

# Fuzzy Framework of Health Risk Assessment of Nitrates in Groundwater

Samia Chiban<sup>1</sup>, Adel Awad<sup>2</sup>, Ahmad Wazzan<sup>2</sup>, Hanan Sheikh-Youssef<sup>3</sup>

<sup>1</sup>Higher Institute of Environmental Research, Tishreen University, Latakia, Syria

<sup>2</sup>Faculty of Civil Engineering, Tishreen University, Latakia, Syria

<sup>3</sup>Environmental Engineering, Department of Water Safety, Environment Directorate, Tartus, Syria

Email: samia982@yahoo.com

**How to cite this paper:** Chiban, S., Awad, A., Wazzan, A. and Sheikh-Youssef, H. (2018) Fuzzy Framework of Health Risk Assessment of Nitrates in Groundwater. *Applied Mathematics*, 9, 586-601.  
<https://doi.org/10.4236/am.2018.96041>

**Received:** April 3, 2018

**Accepted:** June 9, 2018

**Published:** June 12, 2018

Copyright © 2018 by authors and Scientific Research Publishing Inc.  
This work is licensed under the Creative Commons Attribution International License (CC BY 4.0).  
<http://creativecommons.org/licenses/by/4.0/>



Open Access

## Abstract

This study provides a characterization of potential health risk associated with groundwater Nitrates in Akkar plain in Tatars governorate in Syria. Six villages were chosen for Nitrates analysis in artesian wells. The results show that Nitrates concentrations are above the natural level but still below the level of water potability and measures have to be taken to prevent Nitrates accumulation in groundwater. Comparing Nitrates dose in young children diet with reference dose and calculating the hazard index give the noncarcinogenic risk (Methemoglobinemia). Carcinogenic risk of drinking well waters with Nitrates is calculated in a probabilistic-fuzzy framework. The role of Bacterial contamination of water and its association with Nitrates pollution is discussed and we found that the two pollutants are not correlated in Akkar plain, because Nitrates come principally from fertilizers in this area while fecal coliforms come from landfill leachates and untreated wastewaters. Nitrates and FC play associated roles in gastro-intestinal disturbance and Methemoglobinemia.

## Keywords

Drinking Water, Methemoglobinemia, Carcinogenic Risk, Fuzzy Logic, Probability, Artesian Wells, Fecal Coliforms, Pollution, Potability

## 1. Introduction

It is common to see in the recent scientific researches, hypotheses that highlight the role of environmental factors in cardio-respiratory diseases, breast morbidity, some cancer types: stomach, colon and rectum, liver, pancreas, larynx, lung, bladder, kidney, brain, lymphatic and haematopoietic cancer [1]-[10].

Environmental factors mean a variety of exposure risks include air pollution because of vicinity of industrial complexes, landfills, incinerators, drinking water pollution because of fertilizers, herbicides, chlorine compounds... These pollutants can accumulate in the natural food chain and arrive to human as well.

Groundwater Nitrates pollution is a worldwide problem. Nitrates are soluble in water and move easily through the soil and into the groundwater. Because of development of farmlands and application of chemical fertilizers, particularly azote fertilizers, Nitrates became one of the main sources of soil and water pollution. Therefore, it is necessary to investigate Nitrates pollution of groundwater [11]. Nitrates accumulate in groundwater according to two factors, Nitrates sources from surface layer and the vulnerability of aquifer to Nitrate. Vulnerability of an aquifer is a term used for describing the easiness of pollutants of soil surface to reach the groundwater. High-Nitrates drinking water is most often associated with privately-owned wells.

In the European Union, groundwater protection against pollution is regulated by the European directive (2006/118/EC) [12] that is abbreviated by GWD “Ground Water Directive”. Regarding Nitrates, the GWD establishes the quality standard for assessing groundwater chemical status of 50 mg/L, in addition to other criteria for the definition of groundwater status (quality and quantity). Nitrates status forms a fundamental part of the GWD and directives concerning surface water protection against pollution. The Nitrates directive (1991/676/EEC) [13] was drawn up with the specific purpose to reduce water pollution caused by Nitrates from agricultural sources and prevent further such pollution [14]. Moreover, according to the directive, EU members are asked to identify waters affected by Nitrates pollution called Nitrates Vulnerable Zones (NVZs). The NVZs are defined as areas where the groundwater contains or could contain (if no action is taken to reverse the trend) more than 50 mg/L of Nitrates.

