Original Article

Examining Changes Trend of Fluoride Concentration in Groundwater Using Geo-Statistical Technique Case Study: Drinking Water wells in Yazd-Ardakan Plain

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Abstract

Introduction: Studies have demonstrated that the presence of standard amount of fluoride in drinking water can prevent tooth decay. Meanwhile, higher concentration than permitted amount in drinking water causes bone disease and dental fluorosis. Given the importance of fluoride in drinking water as well as GIS ability in spatial analysis of various factors in the groundwater, this study aimed to evaluate concentration of fluoride in the plain of Yazd-Ardakan using two IDW (Inverse Distance Weighting) and Kriging models.

Materials and Methods: In this descriptive study, the applied data on the water quality of underground water of Yazd Regional Water Company and the average annual fluoride in 2014 were related to 76 rural wells and 107 urban wells in the studied area. The fluoride rate in groundwater in rural and urban areas was compared to the standard amount of Institute of Standard and Industrial Researches and then was analyzed via Arc GIS software using IDW and Kriging interpolation methods.

Results: The mean concentration of fluoride was 0.68 mg/l in rural areas and 0.7 mg/l in urban areas. With regard to the minimum and maximum concentrations of fluoride as well as standard deviation of 0.364, fluoride concentration fluctuations in rural areas were reported higher than urban areas, For both rural and urban areas, Kriging interpolation method was more efficient than IDW method.

Conclusion: The study findings revealed shortage of fluoride in drinking water and the risk of tooth decay within the residents of southern districts of Yazd-Ardekan which necessary management needs to be taken in regard with these areas. It is worth mentioning that Geo-statistical methods can greatly assist in understanding the environmental issues.

Keywords: Drinking water; Fluoride concentration; Geographic information systems; Interpolation

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Introduction

Fluorine is regarded as a halogen being yellow in its gas form. It is a strong oxidizing element so that itcan react explosively with hydrogen gas in cold and dark conditions ^[1]. Fluoride can be effective in preventing tooth decay, that is why it is added to toothpaste and drinking water ^[2]. The conducted studies have proved the effectiveness of this element in prevention and control of tooth decay ^[3, 4]. Fluoride accumulates in bones and teeth of the growing children that contributes to the strength of teeth enamel before their emergence of decay. It also aids with

hardening the teeth enamel within the adults ^[5]. Despite the benefits and the necessity of its existence in body, exposure to high concentration of fluorine can be very are required to be monitored based on a regular schedule. Drinking water can be obtained through municipal water systems, private wells, water reservoirs, springs, and bottled water. The fluoride rate in such sources might be remarkably different depending on the source, season, and treatment level. For instance, an inverse relationship can be observed between the concentration of the ion and temperature so that according to World Health Organization (WHO) standard, the maximum acceptable fluoride in drinking water at 8-12°C is 1.5 mg/l and at 25-30°C is 0.7 mg/l [12]. The amount of fluorine is different in the groundwater depending on the type of soil layers. Its concentration in

dangerous. According to the experiments conducted on animals in connection with high exposure to fluoride in drinking water, the results suggest the possibility of adverse effects on the nervous system as well as cumulative toxic symptoms such as dental and skeletal fluorosis ^[6, 7]. Furthermore, drinking over-fluoridated water can be followed by bone diseases, including bone pain hypersensitivity in some people. Epidemiological studies have confirmed that exposure to excessive fluoride in drinking water can lead to a decrease in mental ability and an increase in the prevalence of dental fluorosis in children [8-^{11]}. Therefore, in order to control fluoride concentration and to match with the standards, public water supply systems groundwater in areas with iron ore, dolomite, and clay is 0.3-0.4 mg/l, with alkaline rocks is 7.8 mg/l, with basaltic rocks is less than 0.1 and with granite rocks is 9.2 mg/l. Since fluorine concentration in atmosphere is so low (0.5 mg/l), it can be negligible compared with the total fluorine to be absorbed into the body ^[13].

