A lumped-parameter model for investigation of nitrate concentration in drinking water in arid and semi-arid climates and health risk assessment

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Abstract

Purpose This study was conducted to assess the capability of the lumped parameter model (LPM), an efficient model due to its analytical nature and the limited data requirements, to estimate health risks from nitrate in groundwater in arid and semi-arid climates.

Methods To assess the capability of LPM, two scenarios were established: one for estimation of hazard quotient (HQ) via monitoring nitrate concentration in groundwater and the other using the LPM. After nitrate was monitored in 148 randomly-selected wells, a modified LPM was used to estimate water volume and nitrate concentration, which ultimately led to the development of a model for estimating HQ. The performances of LPM were assessed using the coefficient of determination, percentage standard deviation, and root mean square error. To compare health risk maps Kriging, Spline, Inverse distance weighted, and natural neighbor models were run using geographical information system (GIS).

Results Linear analysis revealed a strong correlation between HQ values estimated in LPM and monitoring scenarios in arid climate compared to semi-arid ($r = 0.962$, $n = 22$, $p = 0.00$), suggesting that the LPM was more accurate in predicting nitrate concentration in the arid climate. Uncertainty analysis showed that LPM outputs were sensitive to several parameters, especially leakage from cesspits, which are involved in the sources and sinks of nitrate in the groundwater. In addition, it was found that the natural neighbor was the most appropriate model with the lowest errors for preparing health risk maps from nitrate.

Conclusions The obtained results revealed that LPM can be effectively used to estimate nitrate concentration in groundwater in arid climates and thereby LPM is an appropriate model to estimate health risk from nitrate in this climate.

Keywords Climates · Groundwater · Health risks · Lumped parameter model · Nitrate

Introduction

Groundwater is the main source of drinking water for many people around the world, particularly in rural areas. Urban, commercial, industrial, and agricultural activities can affect the quality of groundwater. In addition, population growth,

land change, and droughts bring about increased mineral and chemical groundwater pollutions [1, 2]. Pollution of groundwater can reduce the quality of drinking water, rise the costs of alternative methods of supplying drinking water, and increase the potential health problems [2].

Nitrogen is an essential nutrient in plant growth and sustainable agriculture and naturally occurs in the soil. Moreover, it occurs anthropogenically as a by-product of human waste [3]. The excess nitrogen may leach into groundwater usually in the form of nitrate [4]. Since the 1970s, pollution of groundwater with nitrate has become a key environmental issue in many parts of the world [5]. Groundwater nitrate concentrations in Europe exceed the WHO recommendations for drinking water in 22% of cultivated land [6]. Similar results have been reported in other studies, especially in arid climates, which indicate nitrate concentrations have increased in groundwater in past decades [7–9].

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The consequences of increased nitrate in water bodies are related to long-debated health concerns, cancer, and environmental impacts such as eutrophication [10, 11]. In addition, estimation of health risk from nitrate in drinking water resources are presented in many previous studies [12–14]. These concerns have led to the drafting of regulations regarding monitoring and remediation of nitrate in water resources in different countries [2].

Although many complex models have been developed to simulate the behavior of nitrogen in aquifers, the practical use of such model is complicated [15, 16]. The lumped parameter model (LPM) is a parametric model that was first introduced to interpret environmental radioisotopes in groundwater hydrology [17]. This model was developed to evaluate nitrate leakage from the unsaturated zone using limited information [18]. It allows the simulation of a system with fewer data requirements compared to their distributed counterparts [1]. Many studies have utilized the LPM to assess the efficacy of management options in remedying a situation and protecting groundwater quality from nitrate pollution [18, 19].

The present study was implemented to develop a LPM to assess non-cancer risks from exposure to nitrate in groundwater located in arid and semi-arid climates. Generally, risk assessment is a conceptual framework that investigates information related to the estimation of health and environmental consequences and operates in four stages, including hazard identification, dose-response assessment, exposure assessment, and risk characterization [20]. For this purpose, the following questions were addressed: (a) how do uncertainty factors affect the estimation of nitrate levels in groundwater?, and (b) do different climatic features affect the health risk from nitrate in groundwater? This study is designed to estimate health risks from nitrate in two scenarios: (a) estimation of hazard quotient (HQ) by monitoring nitrate concentration in groundwater, which is named the monitoring scenario, and (b) estimation of nitrate levels and HQ using a modified LPM, which is named the LPM scenario.