Nitrates has been implicated in methemoglobinemia and also a number of currently inconclusive health outcomes [15]. These include proposed effects such as cancer (via the bacterial production of N-nitroso compounds), hypertension, increased infant mortality, birth defects, diabetes, spontaneous abortions, respiratory tract infections, and changes to the immune system (Centers for Disease Control and Prevention CDC 1996) [16].

Categorically, Nitrates in drinking water is a contributing factor not a unique cause of methemoglobinemia and gastric cancer. Methemoglobinemia has several causes as hereditary, Drug/Chemical induced and Diet induced [15]; these causes and more can induce gastric cancer. Complex co-factor relationships do not currently allow the establishment of a quantitative exposure-response relationship for human exposure to Nitrates in food or water and the subsequent development of methemoglobinemia or cancer. Nitrates or nitrite (ingested), under conditions that result in endogenous nitrosation, are registered in Group 2A “Probably carcinogenic to humans” by the IARC (International Agency for Research on Cancer).

Recently, Researchers around the world are working continuously to assess the health risk of long term exposure to Nitrates in drinking water. Sadler *et al.* 2016 [17] used probabilistic techniques in order to assess health risk for the local populations in rural areas in Indonesia. Their results indicate a low risk of infant methaemoglobinaemia for the whole population, but some risk for the sensitive portion of the population and an elevated risk for birth defects, in particular for the more sensitive population. According to the study, a sanitation improvement program in the study area had shown a positive effect in reducing nitrate levels in wells and the corresponding risk for public health. In the study of Schullehner *et al.* 2017 [18], Nitrate was used as an example contaminant in presenting an approach making drinking water quality data from the Danish national geodatabase Jupiter available for epidemiological studies on long-term health effects of drinking water quality. Espejo-Herrera *et al.* 2015 [19] suggested in their study, about Nitrate in drinking water and bladder cancer risk in Spain, that elevated risk of bladder cancer is found only among subjects with longest exposure duration to the highest levels of Nitrates. Stayner *et al.* 2017 [20] investigated the role of Atrazine and nitrate in drinking water and the risk of preterm delivery and low birth weight and they found a strong interaction between atrazine and nitrate for preterm delivery.

In this research, we try to assess the noncarcinogenic and carcinogenic risks of drinking waters with high Nitrates concentrations. We evaluate the risk of drinking water Nitrates in wells in South of Tartus governorate in Syria, and we try to count the principal pollution sources in Tartus governorate that has the highest cancer rates in Syria.

## 2. Noncarcinogenic Health Effects of Nitrates in Drinking Water

Infants under 3 months of age are particularly susceptible to methemoglobinemia. Because the pH of the infant's gastric juice is relatively high (from 5 to 7), Nitrates is reduced to nitrite by the Nitrate-reducing bacteria in the stomach, and the nitrite is absorbed in the blood and then converts hemoglobin (a protein in red blood corpuscles) to methemoglobin (MetHb), which cannot carry oxygen to the body's tissues [21].

The methemoglobin levels of normal healthy individuals are about 0.5% - 2.0% of the total hemoglobin. At levels above 10%, clinical signs are evident (*i.e.*, a bluish color in the skin and lips is produced) and the condition is termed methemoglobinemia. Generally, methemoglobinemia is not a notifiable disease; and definitions of methemoglobinemia (in terms of the required level of MetHb) vary in the literature [15]. In the USEPA 2008 report [22] on Dose-Response of Nitrates and other Methemoglobin inducers on Methemoglobin levels of infants' risk, the risk factors are studied in the case of MetHb level is greater than 2% in the child blood and in the case of MetHb level is greater than 3% in the child blood. However, methemoglobinemia responds rapidly to treatment.

Current methods for estimating noncarcinogenic health effects (e.g., methemoglobinemia) from human exposure to chemicals rely on the concept of a Reference Dose (*RfD*). The *RfD* is an estimation of daily exposure that is not expected to produce an adverse effect over a lifetime, and it can be derived using the following formula [23]:

$$RfD = \frac{NOAEL \cdot DW}{UF} \quad (1)$$

where:

The No Observed Adverse Effect Level (*NOAEL*) is an exposure level (mg/l) at which there are no statistically or biologically significant increases in the frequency or severity of adverse effects for the exposed population compared to its appropriate control group. It is assumed from laboratory experiments of (Daurson *et al.*, 1991) [23] that the value of *NOAEL* = 10 mg/l NO<sub>3</sub>-N (WHO and USEPA standard); *DW* refers to daily water consumption of an infant (L/day/infant); this value is taken for a 4 kg-weight infant = 0.64 l/day. Infant can be written as 0.16 L/kg-d.

