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Nitrate health risk assessment and its spatial distribution in drinking water in Arak, Iran

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ABSTRACT

Potable water pollution with nitrate (NO_3^-) is a global concern and related to human health outcomes. In the present study, the health risk assessment of nitrate in drinking water resources of Arak, Iran, was implemented through ordinary kriging (OK) and empirical Bayesian kriging (EBK) models. Water samples were collected from 61 locations and analyzed for nitrate and other water quality parameters during the two statistical periods of 2011 and 2018. Health risks of nitrate were estimated by using the hazard index (HI) for children, males and females and interpolation models which include OK and EBK were used to the expansion of nitrate pollution. The mean concentrations of nitrate were varied from 4.5 to 56 mg/L, with a mean of 29.5 mg/L. furthermore, the means HI for children, adult males, and adult females were 1.12 (0.43 to 1.87), 0.78 (ranged from 0.2 to 1.47) and 0.86 (0.25 to 1.35), respectively. The spatial distribution of nitrate concentrations towards the central and southeast part of the study area was the highest so that residents in these regions were at the highest health risk and children were more exposed. Subsequently, there is necessary for applying effective strategies to protect drinking water quality and to better manage and control nitrate pollution sources.

Keywords: Chemical parameters; Drinking water quality; Cross-validation; Nitrate

1. Introduction

Nitrate is a widespread pollutant in drinking water [1]. Common anthropogenic sources of nitrate include septic systems and wastewater discharges from sewage treatment plants, fertilizers used for agricultural production and landscaping, animal manure, fossil fuel combustion, and human waste [1,2]. Based on epidemiologic and animal studies, some health effects such as infant methemoglobinemia ("blue baby syndrome"), gastric cancer, stomach and esophagus cancers, spontaneous abortion, birth malformations, goiter, and hypertension are related to high concentration of nitrate in drinking water [3]. Additionally, recent epidemiological studies have found associations between nitrate concentrations in drinking water and bladder cancer [4,5], colon cancer [6], kidney cancer [7], birth defects [8], preterm birth [9], and thyroid dysfunction [10], although the Agency for Toxic Substances and Disease Registry (ATSDR) concluded that there is "limited evidence" for nitrate-induced cancer [11]. Finally, International Agency for Research on Cancer (IARC) classified "ingested nitrate or nitrite under conditions that result in endogenous nitrosation" as a probable human carcinogen (Group 2A) [12]. Hence, the regulatory limit 50 mg/L as NO_3^- and 10 mg/L as NO_3^-–N of nitrate in drinking water is set by the World Health Organization (WHO) and Environmental Protection Agency (EPA) to protect human health [13,14]. Similarly, the Institute of Standard and Industrial Research of Iran (ISIRI) sets the similar standard limit [15].

Due to the increase of anthropogenic nitrogen inputs, high nitrate concentrations in water resources have been widely reported in various parts of the world [1,16,17]. Rivett et al. [16] reported that the average concentration of nitrate was exceeded than 25 mg/L in >50% of water resources in Spain, United Kingdom, Germany, France and Italy. In drinking water resources in North China [17] and 21 rivers of India [1], the concentration of nitrate reported to be slightly higher than 50 mg/L as NO_3^- [1,17]. In parallel to other countries, the high concentration of nitrate in drinking water resources has been reported in Iran [18]. For example in Hamadan, the nitrate concentration over 5 years elevated roughly twofold (from 24 to 43 mg/L) and in some areas, it was eightfold (from 24 to 195 mg/L) [19]. The same high concentrations of nitrate were reported in some other areas of Iran such as Mashhad, Isfahan, and Tehran [18]. Thus, drinking water resources contaminated with nitrate led to reduce the quality of water and subsequently associated with health risk [20]. To accurate health risk assessment of nitrate, it is necessary to collect numerous samples from different space and time [21]. Since collecting many samples is costing and time-consuming, the geostatistical interpolation methods such as ordinary kriging (OK) and empirical Bayesian kriging (EBK) are employed to estimate the nitrate concentration. The purpose of this study was to determine the health risk assessment of nitrate in drinking water resources used in Arak, Iran, through the ordinary kriging and empirical Bayesian kriging methods.

2. Methods

2.1. Study area

A study area map with the sampling locations is drawn in Fig. 1. The source of drinking water in most cases is supplied from groundwater. According to the latest statistics, there are about 600,000 people in the 98.8 km^2 of the study area. The study region is geographically located in cold semi-arid region in the western part of Iran and has mainly cold and dry climate. The mean monthly temperature changes from –4°C to 40°C during January to December. In the study region, the annual rainfall ranged from 160 to 550 mm.