Materials and methods

Nitrate analysis

A cross-sectional study was performed to monitor nitrate content in randomly-selected groundwater wells in the Qazvin plain, Iran, located in arid and semi-arid climates with an area of about 74,737 km² in 2016 and 2017. The climates were specified via the Dommartin method based on temperature and annual rainfall [21]. Initially, approximately 620 underground wells were considered. Then, determination of sample size was done based on

Cochran formula [22]. Finally, 148 wells were randomly selected by setting the confidence level at 95% and the average standard deviation at 1 mg/L. The sampling stations are presented in Fig. 1. A one-liter grab sample was taken in a glass bottle from each selected well. The water samples were transported to a laboratory under controlled temperature conditions. Nitrate analysis was performed as soon as possible in accordance with the standard methods [23] with a spectrophotometer DR6000 (HACH) at a wavelength of 220 nm.

Development of the LPM

After analysis of nitrate in laboratory, the LPM was developed for estimation of nitrate in all selected groundwater wells. Estimation of water volume and nitrate concentration in the selected groundwater was the main concern in the use of the LPM. This study follows the application of the modified LPM, which assesses pollution of groundwater with nitrate [18]. The estimations were under the assumption that nitrate changes take place in the unsaturated zone, and first-order reactions are dominant in the aquifer. Lateral outflow and water pumped for irrigation and domestic uses were the sinks. Nitrate from the lateral flow, artificial recharge, fertilizer surplus, and natural recharge were considered as inputs to the model domain. Furthermore, nitrate lost through lateral outflow, irrigation, domestic use of groundwater, and denitrification were considered as outputs from the aquifer [18, 19].

Estimation of water volume in the aquifer

The water volume (L^3/T) in the aquifer with an elevation of h_1 (V_w) was determined using Eq. 1.

$$V_w = \Delta S_w + V_{w0} \\ = [G_{in} + Q_{Ar} + R_{ra} + R_{ir} + R_{wwl} + R_{wl} + R_{CSPT} - G_0 - Q_{irr} - Q_{DO}] + V_{w0} \quad (1)$$

where ΔS_w is the change in the water storage in the aquifer (L^3/T), V_{w0} is the water volume in the aquifer at the beginning of each time step (L^3), G_{in} is lateral inflow, Q_{Ar} is artificial recharge, R_{ra} is recharge from rainfall, R_{ir} is recharge from irrigation return-flow, R_{wwl} is recharge from wastewater leakage, R_{wl} is recharge to the aquifer from the leakage of water from the distribution network, R_{CSPT} is recharge from cesspits, G_0 is lateral outflow, Q_{irr} is the total monthly volume of water used for irrigation, and Q_{DO} is nitrate lost from the aquifer through water pumped for domestic purposes [18, 19]. The parameters used for “Estimation of water volume in the aquifer” and “Estimation of nitrate concentration in the aquifer” are listed in Table 1.

Fig. 1 Sampling stations in the arid and semi-arid climates in the study area in Qazvin province

Estimation of nitrate concentration in the aquifer

Nitrate concentration in the aquifer was estimated using Eq. 2 (L/T).

$$C = (\Delta S_N + V_{w0}C_0)/V_W \quad (2)$$

where C_0 is average nitrate concentration (M/L^3) at the beginning of each time step, and C is average nitrate level at the end of each time step (M/L^3). At the first time step, C_0 equals the initial concentration in the aquifer. In addition, ΔS_N is the change in the mass of nitrate in the aquifer for each time step (M/T) and is calculated via Eq. 3.