One proper way to prevent groundwater contamination is to investigate spatial variability of groundwater quality as well as to manage utilization of water resources and land use ^[14]. Geographic information system (GIS) is considered as a technology or machine that can be used to identify data (thematic maps), analyze, interpret, and consequence the data, ecological potential

social-economic assessment and requirements of human for the use of land, identify environmental changes, destructives, waste, and pollution, and above all it can be used for regional other planning or in words for [15] environmental planning Inverse distance weighting (IDW) is one of the spatial analysis methods in which the only effective factor is the distance fort weighting. Another method used in spatial analysis is Kriging model. In this interpolation method which is based on regression, the weight is not only allocated to the distance between the surrounding points, but also is based on the correlation between the measured points ^[16, 17].

Several studies have been conducted on the fluoride amount in drinking water in different parts of Iran. Noori Sepehr et al. (2006) investigated the amount of fluorine in drinking water in villages of Semnan and reported its range as 0-1.94 mg/l^[18]. In another study conducted in Sistan and Baluchestan during 2004-2006, fluoride concentration in some areas was 1.9 mg/l^[19]. Asghari Moghadam et al. (2006) stated that the concentration of fluorine in drinking water was above the world standards in Bazargan and Poldasht and all

people who were consuming spring water in that area were suffering from dental fluorosis and skeletal fluorosis in some cases [20]. Pour Islami et al. (2007) studied fluorine concentration in drinking water in Kerman Province and reported that the average concentration was 0.38 mg/l^[21]., As it was mentioned above, since fluorine concentration was regarded important in drinking water, and the ability of GIS in spatial analysis of different factors in groundwater is well established, this study aimed to investigate the fluorine concentration in groundwater in the plain of Yazd-Ardekan using IDW and Kriging model. Ultimately, an optimal model was selected concerning zoning fluorine concentration.

Materials and Methods

The research area involved Yazd province including north to the south of the Yazd province at longitude 53.778 to 54.64 and latitude 32.52 to 31.43. As it is depicted in figure 1, cities of Ardekan, Meibod and Sadough are located in the northern part of Yazd province, Taft is in the southwest and Mehriz is in the south of Yazd province). Due to its geographical location in the central plateau of Iran, Yazd province has a hot and dry desert climate.



Figure (1): Geographic scope of the research site

In this discriptive study, the data related to the quality of underground water of Yazd Regional Water Company as well as the average annual fluoride in 2014 (two sampling period per year) were used., which were related to 76 rural wells and 107 urban wells in the studied area. The fluoride rate of groundwater in rural and urban areas was measured.

GIS Arc software was used from ESRI Company in order to spatially analyze the data concerning the fluoride concentration in ground water in the plain of Yazd-Ardekan.

Krigging method, based on the average moving weighting, is recognized as the best unbiased estimator, that in addition to the estimated values, specifies the estimated error rate in each point.

In the past decade, Kriging has been introduced as a powerful interpolation technique used in various disciplines of earth sciences such as hydrology, soil science, and mining. Despite all the advantages of this method, soft changes during estimation make the variance of the estimated samples change less than the real points; i.e. the amount of changes in the model prediction is less than the reality ^[22]. In general, the success of this method in interpolation of variables completely depends on the accuracy in selecting data model with toxic variogram experiment. If the model is not selected accurately, the interpolation result won't be appropriate. In some cases, the results are not sufficiently accurate in this model due to some reasons as inconsistency of spatial structure and public structure, low accuracy of data and lack of spatial homogeneity of data.