2.2. Water sampling and analysis

The water samples were carried out in the months of October and November in 2011 and October and November in 2018. Drinking water samples were collected from 61 locations in the Arak as an industrial megacity of Iran and analyzed for nitrate and other water quality parameters during the two statistical periods of 2011 and 2018 (Fig. 1). Drinking water samples were collected in 1-L prewashed high quality polyethylene bottles and transported to the laboratory and analyzed for 13 parameters, specifically pH, EC, TDS, cations (Ca^{2+}, Mg^{2+}, Na^+ and K^+) and anions (HCO_3^-, SO_4^{2-}, Cl^-, F^-, NO_2^-, and NO_3^-). The pH and electrical conductivity (EC) were measured in situ at 25°C using a precision pH and EC meter HACH HQ40D portable (Loveland, Colorado, USA). Then the samples were delivered within 48 h to the laboratory under refrigerated conditions (4°C in cold box). All samples were analyzed following the Standard Methods suggested by the American Public Health Association [22]. Nitrate was measured in mg/L using a spectrophotometer [22]. Sodium (Na^+) and potassium (K^+) were analyzed by flame photometer; nitrate (NO_3^-), sulfate (SO_4^{2-}), and fluoride (F^-) by spectrophotometry (DR5000); and calcium (Ca^{2+}), magnesium (Mg^{2+}), bicarbonate (HCO_3^-), and chloride (Cl^-) by volumetric method and total dissolved solid (TDS) was calculated by summing up all major ions. Moreover, Piper diagram was used to classify the dominant type of water resources.

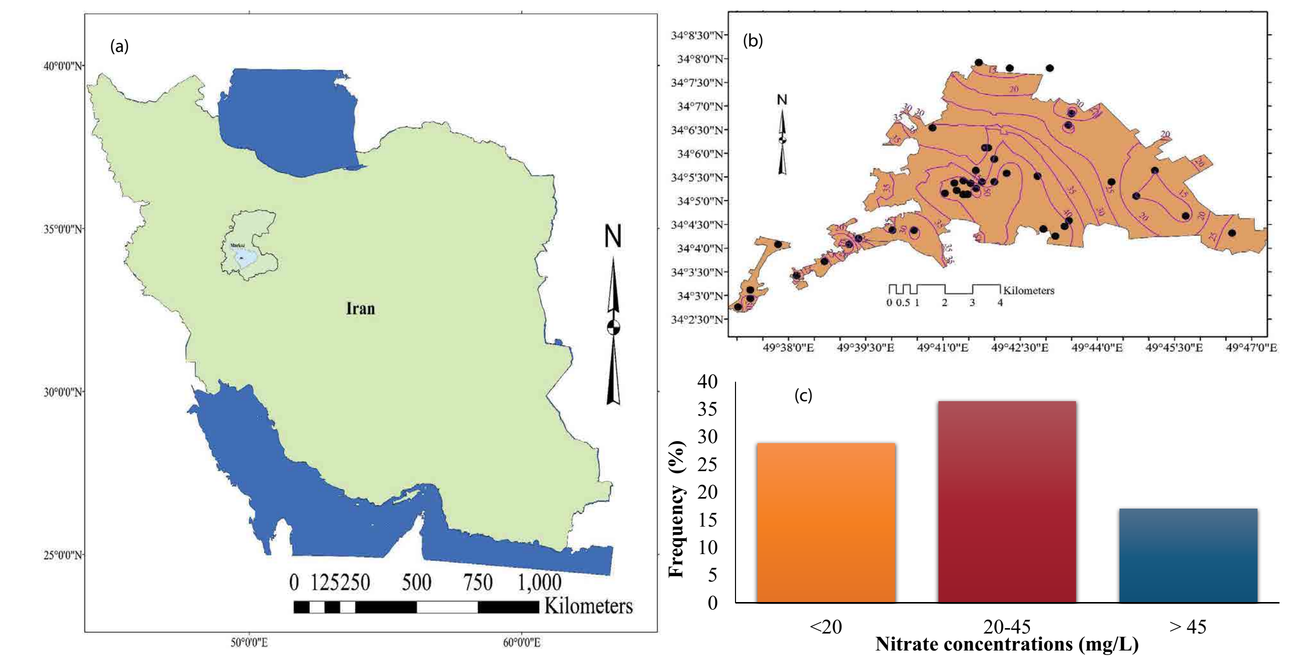

Fig. 1. (a) Geographical study area, (b) sampling locations of drinking water resources with NO_3^- isolines over the study area, and (c) frequency distribution of nitrate concentrations.

2.3. Health risk assessment

The total hazard index (HI_{total}) which represents the cumulative non-carcinogenic risk is estimated by summing up hazard quotients (HQ_{oral} and HQ_{dermal}) and are computed by Eqs. (1) and (2):

$$HI_i = HQ_{oral} + HQ_{dermal} \quad (1)$$

$$HI_{total} = \sum_{i=1}^{n} HI_i \quad (2)$$

Oral and dermal hazard quotient for the nitrate health risk assessment was calculated through non-carcinogens health risk model (US.EPA) by the following expressions:

$$HQ_{oral} = \frac{CDI}{RfD} \quad (3)$$

$$HQ_{Dermal} = \frac{DAD}{RfD} \quad (4)$$

where HQ_{oral} and HQ_{dermal} are the non-carcinogenic for oral and dermal hazard quotient, respectively. CDI and DAD indicate chronic daily intake (mg/kg d) and the dermally absorbed dose (mg/kg d), respectively, and RfD represents the reference dose of a specific contaminant. The oral reference doses of nitrate-nitrogen (1.6 mg/kg/d) were obtained from the database of Integrated Risk Information System (US EPA). Non-carcinogenic risk through drinking water pathway in terms of CDI is computed by Eq. (5) [23,24]:

