**Table 1** Relationship of groundwater nitrate concentration and land-use pattern, type of aquifer, and soil-drainage capacity

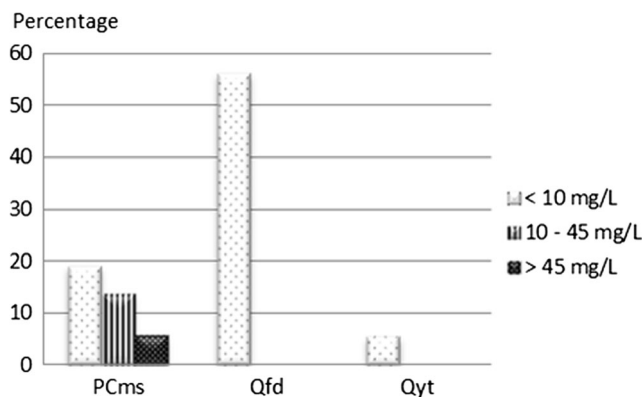
	Number of samples	Mean NO <sub>3</sub> (mg/L NO <sub>3</sub> <sup>-</sup> )	Median NO <sub>3</sub> (mg/L NO <sub>3</sub> <sup>-</sup> )	Range NO <sub>3</sub> (mg/L NO <sub>3</sub> <sup>-</sup> )	<i>p</i> value
<b>Land-use pattern</b>					
Paddy field	9	6.59	5.80	1.10–17.00	.000
Mixed orchard	52	3.66	1.10	1.00–49.00	
Sugarcane	12	28.92	14.50	1.10–110.00	
<b>Type of aquifer</b>					
PCms	28	19.37	10.15	1.10–110.00	.000
Qfd	41	1.21	1.10	1.00–3.20	
Qyt	4	1.05	1.05	1.00–1.10	
<b>Soil-drainage capacity</b>					
Very well drained	4	9.08	8.50	5.30–14.00	.001
Well drained	41	4.82	1.10	1.00–49.00	
Moderately drained	8	17.71	10.25	4.20–51.00	
Poorly drained	20	11.03	1.10	1.00–110.00	

planting areas, mixed orchards, and paddy fields, which were 15, 7, and 6 kg N/Rai, respectively (Department of Agriculture 2014). Although fertilizer application in mixed orchard areas was greater than that in the paddy fields, the median groundwater nitrate concentrations did not correlate, which may be due to excessive fertilizer being applied to paddy fields (Witheetrirong et al. 2011). The result agrees with several studies that found the significant relationship between crop type/land-use and the level of groundwater nitrate (Zhang et al. 1996; Burow et al. 1998; Eckhardt and Stackelberg 1995; Nolan and Stoner 2000; Nolan 2001; Tong and Chen 2002; Jeyaruba and Thushyanthy 2009; Lockhart et al. 2013).

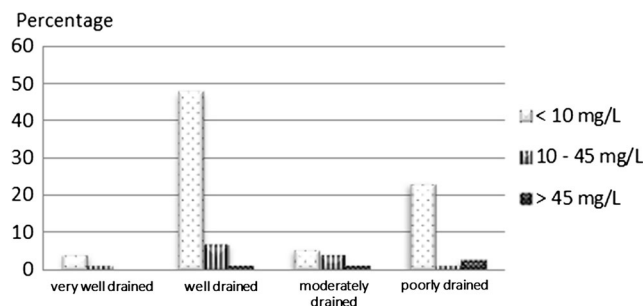
In terms of the type of aquifer, it was found that nitrate concentrations at sites located in a consolidated aquifer (PCms) were higher than those in an unconsolidated aquifer (Qfd and Qyt); the Kruskal-Wallis test showed a significant difference in nitrate concentrations in groundwater by aquifer type (*p*<0.05). This finding does not support the theory that unconsolidated aquifers facilitate water and contaminant leaching into the water table, leading to an increase in groundwater nitrate concentration (Nolan et al. 2002). This might be

because unconsolidated aquifers are commonly adjacent to underlying rivers so that the groundwater nitrate level might be affected by recharge/discharge mechanism between groundwater and surface water (Nolan 2001; Hatfield and Follett 2008). In addition, the consolidated aquifers have fewer pores than unconsolidated aquifers, resulting in less water being available to dilute the contaminants that might contribute to the higher level of groundwater contamination (Morris et al. 2003).

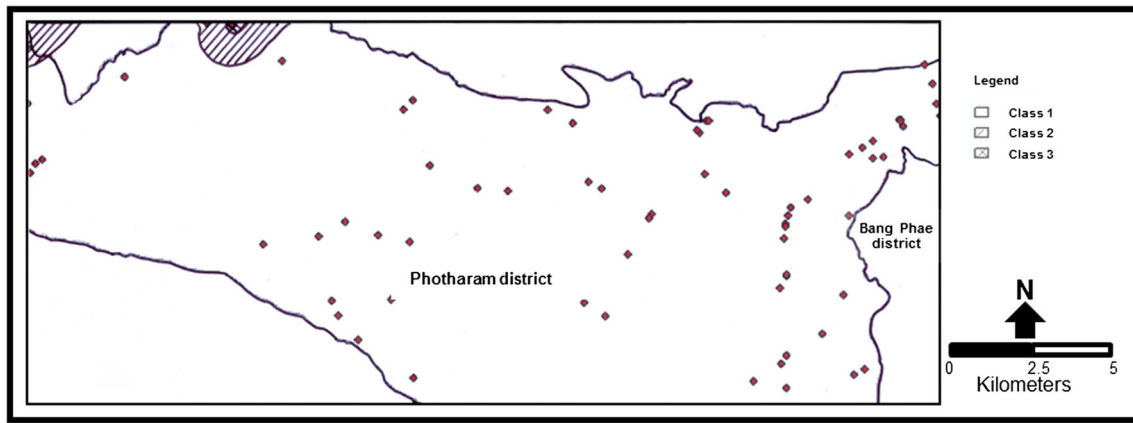
In general, poorly drained soils can restrict the movement of nitrate into the water table (Gaines and Gaines 1994; Mueller et al. 1995) so that groundwater below that area should be at lower risk of nitrate contamination (Sophocleous et al. 1990; Townsend and Marks 1990). However, our study found that the median nitrate contamination levels at sites located in moderately drained soil were higher than those at sites located in very well-drained soil and well-drained soil; this might be due to the influence of other soil characteristics including soil depth and organic matter content on the possibility of nitrate percolation through the soil (Cook 1990; Gardner and Vogal 2005; Meinardi et al. 1995; Fewtrell 2004; Dubrovsky and Hamilton 2010). In