In this study, ordinary Kriging models have been applied to investigate concentration changes of fluorine in groundwater in Yazd in terms of space and time. Kriging is a method that makes use of spatial correlation structure of phenomena in spatial estimation. In ordinary Kriging method, a certain range of points can be considered in interpolation for each destination point. It should be noted that coordinate is the points of sample relationship in UTM system. The general equation is as follows:

Equation (1) $Z * (xi) = \sum_{i=1}^{n} \lambda i z(xi)$ Where $Z^*(xi)$ is the estimation grade, λi is the weight or importance of the amount related to I sample and z(xi) is the value of measured variable.

The other zoning method applied in this study was IDW. IDW for each measurement point considers a weight according to the distance between that point and the position of unknown point. Then the weights are controlled by the weighting so as larger powers reduce the effect of farther points from the point under estimation and smaller powers distribute the weights more evenly between adjacent points. It should be noted that this method only considers the distance between points regardless their position of and

arrangement. That is, the points with similar distance from the estimate point have the same weight. The weight factor is calculated using the following equation:

Equation (2)
$$\lambda i = \frac{Di - \alpha}{\sum_{i=1}^{n} Di - \alpha}$$

Where λi is the weight of i station, D_i is the distance of I station from the unknown point and α is the weighting power.

Model validation was fulfilled using the following evaluation criteria [23].

Equation (3) $_{\text{RMSE}} = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (q_i - q_i^{*})^2}$ Equation (4) % $_{\text{RMSE}} = \left(\frac{_{\text{RMSE}}}{_{\mu}}\right) \times 100$ In these equations n is the number of data, qi is the measured value and q_i^{*} is the value predicted by the model. μ is referred to as the mean of each measured component.

The RMSE (Root Mean Square Error) is zero in the optimal state or when the estimated and measured values equal to zero. The smaller is RMSE, the more accurate is the prediction of interpolation model. RMSE is sensitive to outliers, therefore, RMSE% can be used. Smaller amount of this feature indicates more precise estimates that is to say the little difference between actual and estimated values ^[24]. Acceptable limit of RMSE% is introduced 40% and RMSE% above 70% is the sign of inaccuracy in estimating points and large variations between estimated and measured values.

Results

In the present study, the fluorine concentration of the groundwater was investigated in the plain of Ardekan –Yazd.

Thus, the sampling results of 76 wells in rural areas and 107 wells in urban areas were analyzed. Statistical description (mean, minimum, maximum, standard deviation) related to fluorine concentration in the water of urban and rural wells are displayed in Table (1). . In order to compare fluorine concentration with the standards in the studied area, Iran's 1053 standard for drinking water was utilized[25]. The standard is displayed in Table (2).

Table (1): Statistical description (mean, minimum, maximum, standard deviation) of fluorine in the water

of wells							
Standard deviation	Minimum concentration of fluorine (mg/l)	Maximum concentration of fluorine (mg/l)	Mean concentration of fluorine (mg/l)*	Type of well			
0.278	0.16	1.5	0.7	Urban			
0.364	0.02	1.96	0.68	Rural			

*confidence interval (CI) 95%

Table (2): Water Standard (1053), minimum and maximum concentrations of fluoride in drinking water

The annual maximum of daily air temperature*	Minimum permitted amount of fluorine(mg/l)	Average amount of fluorine (mg/1)	Maximum permitted amount of fluorine(mg/l)
10-12	1.1	1.2	2.4
12-14.6	1	1.1	2.2
14.6-17.7	0.9	1	2
17.7-21.5	0.8	0.9	1.8
21.5-26.3	0.7	0.8	1.6
26.3-32.5	0.6	0.7	1.4

* Average temperature is based on the average maximum temperature of the region in five consecutive years.