$$CDI = \frac{CPW \times IR \times ED \times EF}{ABW \times AET} \quad (5)$$

where CDI is the chronic daily intake (mg/kg day); CPW is the concentration of a particular contaminant in groundwater (mg/L); IR is the human ingestion rate (2.5 L/d for adults, and 0.78 L/d for children); ED is the exposure duration (64, 67, and 12 y for men, women, and children, respectively); EF is the exposure frequency (365 d for children and adults); ABW is the average body weight (65, 55, and 15 kg for men, women, and children, respectively), and AET is the average time (23,360; 24,455; and 4,380 d for men, women, and children, respectively). Dermal contact pathway was estimated by using the following equation:

$$DAD = \frac{TC \times IR \times ED \times EF \times SSA \times CF}{ABW \times AET} \quad (6)$$

where DAD is the dermally absorbed dose (mg/kg d); TC indicates the contact duration (h/d: 0.4 h/d for adults and children); Ki is the dermal adsorption parameters (0.001 cm/h); EV is the bathing frequency (considered as 1 time/d); SSA is the skin surface area available for contact (16,600 and 12,000 cm^2 for adults and children, respectively); CF is the unit conversion factors (0.001); ED is the exposure duration (64, 67, and 12 y for men, women, and children, respectively); EF is the exposure frequency (365 d for children and adults); ABW is the average body weight (65, 55, and 15 kg for men, women, and children respectively), and AET is the average time (23,360; 24,455; and 4,380 d for males, women, and children, respectively). Based on the HI_{total} values, no significant risk of non-carcinogenic effects are anticipated if the value is less than one ($HI_{total} < 1$). However, in the case of HI_{total} value that exceeds one ($HI_{total} > 1$), residents are exposed to non-carcinogenic risk [23,24].

2.4. Geostatistical analysis

Interpolation methods, including ordinary kriging (OK) and empirical Bayesian kriging (EBK), were used to estimate the concentrations of nitrate, using ArcMap 10.3 (ArcGIS, ESRI, Redlands, CA, USA). Before the application of interpolation methods, the normality of the data was investigated through the Shapiro–Wilk test. Ordinary kriging used to estimate the values of a spatial variation of nitrate at unsampled locations. Semi-variogram of samples is defined as the expected squared difference between pairs of data by increased distances as given by the following expression:

$$\lambda(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} \left[Z(Xi) - Z(Xi + h) \right]^2 \quad (7)$$

where λ(h) is the semi-variance of the sampling sites separated by a distance h; Z(xi) are the sampling values at points xi with data in xi and xi + h; $N(h)$ is the number of paired data separated by a h distance. In the EBK, parameters are automatically adjusted during the modeling process. Consequently, the uncertainty in semi-variogram estimation will be diminished and the standard error will be reduced.

2.3. Error measures

The accuracy of nitrate risk estimates was assessed using error measurements, including root mean square error (RMSE), mean standardized error (MSE), root mean square standardized error (RMSSE), and average standard error (ASE). These error measurements are defined as follows:

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} e_i^2} \quad (8)$$

$$MSE = \frac{1}{n} \sum_{i=1}^{n} \frac{e_i}{s_i} \quad (9)$$

$$RMSSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} \left(\frac{e_i}{s_i} \right)^2} \quad (10)$$

$$ASE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} s_i^2} \quad (11)$$

where e_i and s_i^2 are error and variance of an estimate at distance x_i, respectively. Correlation coefficient (R^2) was also calculated by establishing a regression between observed and estimated values of nitrate.

3. Results and discussion

3.1. Drinking water types

The Piper trilinear diagram is drawn in order to classify the drinking water based on chemical characters (Fig. 2). On the basis of Piper diagram, drinking water in the study area is classified mainly into two types: calcium–sodium–bicarbonate (Ca^{2+}–Na^{+}–HCO_3^-) and calcium–magnesium–chloride–sulfate types (Ca^{2+}–Mg^{2+}–Cl^-–SO_4^{2-}).

3.2. Drinking water quality

The statistical distribution (minimum, maximum, mean, and standard deviation) of various physico-chemical parameters of 61 drinking water samples are shown in Fig. 3. The pH value of water in the study area differs between 6.5 and 8.4, with an average of 7.40, and only few samples have values less than 7, which shows a slightly alkaline environment. Electrical conductivity (EC) was in the range of 170–1,881 µS/cm and TDS in the range of 195–967.5 mg/L, with an average of 474 mg/L (Table 1).

The Shapiro–Wilk test of normality indicated that the NO_3^- was not normally distributed (p = 0.09), with a little skewness. The frequency distribution of nitrate concentration is presented in Fig. 1c. It was appeared that 29% of the nitrate concentration had values below 20 mg/L, 36.5% were between 20 and 45 mg/L and 17% of the samples exceeded from MCL (maximum acceptable limit) of 45 mg/L. The median and standard error for nitrate in water samples during the 2-year period was 27.5 ± 1.6 and 29.5 ± 1.6 mg/L, respectively, which suggested that water samples from the study area have high pollution. Spatial distribution of nitrate shown an increase in the average nitrate concentration in central and southern part of Arak, which shifted from 28.5 ± 13.5 mg/L in 2011 to 30.7 ± 12.8 mg/L in 2018 (Fig. S1). A summary of the accuracy metrics of NO_3^- concentration from OK and EBK models is presented in Table 2. For both models, mean error (MSE) ranged from –0.02 to 0.05.