*UF* is an uncertainty factor, which usually ranges from 1 to 1000 [24]. The appropriate value of the uncertainty factor (*UF*) for infants is 1.0 because they are the most sensitive population.

Thus, we find:

$$RfD = (10 \times 0.64) / 1 = 6.4 \text{ mg NO}_3\text{-N/day} \cdot \text{infant}$$

As 10 mg/L Nitrate-N is equivalent to 44 mg/L Nitrate, the reference dose of Nitrates can be written for every kg of the infant weight:

$$RfD = (44 \times 0.16) / 1 = 7.04 \text{ mg NO}_3\text{/kg} \cdot \text{day}$$

The Average Daily Dose *ADD* of Nitrates taken by every kg of infant weight can be calculated:

$$ADD(\text{mg NO}_3\text{/kg} \cdot \text{day}) = C(\text{mg NO}_3\text{/l}) * DW(\text{l/kg} \cdot \text{d}) \quad (2)$$

*C* is Nitrates concentration in drinking water.

*DW* is the daily water consumption of an infant “already defined”.

The ratio of *ADD* to *RfD* is called the Hazard Index *HI*.

$$HI = ADD / RfD \quad (3)$$

Methemoglobinemia risk does not exist if  $HI < 1.0$ .

But the risk exists if  $HI > 1.5$ .

While one may say, on the basis of strong evidence, that methemoglobinemia cannot occur in infants if  $1.0 < HI < 1.5$ .

### 3. Carcinogenic Effects of Nitrates in Drinking Water

To compensate for the current lack of data related directly to cancer incidence in humans, Nitrate-induced human cancer risk may be estimated based on tests performed in laboratory animals. Though this interspecies conversion is a controversial issue, it may be the only method for modeling the relationship be-

tween Nitrates dose to humans and its corresponding cancer response [25].

Lee 1992 performed a study to estimate human cancer risk corresponding to a particular Nitrates dose. In the study, animal data “rats” obtained from (Terracini *et al.*, 1967) [26] are used for the interspecies conversion, in addition to a combined probabilistic/fuzzy-set framework employed to deal with uncertainty. The Nitrates Dose-Cancer Response relationship is as follows:

$$\frac{1}{1 + \exp(Z_1)} \leq Y \leq \frac{1}{1 + \exp(Z_2)}$$

$$Z_1 = 3.331 + (1 - p)1.138 - 3.429(\ln X) + (1 - p)0.881|\ln X| \quad (4)$$

$$Z_2 = 3.331 - (1 - p)1.138 - 3.429(\ln X) - (1 - p)0.881|\ln X|$$

where

$X$  = total Nitrates intake by a human (g/day),  $X = NW + NF$ ;

$NW$  = Nitrates intake from drinking water (g/day);

$NF$  = Nitrates intake from sources other than drinking water and it is taken as an upper estimate about 0.15 g/day (including the amount obtained by the conversion of ingested nitrite to Nitrate);

$Y$  = lifetime probability of developing human gastric cancer;

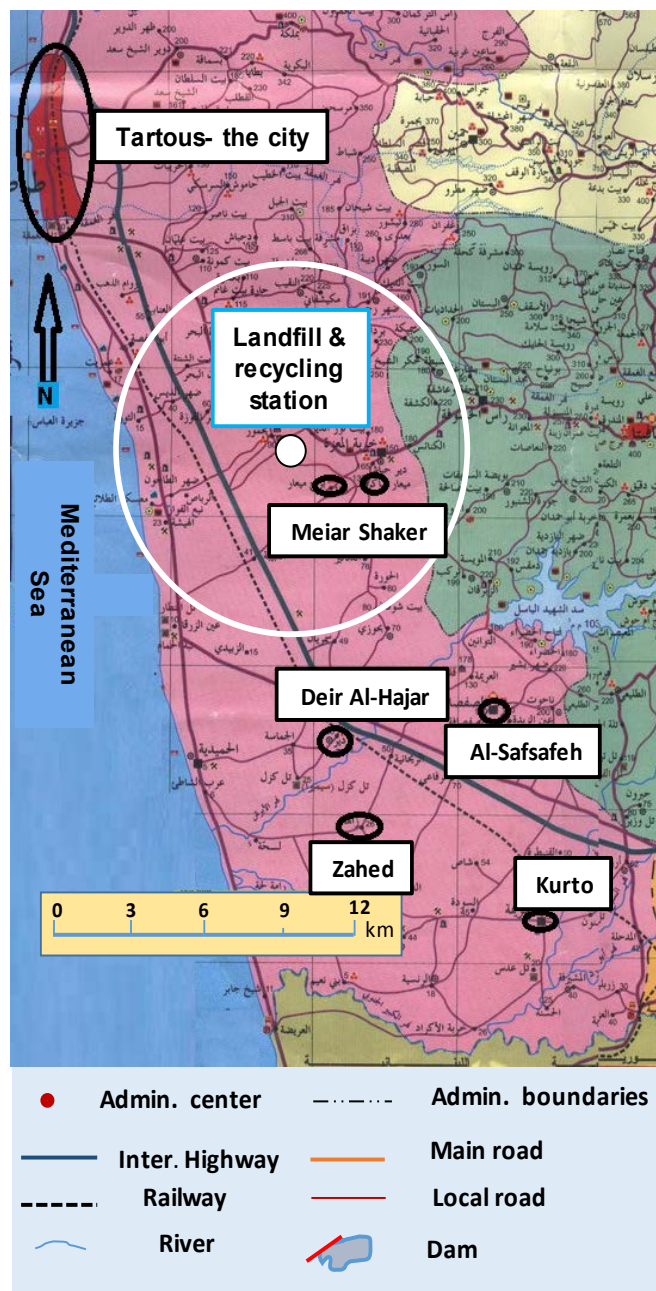
and  $p$  ( $0 < p < 1$ ) = the membership degree, which is used to define the domain of the response  $Y$  at various degrees of membership. The value  $p = 0.5$ , is normally taken which is neither optimistic nor pessimistic.