According to Iran's standard of drinking water within the studied region as well as considering weather conditions, fluorine concentration should range from 0.6 to 1.4. Table 1-4 demonstrates that the mean concentration of fluorine in urban and rural areas was reported in the standard range, though with regard to the minimum and maximum concentration presented in some areas, the fluorine rate was less than the allowable amount and in some areas fluride concentration was exceeds. In general, the total concentration of fluorine in urban areas is more than that of rural areas. However, fluctuation in fluorine concentration in rural areas with regard to minimum concentration of 0.02, maximum concentration of 1.96 mg/l and standard deviation of 0.364 was more than that of urban areas. The main reason of greater fluctuation in rural areas isprobably related to more distribution of wells in these areas as well as more varied texture of soil in comparison to urban areas. In urban areas, the fluorine concentration in 38 wells was lower than the permitted limit (0.6 mg/l), though the fluorine concentration in 2 wells exceeded the maximum permitted limit (1.4 mg/l). In rural areas. the fluorine concentration in 36 wells was lower than the permitted limit (0.6 mg/l), whereas the fluorine concentration in 2 wells exceeded the maximum permitted limit (1.6 mg/l).

To select the optimum conditions for Kriging model, at first the date was normalized via Box-Cox model and then all the models in the software were analyzed. Ultimately, given the amount of RMSE it was found that Gaussian model was the best model for zoning fluorine concentration in rural areas through Kriging method. In IDW method, 1, 2, and 3 powers were used to select the optimum conditions. Figures 2-A and 2-B respectively indicate zoning via Kriging method and IDW in rural areas and Figures 3-A and 3-B show zoning in the urban areas.



Figure 2 (a): Zoning fluorine concentration in rural areas using kriging method



Figure 2 (b): Zoning fluoride concentration in rural areas using IDW



Figure 3 (a): Zoning fluorine concentration in urban areas using kriging method



Figure 3(b): Zoning fluorine concentration in urban areas using IDW

In order to compare Kriging and IDW methods, RMSE criterion was used which was equal to 0.378 for Kriging model and 0.447 for IDW in rural areas. In urban areas, it was equal to 0.266 and 0.294 for Kriging model and IDW, respectively. In this way, Kriging model had better conditions than IDW in regard with zoning fluorine concentration in both rural and urban areas. RMSE percentage for rural

areas was 55% and 65% respectively, which indicated the average performance of models fluorine the two on the concentration data in rural areas. This may be due to outliners in the zoning of rural areas. In Urban areas, RMSE was equal to 38% for Kriging method and 42% for IDW method which indicated good conditions of Kriging method in regard with zoning fluorine concentration (Table 3).

Table 3: comparing interpolation methods in rural and urban areas

Kind of area	Interpolation method	RMSE	%RMSE
Rural	IWD	0.447	65
	Kriging	0.378	55
Urban	IDW	0.294	42
	Kriging	0.266	38

In order to show zoning maps better and more simply in both urban and rural areas, the maps were classified according to standard rate in Iran as shown in Figures 4 and 5. According to Figure 4, it can be concluded that the main problem of fluoride shortage in drinking water is related to northeastern villages and parallely, to the southwestern villages of the studied region. Figure (5) also reportsthat lack of fluoride is more related to cities in central areas such as Yazd southeast area.



Figure (4): The classification of fluorine concentration based on the relative standard of rural areas and Kriging method



Figure (5): The classification of fluorine concentration based on the relative standard of urban areas and Kriging method

Discussion

In this study, the concentration of fluorine in the wells in rural and urban areas in Yazd - Ardekan was studied. Given the average concentration in 2014, it was found that concentration fluctuations of fluoride in various rural areas were more than those of urban areas. In urban areas, wells with standard concentration were more than those of rural areas. According to the maps of fluorine concentration in urban and rural areas, it can be concluded that in urban and rural areas no problem fluorine concentration over of the permitted limit (more than 1.4 mg/l) was

observed and therefore there is no concern regarding diseases such as dental or skeletal fluorosis.