$$\Delta S_N = NO3(G_{in} + Q_A + SURP + R_{ra} + R_{ir} + R_{wwl} + R_{wl} + R_{CSPT}) / NO3(G_0 + irr + DO + DEN) \quad (3)$$

where $NO3_{Gin}$ and $NO3_{SURP}$ are the mass of nitrate that enters the aquifer through lateral inflow and fertilizers, respectively. Moreover, $NO3_{QA}$, $NO3_{Rra}$, $NO3_{Rir}$, $NO3_{RWWL}$, $NO3_{RWL}$, and $NO3_{CSPT}$ denote the monthly amount of nitrate that enters the aquifer through artificial recharge, recharge from rainfall,

irrigation return-flow, leakage of wastewater, leakage from the water distribution network, and leakage from cesspits, in that order. Furthermore, $NO3_{Go}$, $NO3_{Ir}$, $NO3_{DO}$, and $NO3_{DEN}$ indicate the nitrate mass that leaves the aquifer through lateral outflow, water pumped for irrigation, water pumped for domestic purposes, and denitrification, respectively [18, 19].

Health risk assessment

The health risks associated with exposure to nitrate in groundwater resources were assessed by calculating the hazard quotient (HQ) using a modified USEPA health risk assessment model as shown in Eq. 4. To estimate health risk, in this equation stochastic model is integrated with parametric model.

$$HQ = [\Delta S_N + V_{w0}.C_0].IR.EF.ED / [V_{w1}.Bw.AT.RfD] \quad (4)$$

where RfD is a nitrate reference dose which is set to 1.6 mg/kg/day following the Integrated Risk Information System [24], IR is water intake rate (L/day), EF is exposure

Table 1 The parameters used to estimate water volume and nitrate concentration in the aquifer in the study area

Parameter	Value/description
Climate	arid, semi-arid
Land use	Agricultural, desert, residential
Water resources	Agricultural, drinking
Cultivated products	Wheat, corn, grape, potato, alfalfa, tomato, watermelon
Area around wells [km^2]	0.785

frequency (day/year), ED is exposure duration (years), BW is body weight (kg), and AT is averaging time (days/year).

To assess IR, in the study area, 836 precipitants were randomly selected. Then, following the protocol used by Karyab et al. (2016), an expert interviewed the participants regarding the amount of tap water, bottled water, and heated tap water consumed in the last 24 h prior to the interview [25]. Furthermore, based on age-dependent adjustment factors (ADAFs) and physiological differences, exposure parameters were considered in three different age groups of fewer than 2 (infants), 2–16 (children), and older than 16 years old (adults). While an HQ value of *less than* or equal to 1 indicates that adverse effects are not likely to occur, an HQ value of higher than 1 showed a significant risk level. The higher the HQ value attributed to nitrate, the greater the likelihood of adverse non-carcinogenic health impact of nitrate in groundwater [26].

Model validity and data analysis

The validity of estimating nitrate content in the LPM scenario was assessed by the correlation coefficient (R^2), mean errors (ME), average relative error (ARE), percentage standard deviation (PSD), and Root Mean Square Error (RMSE) values [27], which are given in Table 2.

Descriptive statistical analysis was performed using SPSS (version 23). The Kolmogorov-Smirnov test was used to determine the normality of the distribution of the variables. Linear relationship between HQ estimated by monitoring and HQ estimated by the LPM was determined using the Pearson product-moment correlation coefficient. Moreover, the Kruskal-Wallis test was used to determine variation of nitrate concentration in arid and semi-arid climates. Significance was set at 95% ($\alpha = 0.05$).

In addition, to compare health risk maps from monitoring and LPM scenarios, four interpolation models including Kriging, Spline, inverse distance weighted (IDW), and natural neighbor models were run using ArcGIS software, version 10. Interpolation performance in preparing groundwater nitrate map is affected by several factors, including sampling density, sample distribution, and homogeneity or heterogeneity.

Subsequently based on Eqs. 5 and 6, the mean relative error (MRE) and Root Mean Square Error (RMSE) were used to validate the used interpolation models [7, 28].

$$MRE = \frac{1}{n} \sum_{i=1}^n \left| \frac{z^*(x_i) - z(x_i)}{z(x_i)} \right| \quad (5)$$

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n [z^*(x_i) - z(x_i)]^2} \quad (6)$$

Where $z[x_i]$ is the observed value at location i , $z^*(x_i)$ is the interpolated value at location i , and n is the sample size.