#### 4. Study Area

The coastal area in the eastern coast of the Mediterranean is characterized by a large population density and relies mainly on agriculture. Akkar plain is considered one of the most fertile agriculture land in the region. Its total area in Syria is about 26,600 ha, with 22,300 ha of arable land. In this research, we consider the zone of Akkar plain located in Tartus District (North Western Syria) with area of 18,600 ha (Figure 1).

The manifestations of water saturation are observed clearly in this zone of plain especially in lowlands. The problem of water saturation can be linked to the existing poor surface drainage system that needs rehabilitation while there is no subsurface drainage system. The absence of efficient drainage system and the intensive use of fertilizers in agriculture have serious health consequences presented by the elevated concentration of Nitrates in groundwater used for drinking and irrigation in this vital area. The elevated Nitrates concentrations can be referred also to the municipal wastewater disposal without treatment and breeding activities in the rural areas.

In the plain area, many rivers flow, the most remarked ones are Nahral-Kabir al-Janoubi “Arabic for the Southern Great River”, Al-Abrash river and Al-Arous river. A lot of small dams are constructed on rivers in the area. In the past, there were about 4500 wells in the plain, of which 3700 wells are in Tartus District alone. For many reasons, most of these wells are out of service now, increasing the problem of water saturation.



**Figure 1.** Locations of the studied villages in Akkar plain in Tartus governorate, with 7 km buffer around landfill.

In order to evaluate health and environmental risk assessment of the elevated Nitrates concentrations in Akkar plain, we have selected six villages that depend mainly on groundwater to provide drinking water (Figure 1). We located the wells used as a source for drinking water, and we analyzed Nitrates in water samples taken monthly during one year, from May 2009 to May 2010. The depth of wells serving the studied villages and some other information are given in Table 1.



**Table 1.** Artesian wells characteristics in the studied villages.

Area	Well n.	Depth	Notes	Area	Well n.	Depth	Notes
Al-Safsafah	1	225	Exploited from 2008	Kurto	4	100	Exploited from 2000
Der-Alhajar	2	140	Exploited from 2000	Meiar Shaker	5	125	Exploited from 1990
Zahed	3	100	Exploited from 1982	Meiar Shaker—the plain	6	120	Exploited from 1995

## 5. Analysis of Nitrates Concentration Results

Nitrates concentrations measured monthly in the six areas are depicted in **Figure 2** and the seasonal average of these values are depicted in **Figure 3**. Taking into account that the permitted range of Nitrates in drinking water is 45 milligram per liter, there is no alarm of danger for the aquifer of Akkar plain located in Tartus district, except for Kerto village wells, where high values of Nitrates are noticed in June 2009 and February 2010.

The small values of Nitrates concentrations in groundwater in spite of the excessive use of fertilizers can be referred to the large depth of selected wells. The large depth means a relatively long time of pollutants to travel through soil until reaching groundwater. In this time, the passing water undergoes a real filtration process by natural physical, chemical and biological means in the soil media, results in water arriving to the aquifer with reduced pollutants.