**Fig. 4** Percentage nitrate contamination levels for different aquifer types



**Fig. 5** Percentage nitrate contamination levels for different soil-drainage capacities



**Fig. 6** Children’s health risk areas in Photharam District, Ratchaburi Province, by class

addition, a higher level of groundwater nitrate in the area beneath moderately drained soil than well-drained soil might be affected by the higher nitrate leakage rate at medium depth in clay soil compared with sandy soil (Ehteshami et al. 2013) or a correlation of higher-risk land uses, such as sugarcane on moderately drained soil.

In this study, several interpolation methods were used to create predictive models based on the HQ value of each well. Ordinary kriging was the most appropriate model since it provided the smallest MAPE compared to the others. Several studies found ordinary kriging performed better than other methods (Reza et al. 2010; Baskan et al. 2009). However, its performance might be affected by several factors that characterize the datasets being interpolated, such as sampling density, sample distribution, and homogeneity/heterogeneity (Eldeiry and Garcia 2012). To determine the nugget effect caused by spatial heterogeneity, the ratio of nugget sill was calculated. The ratio of nugget sill in this study was 12.27 %, which indicated low heterogeneity or low nugget effect of the study area, leading to high predictive reliability.

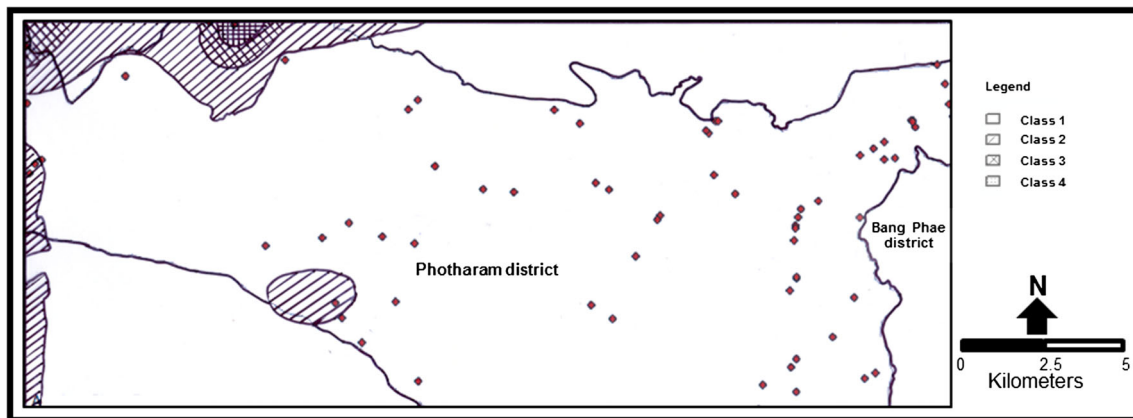
The health risk maps created by ordinary kriging using spherical semivariogram parameters with the range of 13, 825 m, nugget of 0.034, and partial sill of 0.277 showed that

some part of the study area had  $HQ > 1$ , which indicated the residents in that area are prone to the adverse health effects of daily intake of groundwater contaminated by nitrate. In addition, the area with  $HQ > 1$  that mostly overlaps with sugarcane planting area also indicates the potential effect of land use on nitrate contamination in groundwater as previously mentioned.

Our study mainly focused on determining the effect of land-use pattern, type of aquifer, and soil-drainage capacity on groundwater nitrate contamination in the study area. However, several other factors are related to nitrate contamination level in groundwater, i.e., climate, precipitation, cropping system, agricultural practice, irrigation methods, well depth, and aquifer yield (Lichtenberg and Shapiro 1997; Nolan 2001; Debernardi et al. 2008; Jones and Olson-Rutz 2011; Letey and Vaughan 2013). Further research should be conducted that encompasses all potentially significant factors.

**Conclusion**

The study results indicate that fertilizer application contributes to groundwater contamination by nitrate. Regarding potential



**Fig. 7** Adults’ health risk areas in Photharam District, Ratchaburi Province, by class

health risk from exposure to nitrate in groundwater, although the result from this study indicated that most residents in Photharam District were not at risk, some areas had HQ >1. Therefore, it is recommended that nitrogen management practices should be implemented, and groundwater nitrate should be monitored to prevent the adverse effect to human health.

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**Compliance with ethical standards** The authors declare that the manuscript is in compliance with ethical standards.

**Conflict of interest** The authors declare that they have no conflict of interest.

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