Results

Health risk characterization

As presented in Table 3, average nitrate concentration in the monitoring scenario was 27.59 and 18.68 mg/l in semi-arid and arid climates, respectively. On the other hand, average concentration was estimated to be 30.96 and 18.29 mg/l in semi-arid and arid climates in the LPM scenario, in that order.

In the monitoring scenario, 2.7% of the water aliquots (4 wells) had a nitrate pollution of less than 10 mg/L, 40.5% (60 wells) had nitrate levels of between 10 and 20 mg/L, and 52.1% (77 wells) had levels of between 20 and 50 mg/L. Further, the concentration of nitrate in 4.7% of groundwater (7 wells) was higher than the acceptable limit of 50 mg/l [29].

IR was determined using data gathered from the participants concerning their direct consumption of water (including tap, bottled, and boiled water). Average IR was 0.07 ± 0.02 , 0.97 ± 0.15 , and 1.20 ± 0.18 L/day for infants, children, and adults, respectively. The value of BW was 4.5 kg for infants, 30 kg for children and 63.5 kg for adult. Since groundwater was the sole source of potable water for the participants in this study, EF was assigned 365 days per year. ED was considered 1 year for infants, 12 years for children, and 30 years for adults. AT was considered 365 days/year $\times$ 1 year for infants, 365 days/year $\times$ 12 years for children, and 365 days/year $\times$ 30 years for adults [30].

Table 2 Validation analysis and their expressions applied to assess precision of nitrate anticipation by modified LPM

Analysis	Equations	Analysis	Equations
ME	$\sum_{i=1}^n (q^{LPM} - q^{det}) / n$	PSD	$100 \sqrt{\frac{1}{n} \sum_{i=1}^n \left(\frac{q^{LPM} - q^{det}}{q^{LPM}} \right)^2}$
ARE	$\frac{100}{n} \sum_{i=1}^n \left \frac{q^{LPM} - q^{det}}{q^{LPM}} \right $	RMSE	$\sqrt{\frac{\sum_{i=1}^n (q^{LPM} - q^{det})^2}{n}}$
Expressions	q^{LPM} is anticipated nitrate concentration by lump parametric model; q^{det} is determined nitrate concentration in laboratory; n is number of sample		

Table 3 Nitrate concentration in water resources with LPM and monitoring

Climates	Scenario	n	Nitrate concentration (mg/L)				
			Range	mean	Std. Err. of mean	Std. Dev.	95% Conf. Interval
Semi-arid	Monitoring	126	5.40–76.60	24.84	1.12	12.61	22.61–27.07
	LPM		4.40–92.20	27.84	1.53	17.22	24.80–30.87
Arid	Monitoring	22	10.49–43.99	18.68	2.05	9.65	14.41–22.96
	LPM		9.22–52.12	18.29	2.49	11.68	13.11–23.47

As shown in Fig. 2, the estimated HQ using the LPM scenario in the semi-arid climate was 0.17 for infants, 0.64 for children, and 0.25 for adults. These values were estimated to be 0.10, 0.37, and 0.147 in arid climate. Evidently, HQ induced by nitrate in drinking water in the arid climate was lower than in the semi-arid climate. The use of the LPM scenario revealed that 100% of the consumers had HQ values lower than the permissible level of 1, implying that none of the consumers suffered from potential adverse effects of nitrate in drinking water.

Similar HQ was estimated in the monitoring scenario for arid and semi-arid climates. More particularly, in the semi-arid region, it was calculated to be 0.16, 0.57, and 0.23 for infants, children, and adults, respectively. Likewise, the values for these age groups in the arid climate were 0.1, 0.40, and 0.15, in that order. Like the LPM scenario, this shows that there was no threat to human health. It is worth noting at this juncture that although both scenarios yielded estimated HQ values lower than the acceptable level, the HQ estimated by the LPM scenario can lead to a slight to moderate overestimation, which can in turn give rise to errors, especially near the cut-off point of 1. Average difference between the HQ estimated in the LPM scenario and that in the monitoring scenario was $19.39 \pm 19.7\%$ and $12.86 \pm 7.48\%$ in semi-arid and arid climates, respectively. It was also found that the percentage of difference increases with a rise in HQ levels.