3.3. Health risk of nitrate

The spatial distributions of nitrate hazard by OK and EBK interpolation maps for adults in 2011 and 2018 are presented in Figs. 4a–c, respectively. The mean hazard index for children, adult females, and adult males were 1.12 (0.43 to 1.87), 0.86 (0.25 to 1.35), and 0.78 (ranged from 0.2 to 1.47), respectively (Table 3). Over 15% samples were higher than the acceptable limit for noncarcinogenic risk (HI > 1). The mean hazard index of nitrate has been increased in 2018 compared with 2011 (2018 vs. 2011: concentration of nitrate 28.5 vs. 30.7 mg/L; HI, 0.7 vs. 0.85).

4. Discussion

The concentration of TDS with the highest value of 1,881 mg/L in the north part of Arak, reflecting the content of drinking water TDS in the study area is still good for consumers. As per the WHO guideline, the maximum permissible limit (MPL) of TDS is 1,500 mg/L for drinking

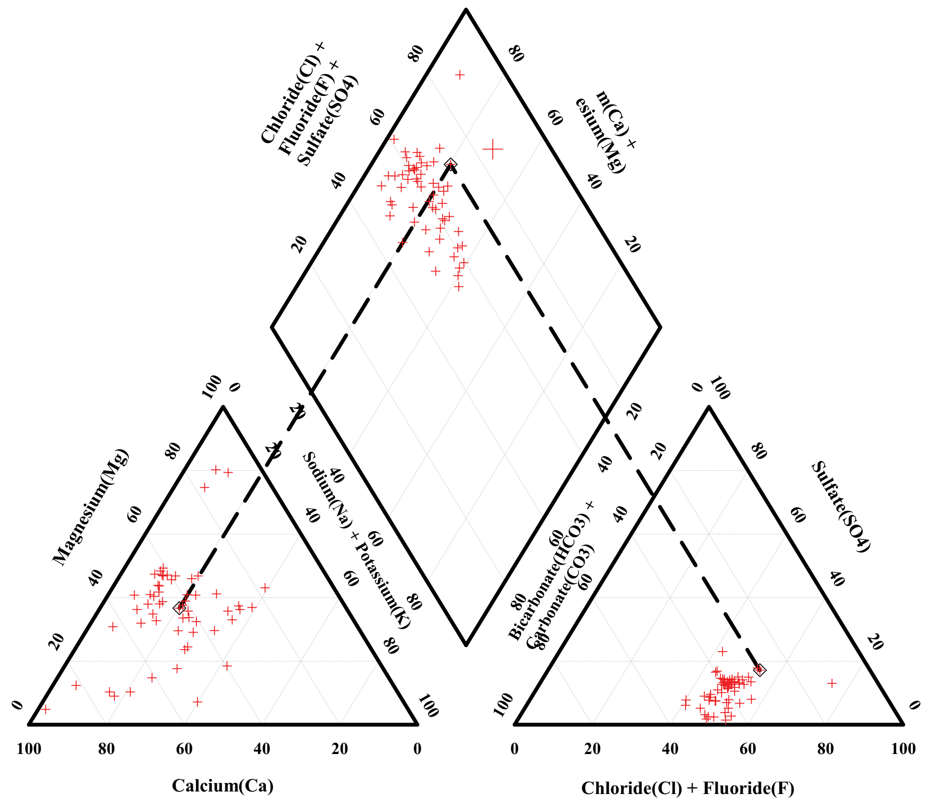

Fig. 2. Piper diagrams for the drinking water major cations and anions.

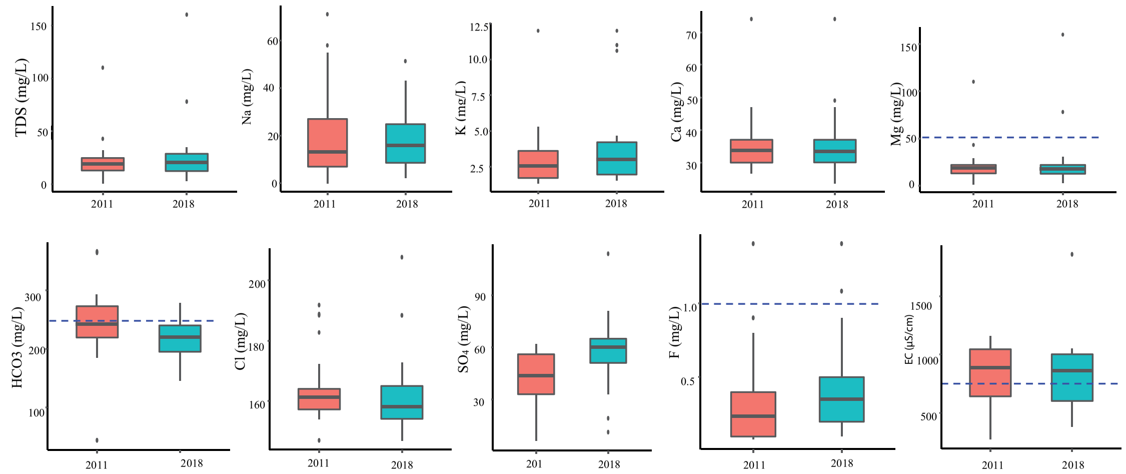

Fig. 3. Box plots of the main chemical parameters of drinking water in years 2011 and 2018.