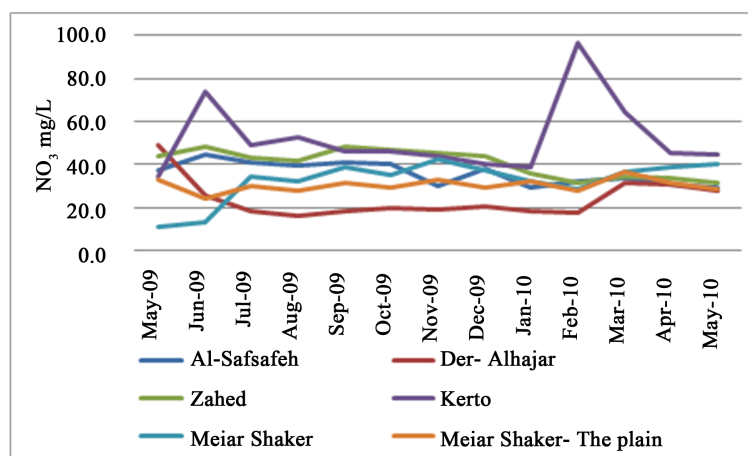
### 5.1. Noncarcinogenic Effects of Nitrates in the Studied Area

We use Equation (3) to evaluate the non-carcinogenic risk from drinking groundwater. The results are shown in **Table 2**. We find that the risk is negligible except for Kerto. Thus, the groundwater quality has to be monitored regularly if it is used as drinking water and farther investigations can be led to see whether the reasons of water pollution can be treated or the population of Kerto has to depend on alternative source to drink.

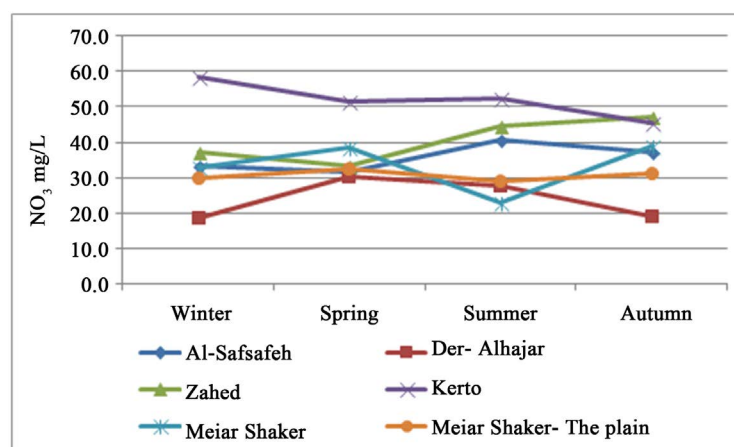
### 5.2. Nitrates and Bacterial Pollution

While Nitrates exist naturally in groundwater, concentrations over 1 mg/L Nitrates indicate human activity according to a study conducted in the United States [27]. However, no cases of methemoglobinemia occurred in infants consuming water containing concentrations less than 10 mg/l  $\text{NO}_3\text{-N}$  unless the water was bacterially contaminated [28]. In the report of USEPA 2008 [22], Nitrates intake and exposure to fecal water contamination were considered the major exposure factors. It was noted that they are generally correlated and it is very challenging to separate the effects of these risk factors and measure their actual and independent effects.

In our research, no bacterial analysis in groundwater was performed. It is worth to mention that this analysis was included in the study of Shaheen *et al.* 2013 [29] of some groundwater pollution indicators in artesian wells, located in



**Figure 2.** Monthly Nitrates results for the studied villages.



**Figure 3.** Average seasonal variations of Nitrates in Artesian wells in the studied villages.

Akkar plain in the neighboring of Wady-Alhedda Center for solid wastes disposal and recycling (**Figure 1**). Their study shows Nitrates concentrations between 0.2 and 31.4 mg/L (**Figure 4**). The high Nitrates values were observed in wells located in agriculture land. The fecal coliforms count was in the interval 10 - 150 FC/100 mL (**Figure 5**). According to the authors, the elevated microbial contamination is linked with landfill and discharge of untreated domestic wastewater. Thus, the groundwater was not suitable as a source for drinking water in that zone. However, in the studied villages, the problems of small-scale landfills and untreated wastewater increase the probability of groundwater microbial contamination. Several cases of gastro-intestinal cases were reported among population inciting them to use the municipal potable water as a unique source of drinking water and use the ground water from the artesian wells for irrigation only.