Discussion

Risk characterization

By comparison, in the LPM scenario, the corresponding estimated nitrate levels were 4.7% (7 wells), 45.3% (67 wells), and 35.8% (53 wells), respectively. Moreover, in 14.2% of the specimens (21 wells), the nitrate concentration estimated in this latter scenario was higher than the admissible threshold. These values indicate a moderate overestimation of nitrate levels when using the LPM scenario. This can be attributed to uncertainties of parametric models, which draw upon limited information [19].

Nitrate concentration was significantly different in arid and semi-arid climates in both monitoring and LPM scenarios. This is possibly due to different rates of rainfall, land use, and soil characteristics. This finding agrees with several studies which concluded that climate could be a contributing factor in nitrate levels in groundwater. According to the zoning maps of groundwater by Mohammadi et al. (2017), nitrate concentration in the dry season in the water resources of Bandar-e Gaz City, Iran, was greater than rainy seasonal [31]. This finding also supports the theory that more precipitation in semi-arid climates facilitates land change and results in greater nitrate leaching [32, 33].

Fig. 2 Health risks from exposure to groundwater nitrate in different age groups

Table 4 The validity of estimated nitrate contents by modified LPM in different climates

Climates	R ²	Nitrate _{est} equation	ME	ARE	PSD	RMSE
Semi- arid	0.85	1.25 Nitrate _{det} - 3.29	2.99	16.63	19.92	8.12
Arid	0.93	1.16 Nitrate _{det} - 3.47	-0.39	14.29	17.59	3.49

The estimated HQ using the LPM scenario in the semi-arid climate was 0.17 for infants, 0.64 for children, and 0.25 for adults. These values were estimated to be 0.10, 0.37, and 0.147 in arid climate. The obtained results indicated that the highest health risk from exposure to nitrate was observed for children in the semi-arid climate. Similar finding was reported by Yousefi et al. They showed the highest fluoride exposure for different regions was observed in children and teenager's groups [34]. Therefore, it is imperative to reduce nitrate concentration in groundwater using new and effective methods [35–37] to avoid the potential risk to the population.

All groundwater aliquots obtained from the arid climate (7 wells) containing nitrate more than the acceptable level were only identified by the LPM scenario. However, as for the groundwater specimens obtained from the semi-arid climate containing nitrate more than the permissible limit, 7 wells were identified in the monitoring scenario, and 19 wells in the LPM scenario.

The obtained values of ME, ARE, PSD, and RMSE are listed in Table 4. Lower values imply more accurate estimations [27]. The results indicate that climate could affect the validity of LPM outputs, in such a way that the calculated ME, ARE, PSD, and RMSE values in the arid region were lower than in the semi-arid climate.

The results of coefficient of determination in the linear analysis demonstrated very high positive correlation ($R^2 >$

0.9) in both climates [38]. Additionally, the correlation between the nitrate levels obtained via the LPM scenario and the levels observed in the monitoring scenario was higher in the arid climate than in the semi-arid region, suggesting that the modified LPM was more accurate in predicting nitrate concentration in the arid climate.

As is clear in Fig. 3, in both climates, a strong correlation was observed between estimated HQ in the LPM scenario and that in the monitoring scenario. Specifically, in the arid climate ($r = 0.962$, $n = 22$, $p = 0.00$), a linear equation “Estimated HQ by LPM = 1.16 (estimated HQ by monitoring) - 0.027” was obtained. This correlation was weaker in the semi-arid region ($r = 0.917$, $n = 126$, $p = 0.00$).

Health risk mapping

In order To compare accuracy of interpolation models, the obtained MRE and RMSE ranged from 0.01 to 0.21 and from 5.25 to 28.49, respectively. Unlike other studies, which reported that Kriging outperforms other models [39], we found that the natural neighbor was the most appropriate model as it had the least error. Therefore, this model was employed to develop a health risk map. The natural neighbor is a weighted-average model that is used for both interpolation and extrapolation and that can efficiently handle large input points [40]. Figure 4 is the spatial interpolation map of HQ obtained from monitoring and LPM scenarios for adults. This map shows that both scenarios present similar HQ values in most locations.