Table 1
Summary statistics for drinking water samples of study area exceeding the maximum permissible limits (MPLs) prescribed by WHO 2006 in Arak, Iran

Water Quality parameters	Units	2011					2018					WHO guideline	
		Mean	SD	Max	Min	Percentage of samples exceeding (MPL)	Mean	SD	Max	Min	Percentage of samples exceeding MPL	Highest desirable limit (HDL	Maximum permissible limit (MPL)
EC	µS/cm	931.3	86.0	1,860.0	172.0	8	952.8	107.5	1,881.5	193.5	6	750	1,500
TDS	mg/L	464.2	22.0	931.0	195.0	0	499.4	57.2	966.2	230.2	0	500	1,500
pH	–	7.3	0.1	8.5	7.1	0	7.4	0.1	8.6	7.0	2	7	8.5
TH	mg/L	349.6	5.5	548.0	36.0	15	391.6	47.5	590.0	78.0	18	100	500.0
TA	mg/L	191.8	56.0	300.0	120.0	12	203.8	68.0	312.0	132.0	13	250	
Ca^{2+}	mg/L	92.9	8.4	154.0	53.0	0	101.1	16.6	162.2	61.2	0	75	200
Mg^{2+}	mg/L	30.0	2.5	42.0	17.0	0	32.5	5.0	44.5	19.5	0	30	150
Na^{+}	mg/L	18.5	15.2	85	0	0	20.5	16.5	91	0	0	–	200
K^{+}	mg/L	3.6	2.85	12	1.3	0	4.5	2.91	15	1.1	–	–	–
HCO_3^-	mg/L	230.4	3.0	366.0	44.0	14	240.9	13.5	376.5	54.5	16	–	250.0
Cl^-	mg/L	163.5	12.5	210	146	0	172.5	13.2	215	140	0	200	600
SO_4^-	mg/L	48.7	1.5	114.0	6.0	0	52.2	5.0	117.5	9.5	0	200	400
F^-	mg/L	0.2	0.1	0.4	0.0	0	0.3	0.1	0.5	0.0	0	1	1.5
NO_2	mg/L	0.032	0.01	1	0		0.035	0.01	1	0			3
NO_3^-	mg/L	28.5	13.5	53.5	5	10	30.7	12.8	62.0	4.5	12	–	50

Table 2
Cross validation error measures of nitrate estimates

Error measure	OK				Co-kriging (2011 + 2013)	EBK
	Spherical	Exponential	Gaussian	Linear	Exponential	–
RMSE	6.3	5.96	6.53	6.37	8.31	5.15
MSE	–0.02	–0.01	–0.02	–0.02	0	0.05
RMSSE	0.65	0.6	0.86	0.66	1.05	1
ASE	9.67	10.13	7.64	9.59	8.01	7.17
R^2	0.56	0.75	0.62	0.58	0.6	0.7