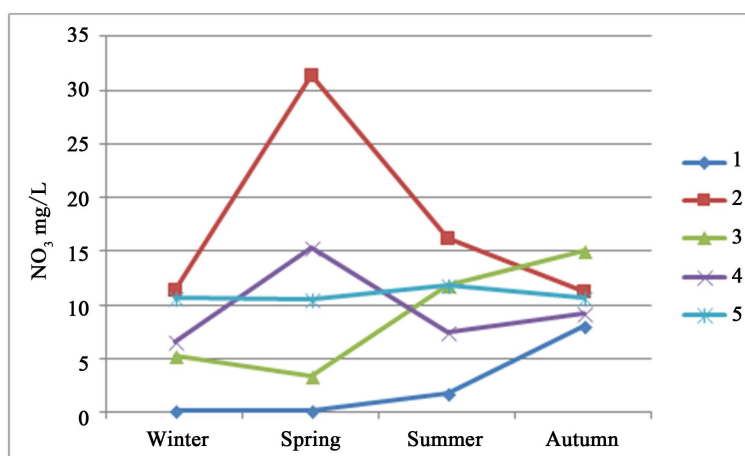
From **Figure 4** and **Figure 5**, we note that Nitrates and FC concentrations are not correlated and vary independently with season, what probably mean that they came from different sources (well 2 is located downstream the landfill



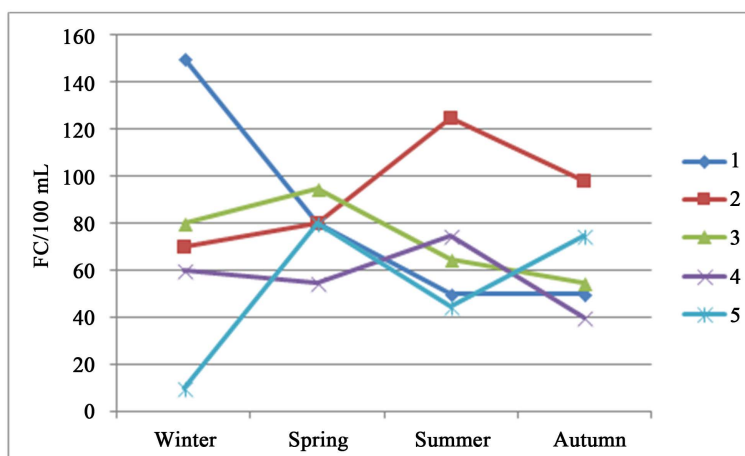
**Table 2.** Nitrates concentrations and associated Methemoglobinemia risk.

		Al-Safsafteh	Der-Alhajar	Zahed	Kerto	Meiar Shaker	Meiar Shaker—the plain
May-09	NO <sub>3</sub>	37.4	48.9	43.7	34.5	11.0	33.0
	ADD	6.0	7.8	7.0	5.5	1.8	5.3
	HI	0.9	<b>1.1</b>	<b>1.0</b>	0.8	0.3	0.8
Jun-09	NO <sub>3</sub>	45.0	25.8	48.7	73.9	13.0	24.0
	ADD	7.2	4.1	7.8	11.8	2.1	3.8
	HI	<b>1.0</b>	0.6	<b>1.1</b>	<b>1.7</b>	0.3	0.5
Jul-09	NO <sub>3</sub>	40.9	18.6	43.5	48.8	34.8	30.3
	ADD	6.5	3.0	7.0	7.8	5.6	4.8
	HI	0.9	0.4	<b>1.0</b>	<b>1.1</b>	0.8	0.7
Aug-09	NO <sub>3</sub>	39.7	16.6	41.7	52.4	32.3	28.2
	ADD	6.4	2.7	6.7	8.4	5.2	4.5
	HI	0.9	0.4	0.9	<b>1.2</b>	0.7	0.6
Sep-09	NO <sub>3</sub>	41.1	18.4	48.4	46.2	38.7	31.4
	ADD	6.6	2.9	7.7	7.4	6.2	5.0
	HI	0.9	0.4	<b>1.1</b>	<b>1.1</b>	0.9	0.7
Oct-09	NO <sub>3</sub>	40.4	19.8	47.2	46.0	35.4	29.3
	ADD	6.5	3.2	7.6	7.4	5.7	4.7
	HI	0.9	0.5	<b>1.1</b>	<b>1.0</b>	0.8	0.7
Nov-09	NO <sub>3</sub>	29.8	18.9	45.8	43.8	42.7	32.8
	ADD	4.8	3.0	7.3	7.0	6.8	5.2
	HI	0.7	0.4	<b>1.0</b>	<b>1.0</b>	<b>1.0</b>	0.7
Dec-09	NO <sub>3</sub>	37.8	20.4	43.7	40.1	37.2	29.4
	ADD	6.0	3.3	7.0	6.4	6.0	4.7
	HI	0.9	0.5	<b>1.0</b>	0.9	0.8	0.7
Jan-10	NO <sub>3</sub>	29.4	18.4	36.3	39.1	32.4	32.5
	ADD	4.7	2.9	5.8	6.3	5.2	5.2
	HI	0.7	0.4	0.8	0.9	0.7	0.7
Feb-10	NO <sub>3</sub>	32.5	17.4	31.5	96.4	28.4	27.8
	ADD	5.2	2.8	5.0	15.4	4.5	4.4
	HI	0.7	0.4	0.7	<b>2.2</b>	0.6	0.6
Mar-10	NO <sub>3</sub>	33.6	31.7	34.7	64.4	36.4	36.7
	ADD	5.4	5.1	5.6	10.3	5.8	5.9
	HI	0.8	0.7	0.8	<b>1.5</b>	0.8	0.8
Apr-10	NO <sub>3</sub>	30.8	30.9	33.5	45.7	38.7	31.6
	ADD	4.9	4.9	5.4	7.3	6.2	5.1
	HI	0.7	0.7	0.8	<b>1.0</b>	0.9	0.7
May-10	NO <sub>3</sub>	29.7	27.6	31.9	44.6	40.4	28.8
	ADD	4.8	4.4	5.1	7.1	6.5	4.6
	HI	0.7	0.6	0.7	<b>1.0</b>	0.9	0.7