Uncertainty analysis

Addressing uncertainties in risk analysis is a critical issue when evaluating the impacts of pollutants on public health [41]. Analysis of uncertainty can be carried out using

Fig. 3 Linear relationships between hazard quotient values estimated in monitoring and LPM scenarios

Fig. 4 Distribution of HQ for adults in study area, **a** from monitoring scenario and **b** from LPM scenario

quantitative methods such as sensitivity analysis. This method identifies the sensitive parameters of the system, evaluates how changing inputs of the LPM scenario can influence the results, and allows us to identify the key parameters with the greatest potential to increase human health risks [42]. Like the previous research, the present study found that risk assessment involves some inevitable uncertainties, including errors in water sampling and nitrate measurement, distribution of nitrate content over time, and the assignment of HQ to drinking water only [43]. In addition, because of the differences identified

between the two scenarios, groundwater nitrate estimated by the LPM scenario was regarded as an uncertainty in the estimation of human health risks.

Sensitivity analysis was also used for analyzing the input and output parameters in the LPM scenario that affect nitrate concentration. It was found that nitrate estimation in the LPM was sensitive to nine variables, which were involved in the source and sink of groundwater nitrate. Analysis of variables affecting nitrate content in water bodies showed that the share of nitrate sources in both climates was as follows: $R_{CSpt} >$

$R_{wwl} > R_{wl} > R_{ir}$. Additionally, Q_{D0} had the greatest impact on nitrate sinks (Table 5).

Sensitivity analysis shows that in the LPM scenario, nitrate recharge from cesspits had the greatest potential to pollute groundwater, thereby increasing HQ attributed to groundwater nitrate. Additionally, the largest reduction in groundwater nitrate was due to pumping water for domestic purposes. The main limitation of this analysis is that interactions between variables were not considered. This warrants further research into estimation of health risks that encompasses all potentially significant variables in the LPM.

Conclusion

This study set out to estimate health risks from nitrate in drinking water in arid and semi-arid climates. For this purpose, two scenarios were established, one for determination of nitrate concentration via water monitoring and the other using an LPM. The results demonstrate the influence of climate on the nitrate content of groundwater. The highest health risk from exposure to groundwater nitrate was observed for children in the semi-arid climate. Due to land change, abundance of cesspits, and agricultural activities, the HQ induced by nitrate in the semi-arid climate was higher than in the arid climate. Both scenarios presented an HQ value of lower than the permissible level, implying no adverse health effect. Although in none of the wells under investigation the HQ estimated by the LPM scenario was higher than the acceptable level, this scenario gives rise to a slight to moderate overestimation, which can cause errors near the cut-off point of 1.

The validation of the modified LPM showed that climate could impact upon outputs in such a way that the calculated validity indices in the arid region were lower than in the semi-arid climate. Linear analysis revealed a strong correlation between the two scenarios in estimation of HQ attributed to nitrate in groundwater. However, correlation in the arid area was higher than in the semi-arid region, indicating that the modified LPM is more accurate in predicting nitrate concentration and the concomitant health effects in the arid climate. Sensitivity analysis showed that wastewater leakage from cesspits contributed the most to health risks associated with nitrate in groundwater.

Table 5 Share of sources and sinks of nitrate from the model domain by LPM scenario

Climates	Share of source %						Share of sink %		
	R_{ra}	R_{ir}	R_{wwl}	R_{wl}	R_{cspt}	Surp	Q_{irr}	Q_{D0}	Q_{DEN}
Semi- arid	1E-5	1.81	28.33	1.91	67.93	1E-2	0.46	99.48	6E-2
arid	3E-5	1.82	27.70	3.67	66.79	2E-2	0.85	99.04	0.11

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Compliance with ethical standards

Conflict of interest There are no conflicts of interest in this manuscript.

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