Since groundwater is largely affected by the type of terrain textures through which it passes, it has different mineral components. If the terrain texture through which groundwater passes contains feldspar rocks, the groundwater contains high amount of fluoride, as well. Otherwise, the majority of groundwater entails small amount of fluoride and in order to prevent tooth decay, it is necessary to develop fluoridation unit for such water ^[15, 16]. In Iran, there are some areas where fluoride concentration has exceeded the allowable limit. Generally, northern and central provinces have the lowest and southern provinces have the highest percentage of fluoride in their water. However, in most parts of Iran, fluoride deficiency has been reported via the number of referrals to dentistry ^[18, 26].

Among different studies conducted in Iran, the one conducted by Shams et al. can be referred, in which the rate of fluoride drinking in water in Tabas was examined.It was observed that the fluoride rate in summer and winter months was equal to 0.71 mg/l and 0.58 mg/l respectively which was very close to the rate of fluoride in the present study ^[27]. In another study, the rate of fluoride ion in Maroon, Karun, and Karkhe Rivers was examined. The average concentration of fluoride ion was detected to be equal to 0.5 mg/l, which was smaller than the average rate of fluoride in urban and rural areas in the present study ^[28]. In examining the quality of water in Noor Abad, Mamasani Plain, the minimum and maximum rate of fluoride was reported as 0.19 and 1.36 mg/l, respectively $^{[29]}$. Omrani et al. studied fluoride concentration in drinking water and its relationship with decayed missed filled teeth (DMFT) within the 7-11-year-old students in Shiraz. After measuring fluoride concentration in the studied areas, they found that in some areas with less fluoride concentration than the standard, the fluoride rate was significantly different from other areas (the mean concentration of fluoride in urban drinking water was measured as 0.69 mg/l) ^[30].

Conclusion

Examining the quality of groundwater through GIS-based approaches can greatly contribute to better understanding of spatial conditions of each component in groundwater, which is highly potential in this regard. As the study findings revealed, fortunately fluoride concentration above the standard limit was observed in none of the areas, though northeastern parts, central, and southern parts of rural areas as well as central and southeastern parts of urban areas in the study are facing shortage of fluoride in drinking water. In order to prevent tooth decay and fluoride deficit problems in drinking water, necessary management should be done such as adding fluoride to drinking water, mixing lowfluoride-concentration water with highfluoride-concentration water as well as teaching people to use fluoride mouthwash and toothpaste.

References

- 1. Whitford G.M. Acute and chronic fluoride toxicity. Journal of .Dent Res 1992;71: 1249–54.
- Environmental Protection Agency. Fluoride: Exposure and Relative Source Contribution Analysis. Washington D.C, U.S; 2010.