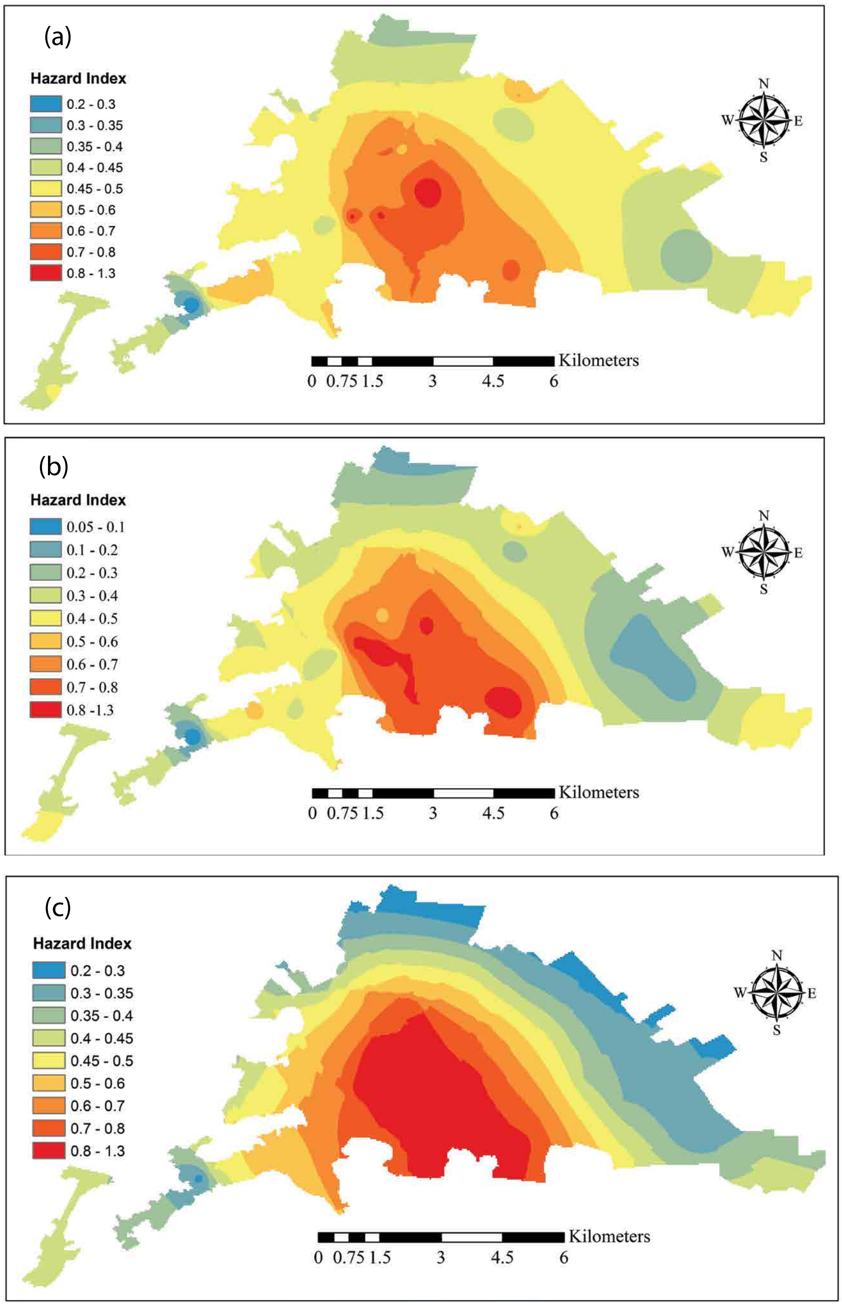

Fig. 4. Spatial variability of nitrate HI in the study area by OK in the 2011 (a), 2018 (b), EBK in the 2011 (c), and 2018 (d).

Table 3
Assessment results of health risks through oral/drinking water intake for adults and children in the Arak, Iran

NO	Longitude	Latitude	NO_3–N (mg/L), 2011	HI, 2011			NO_3–N (mg/L), 2018	HI, 2018		
								Children	Men	Women
1	49.62	34.05	26.5	0.77	0.83	0.96	24	1.28	1.17	0.90
2	49.62	34.05	28.25	1.52	1.09	0.43	26	1.52	0.25	0.27
3	49.63	34.07	25.5	1.19	1.18	0.29	24	1.28	0.36	0.73
4	49.62	34.05	25	1.19	1.47	1.01	24	1.15	0.51	0.58
5	49.64	34.06	21	1.12	0.29	0.75	22	0.64	0.96	1.31
6	49.65	34.06	24.5	1.48	1.51	0.25	22	1.28	0.53	1.11
7	49.65	34.07	4.5	0.67	0.37	0.77	0	1.09	0.33	0.39
8	49.66	34.07	28.5	1.23	0.84	1.19	35	1.76	0.81	0.93
9	49.67	34.07	32.75	0.61	0.68	0.23	35	1.56	0.65	0.54
10	49.67	34.07	25.5	1.15	0.95	1.09	28.5	1.15	1.16	0.27
11	49.68	34.09	23.5	1.74	1.19	0.66	23	0.67	1.11	1.17
12	49.69	34.09	53.7	1.30	0.75	1.03	46.85	1.56	0.31	0.29
13	49.69	34.09	38.5	1.07	1.16	0.56	45.25	0.82	0.89	1.03
14	49.69	34.09	40.5	0.59	1.19	0.86	48	1.63	1.17	0.46
15	49.69	34.09	40	0.67	0.75	0.98	46.5	1.28	1.26	0.31
16	49.69	34.09	40	0.53	0.38	0.25	46.85	1.27	1.57	1.08
17	49.69	34.09	53	1.12	0.84	1.10	53	1.20	0.31	0.80
18	49.69	34.09	40	1.53	0.71	0.49	46.5	1.59	1.62	0.27
19	49.72	34.11	17	0.54	1.04	0.84	19	0.72	0.40	0.82
20	49.71	34.13	12.75	0.72	0.44	1.12	17	1.32	0.90	1.27
21	49.75	34.09	15	1.27	0.40	0.77	25	1.40	1.22	0.27
22	49.75	34.09	14.75	0.75	1.04	0.81	26.5	0.47	1.34	1.21
23	49.74	34.09	22.25	0.49	0.51	0.75	26.5	0.79	1.28	0.52
24	49.70	34.09	44.5	1.08	1.08	0.27	44	1.02	0.81	0.92
25	49.73	34.11	32.5	0.89	1.13	1.04	38.5	0.49	0.45	0.66
26	49.70	34.13	12	1.53	0.87	0.72	14	0.65	0.73	0.25
27	49.72	34.13	13.5	1.64	1.07	1.05	15	1.23	1.02	1.17
28	49.73	34.11	15.25	0.63	0.26	0.55	16.5	1.86	1.27	0.71
29	49.68	34.11	20.75	0.48	0.49	1.30	25	1.39	0.80	1.10
30	49.70	34.10	45	1.05	0.56	0.44	39.5	1.15	1.24	0.60
31	49.78	34.07	27.5	0.58	0.27	1.30	25	0.63	1.28	0.92
32	49.76	34.08	13	1.59	0.39	1.13	14	0.72	0.80	1.05

(Continued)