NO<sub>3</sub>: Nitrates concentration in wells mg/L; ADD: Average Daily Dose; HI: Hazard Index (Equations (1)-(3)).



**Figure 4.** Nitrates concentrations in five artesian wells near solid waste recycling center (adopted from [29]).



**Figure 5.** Fecal coliforms concentrations in five artesian wells near solidwaste recycling center/landfill in Akkar plain in 2011-2012 (Adopted from [29]).

which explain the high values of Nitrates and FC in all seasons). We note also that the interval of concentrations is decreased in Autumn for Nitrates and FC because of the buffering effects of filtered rain water into the ground on pollutants concentrations.

Comparing **Figure 3** and **Figure 4** shows that Nitrates concentrations in Agricultural land in the studied villages are clearly higher than those around the recycling center which also shows that agriculture is the main cause of Nitrates accumulation in groundwater in Akkar plain rather than untreated wastewater or landfill leachate.

### 5.3. Carcinogenic Effects of Nitrates in the Studied Area

**Table 3** shows the Carcinogenic effects resulting from drinking groundwater in the studied villages.  $Y_{\min}$  and  $Y_{\max}$  present the lowest and highest cancer probability by 100,000 persons. We find that the interval  $[Y_{\min}, Y_{\max}]$  is relatively large

**Table 3.** Nitrates concentrations and cancer probability  $Y_{\min}$  and  $Y_{\max}$  in 100,000 person in the studied villages.

		Al-Safsafteh	Der-Alhajar	Zahed	Kerto	Meiar Shaker	Meiar Shaker—the plain
May-09	$\text{NO}_3$	37.4	48.9	43.7	34.5	11.0	33.0
	X	49.070	43.870	34.670	11.170	33.170	0.170
	$Y_{\min}$	8.7	12.4	10.6	8.0	3.4	7.6
	$Y_{\max}$	94.1	123.0	109.3	87.6	45.5	84.3
Jun-09	$\text{NO}_3$	45.0	25.8	48.7	73.9	13.0	24.0
	X	0.260	0.222	0.267	0.318	0.196	0.218
	$Y_{\min}$	11.0	5.9	12.3	24.0	3.7	5.6
	$Y_{\max}$	112.6	69.9	122.5	205.0	48.4	66.6
Jul-09	$\text{NO}_3$	40.9	18.6	43.5	48.8	34.8	30.3
	X	0.252	0.207	0.257	0.268	0.240	0.231
	$Y_{\min}$	9.7	4.6	10.5	12.3	8.0	6.9
	$Y_{\max}$	102.4	57.2	108.8	122.7	88.2	78.7
Aug-09	$\text{NO}_3$	39.7	16.6	41.7	52.4	32.3	28.2
	X	0.249	0.203	0.253	0.275	0.235	0.226
	$Y_{\min}$	9.4	4.2	10.0	13.7	7.4	6.5
	$Y_{\max}$	99.5	53.9	104.3	132.9	82.9	74.5
Sep-09	$\text{NO}_3$	41.1	18.4	48.4	46.2	38.7	31.4
	X	0.252	0.207	0.267	0.262	0.247	0.233
	$Y_{\min}$	9.8	4.5	12.2	11.4	9.1	7.2
	$Y_{\max}$	102.8	56.9	121.6	115.8	97.1	81.0
Oct-09	$\text{NO}_3$	40.4	19.8	47.2	46.0	35.4	29.3
	X	0.251	0.210	0.264	0.262	0.241	0.229
	$Y_{\min}$	9.6	4.8	11.8	11.4	8.2	6.7
	$Y_{\max}$	101.1	59.2	118.4	115.2	89.6	76.7
Nov-09	$\text{NO}_3$	29.8	18.9	45.8	43.8	42.7	32.8
	X	0.230	0.208	0.262	0.258	0.255	0.236
	$Y_{\min}$	6.8	4.6	11.3	10.6	10.3	7.5
	$Y_{\max}$	77.7	57.7	114.7	109.6	106.8	83.9
Dec-09	$\text{NO}_3$	37.8	20.4	43.7	40.1	37.2	29.4
	X	0.246	0.211	0.257	0.250	0.244	0.229
	$Y_{\min}$	8.8	4.9	10.6	9.5	8.7	6.7
	$Y_{\max}$	95.0	60.2	109.3	100.4	93.6	76.9