- World Health Organization. Fluorides and oral health. Geneva: WHO. 1994; (WHO Technical Report Series 846).
- 4. Whitford GM. The Metabolism and Toxicity of Fluoride. 2th Ed. New York: Karger; 1997.
- Centers for Disease Control and Prevention. Achievements in Public Health, 1900-1999: Fluoridation of Drinking Water to Prevent Dental Caries for Water Fluoridation. MMWR 1999; 48: 933-40.
- Mullenix PJ, Denbesten PK, Schunior A, et al. Neurotoxicity of sodium fluoride in rats. Neurotoxicol. Teratol 1995;17(2):169–77.
- Sudhanshu S, Anjali S, Pankaj G .Effect of high fluoride water on intelligence of school children in India. J Neurosci Rural Pract 2012; 3(2): 144–49.
- Chen YX, Han FL, Zhou ZL, et al. Research on the intellectual development of children in high fluoride areas. Fluoride; 2008; 41(2):120–04.
- Sun DJ. Surveillance on endemic fluorosis of drinking water type in China a two-year report of 2003 and 2004. Chin Journal of Epidemiology 2007; 26:161–4.
- Suthar S, Garg VK, Jangir S, et al. Fluoride contamination in drinking water in rural habitations of Northern Rajasthan, India. Environ Monit Assess 2008; 145(1-3):1-6.
- Gupta RK, Agarwal P, Kumar S, et al. Compressive myelopathy in fluorosis MRI. Neuroradiology. 1996 May;38(4):338-42.
- APHA, AWWA, WPCF. Standard methods for the examination of water and wastewater 21th Ed. Washington DC, USA: American Public Health Association; 2005; 1168-9.
- Zandevakili F, Doghashmeh M, Daneshmand Irani K. Investigation of Physico chemical and microbiological quality of mineral waters and packed Iran. The 8th National Congress on Environmental Health, hamedan.2005 (Persian).
- Ehsani H JA, Hasani A, Shariat M, et al. Evaluation of nitrate variation and Total dissolved solids trend in drinking water using GIS Hamedan plain ground. 10th national Conference on Environmental Health. 2007.
- 15. Manimaran D. Groundwater Geochemistry Study Using GIS in and Around Vallanadu Hills, Tamilnadu, India ResJRecent Sci. 2012;15:52-8.
- 16. Cressie N. A. C, Statistics for spatial data; J. Wiley, 1993.
- 17. Schabenberger O, Gotway C A. Statistical methods for spatial data analysis; CRC Press, 2005.
- Noori Sepehr M, Arab F, Shahravi M, et al. investigation of fluoride concentrating in Semnan Village at 2006. Proceedings of the Tenth conference of environmental Health, 12-15 October 2007, Hamadan, Iran: 17-23. (Persian).
- Nori MA, Bazrafshan E, Hosseini AR, et al. survay the level of fluride in drinking water sources in Sistan & Balochestan province Cities in 2003, 2004, 2005. 11th national Conference on Environmental Health, Zahedan univercity of medical science. 2007:87 (Persian).
- Asghari moghadam A, Jamiri R, Mohammadi A. The source of high level concentration of fluride in ground water of Bazargan-Poldasht plain and harm effects on citizenry of this regain journal of environmentology. 2007;41:42.

- poreslami HR, Khazaeli P, Masood por H. survay the level of fluride in drinking water sources in Kerman province Cities. journal of Kerman uiversity of medical science. 2008;15(3):238.
- Xu C. GL, Jiang T, Chen D, et al. Analysis of spatial distribution and temporal trend of reference evapotranspiration and pan evaporation in Changjiang (Yangtze River) catchment. Journal of Hydrology. 2007;327:81-93.
- 23. Mehrjerdi R ZM, Mahmodi Sh, Heidari A. Spatial distribution of groundwater quality with geostatistics (Case study: Yazd- Ardakan plain). World Applied Science Journal. 2008;4(1):9-17.
- Hengel T. HGBMaSA. A generic framework for spatial prediction of soil variables based on regression–kriging. Geoderma. 2004;120:75-93.
- 25. Institute of Standard and Industrial Researches. Drinking water charachtersitics. 5th ed. 2010. (Persian).
- 26. Golshani H. investigation of relationship between fluoride concentration in water with dental caries in Esfahan.1996,Tehran, MPSH Thesis. (Persian).
- Shams M, Mahvi A, Mohammadi A. Investigation of Nitrate and Fluoride concentration in Tabas water supply. Proceedings of the Twelfth conference of environmental Health. 12-15 October 2009. Tehran, Iran: 47-53.(Persian).
- Basir L, Khaneh Masjedi M, Haghighi M, et al. investigation of relationship between fluorosis and DMFT outbreak with fluoride concentration in Khozestan drinking water in 10-12 old children at 2002. Den. J. Shahid Beheshti Un. Med. Sci. 2006; 24: 14-23.(Persian).
- Fallah H, Molana Z, Mir Arab Razi M. investigation of fluoride in Ramian drinking water and determination of DMFT in children. Proceedings of the Twelfth conference of environmental Health, 12-15 October 2009, Tehran, Iran: 59-65. (Persian).
- Emrani M, Dehghani .Detection of DMFT in 7-11 years old student in shiraz. Proceeding of the 12th of environmental health congress. Tehran, Shahid beheshti university, 2009. (Persian).