Table 3 *Continued*

NO	Longitude	Latitude	NO_3–N (mg/L), 2011	HI, 2011			NO_3–N (mg/L), 2018	HI, 2018		
								Children	Men	Women
34	49.69	34.09	42.5	0.80	1.10	0.55	35.5	1.20	0.90	1.18
35	49.70	34.09	56.5	1.08	0.37	0.42	49.25	1.64	0.76	0.53
36	49.70	34.10	41.5	0.65	0.44	1.12	40.5	0.51	1.26	0.85
37	49.70	34.10	35.5	0.99	0.34	0.65	37.85	0.58	1.11	0.90
38	49.71	34.09	41.5	1.34	1.21	1.08	40.5	0.77	0.47	1.20
39	49.70	34.09	41.5	1.64	1.10	0.55	40.5	1.36	0.43	0.82
40	49.69	34.09	39.25	1.51	1.58	0.62	34.75	0.80	1.11	0.87
41	49.70	34.09	43.75	1.64	0.78	0.48	39.25	0.53	0.55	0.80
42	49.70	34.10	29.75	0.48	1.13	0.58	35.25	1.16	1.16	0.29
43	49.70	34.10	34.75	1.19	1.25	0.66	40.25	0.95	1.21	1.11
44	49.71	34.09	38.75	1.31	1.14	0.25	39.25	1.62	1.69	0.66
45	49.70	34.09	45.75	0.44	1.25	1.13	41.25	1.76	0.84	0.51
46	49.69	34.09	45.75	0.74	1.19	0.49	41.25	0.51	1.21	0.62
47	49.72	34.11	15.25	0.95	0.76	0.86	19.75	1.27	1.34	0.71
48	49.71	34.13	5	0.46	0.42	0.62	10	1.64	0.93	0.77
49	49.75	34.09	24.25	1.19	1.09	0.84	18.75	1.76	1.15	1.12
50	49.75	34.09	14.25	1.42	0.23	0.25	8.75	0.68	0.28	0.59
51	49.74	34.09	15.25	1.19	0.34	0.68	9.75	0.51	0.53	1.39
52	49.70	34.09	34.75	1.07	0.48	0.54	37.25	1.13	0.60	0.47
53	49.73	34.11	16.25	0.60	0.90	1.22	10.75	0.62	0.29	1.39
54	49.70	34.13	21.35	1.19	0.49	1.04	15.85	1.70	0.42	1.21
55	49.72	34.13	14.75	1.02	0.31	0.36	9.25	1.86	0.98	0.95
56	49.73	34.11	15.25	1.64	0.76	0.87	9.75	0.86	1.18	0.59
57	49.68	34.11	14.25	1.46	0.61	0.50	8.75	1.16	0.40	0.45
58	49.72	34.07	44.75	1.07	1.08	0.25	52.75	0.70	0.47	1.20
59	49.72	34.08	44.75	0.63	1.04	1.09	48.75	1.06	0.36	0.70
60	49.72	34.07	42.25	1.46	0.29	0.27	43.75	1.44	1.30	1.16
61	49.72	34.07	39.5	0.48	1.18	0.79	46.75	1.76	1.18	0.59
Mean ± SD	-	-	28.57 ± 12.8	1.05 ± 0.39	0.81 ± 0.36	0.73 ± 0.31	30.7 ± 13.5	1.13 ± 0.42	0.87 ± 0.39	0.78 ± 0.33

purpose. Among cations, the concentration of Na^+, K^+, Ca^{2+}, and Mg^{2+} ranged from 2 to 91, 1.1 to 15, 53 to 162 and 17 to 44.5 mg/L, respectively; and among anions, Cl^-, SO_4^{2-}, HCO_3^-, NO_2^-, and F^- concentrations were in the range of 140–215, 6–117.5, 44–376.5, 0–1, and 0–0.5 mg/L, respectively. The agricultural fertilizers are the main source for Na^+ and K^+ in water [23]. Chloride in drinking water may originate from both natural and anthropogenic sources [23]. Bicarbonate and sulfates are originated from the contamination by the domestic wastewaters. The concentration of NO_2^- varies between 0 and 1 mg/L with an average value of 0.03 mg/L, could be associated with the nitrification process due to existing oxic conditions in water. The spatial variation of NO_3^- pollution in drinking water sources could be mainly originated from point sources such as sewage system and septic tanks and nonpoint sources such as chemical fertilizers [23,25]. Subsequently, raised nitrate concentration in the drinking water in the study area is mainly derived from the anthropogenic sources such as domestic sewage, leakage from septic tanks and agriculture [23,26].

The cross-validation for the OK and EBK was compared to determine the best model. The measured error in EBK models were lower than OK models. For both models, mean error (MSE) ranged from –0.02 to 0.05 (Table 2). The smallest difference between RMSE (5.15) and average standard error (ASE) (7.17) linked with EBK model and this model was considered the better model to predict nitrate concentration in the study area (Table 2). The computed hazard index in 2011 and 2018 elevated from 8.2% to 18.1%, which indicated increasing hazard index over the time. The problem of NO_3^- pollution in the drinking water is not only spread all over in Iran but also noticed worldwide [26–29]. Similar noncarcinogenic risks of NO_3^- in drinking water were reported in other studies [23,27,30,31]. The mean hazard index of NO_3^- in this study was higher than those hazard index reported by studies in other countries such as in Iowa, the United States [5,32], South India [23], while they were to some extent lower in some countries such as Saudi Arabia [33], India [34], UK [35], North America [36], Australia [37], and Changshu in China [38]. Similarly, the mean concentrations of nitrate in most large cities of Iranian cities were higher such as Mashhad, Zanjan, Kermanshah, Hamedan, Isfahan, Kerman, and Yazd and in some other cities were lower such as Kerman and Golestan [18].

5. Conclusions

This is the first comprehensive study that evaluates the NO_3^- contamination in drinking water in Arak, Iran. The main conclusions were drawn as follows:

- In order to estimate the nitrate spatial probability in drinking water, EBK model is robust and could be applied to health risk assessment studies.
- In some parts of the central and southern cities, the concentration of nitrate and hazard index was slightly high.
- It was found that children exposed at current nitrate concentrations are at a higher risk than adult females and then adult males. Moreover, hazard index of nitrate exposure in 2018 was higher than in 2011.

The anthropogenic sources, including sewerage, organic garbage, and nitrogenous fertilizers are the main source of NO_3^- in drinking water of this area. Subsequently, it is necessary to apply effective strategies to control and protect nitrate pollution sources in the study area.

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Supplementary information:

Supplementary data to this article can be found in Fig. S1.

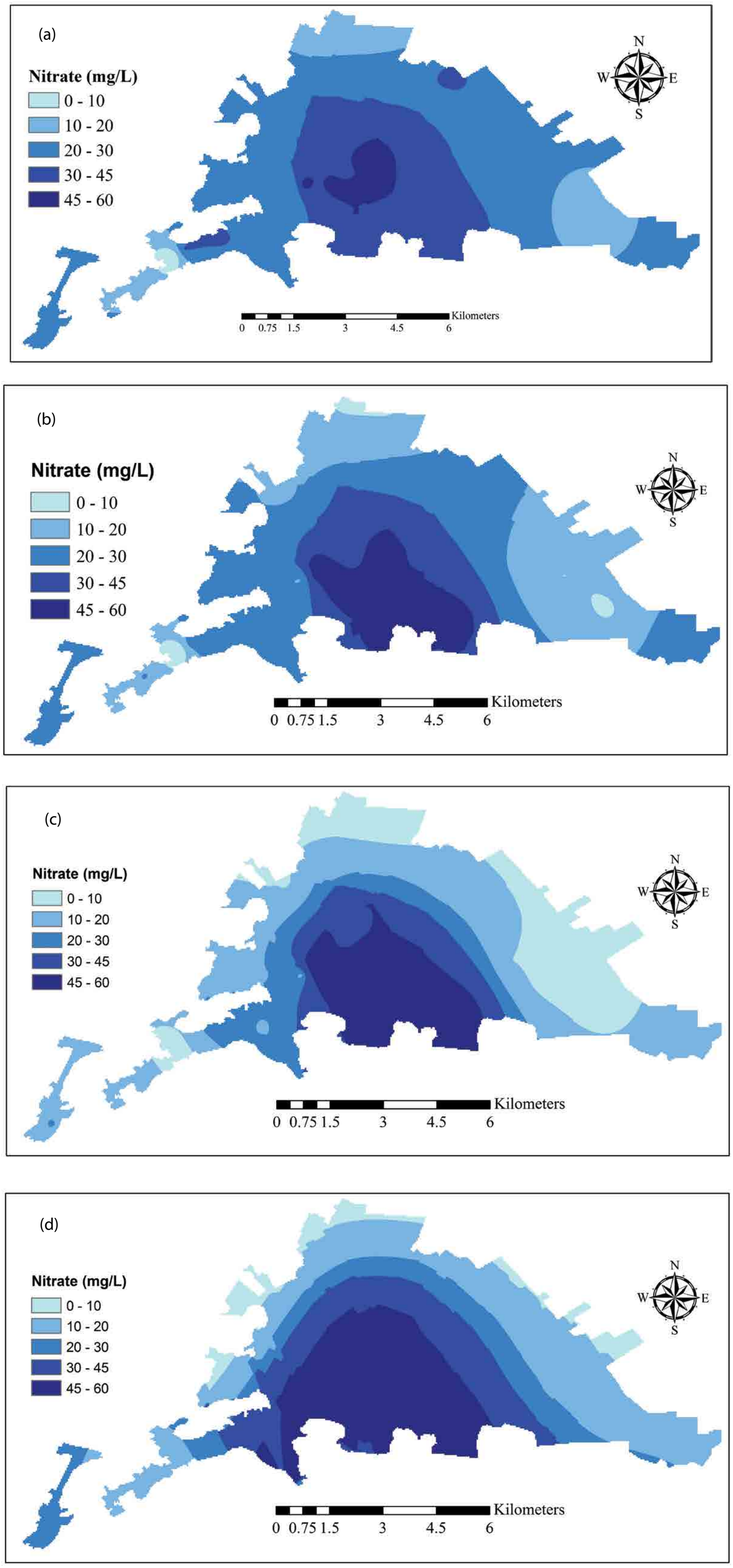

Fig. S1. Spatial distributions of nitrate in Arak by OK in the 2011 (a), 2018 (b), EBK in the 2011 (c), and 2018 (d).