## Continued

Jan-10	NO <sub>3</sub>	29.4	18.4	36.3	39.1	32.4	32.5
	X	0.229	0.207	0.243	0.248	0.235	0.235
	Y <sub>min</sub>	6.7	4.5	8.4	9.2	7.4	7.5
	Y <sub>max</sub>	76.9	56.9	91.6	98.0	83.1	83.3
Feb-10	NO <sub>3</sub>	32.5	17.4	31.5	96.4	28.4	27.8
	X	0.235	0.205	0.233	0.363	0.227	0.226
	Y <sub>min</sub>	7.5	4.4	7.2	40.0	6.5	6.4
	Y <sub>max</sub>	83.3	55.2	81.2	304.2	74.9	73.7
Mar-10	NO <sub>3</sub>	33.6	31.7	34.7	64.4	36.4	36.7
	X	0.237	0.233	0.239	0.299	0.243	0.243
	Y <sub>min</sub>	7.7	7.3	8.0	18.9	8.5	8.5
	Y <sub>max</sub>	85.6	81.6	88.0	170.6	91.8	92.5
Apr-10	NO <sub>3</sub>	30.8	30.9	33.5	45.7	38.7	31.6
	X	0.232	0.232	0.237	0.261	0.247	0.233
	Y <sub>min</sub>	7.0	7.1	7.7	11.3	9.1	7.2
	Y <sub>max</sub>	79.7	79.9	85.4	114.4	97.1	81.4
May-10	NO <sub>3</sub>	29.7	27.6	31.9	44.6	40.4	28.8
	X	0.229	0.225	0.234	0.259	0.251	0.228
	Y <sub>min</sub>	6.8	6.3	7.3	10.9	9.6	6.6
	Y <sub>max</sub>	77.5	73.3	82.0	111.6	101.1	75.7

NO<sub>3</sub>: Nitrates concentration in wells mg/L; X: Total Nitrates intake by a human (from drinking water and other sources) g/d; Y<sub>min</sub> and Y<sub>max</sub>: Minimum and maximum life time probability of developing human gastric cancer by 100,000 persons (Equation (4)).

in all cases and its average is an overestimation of cancer probability. For example, for 45 mg/L Nitrates concentration, which is below the standard, the average cancer probability is 62 per 100,000 persons.

According to the Syrian National Cancer Registry (SNCR) [30], Tartus Governorate has the highest cancer registry in Syria “123 case per 100,000 in Syrian Population 2007”. These data are the last data given by the SNCR, because of the Syrian crisis hitting the country for the last five years. In the SNCR report, it is noted that the gastro-intestinal cancer is the second most common cancer in Syria (12% of female cases and 16% of male cases) after breast cancer (30%) and the third common cancer is lung cancer (5% of female cases and 15% of male cases).

It is better to take the (SNCR) data qualitatively rather than quantitatively, because of the lack of cases registration in hospitals and institutions that can be linked to the novelty of cancer registry in Syria, but unfortunately the crisis comes to vanish the progression of the project. What we want to say is that the SNCR cancer data probably underestimates the real data, but we still can draw

an important information is that Tartus governorate has the highest cancer rates in Syria where the most common cancers are breast cancer, gastro-intestinal cancer and lung cancer respectively.

## 6. Conclusions

Elevated Nitrates concentrations are associated with methemoglobinemia and gastro-intestinal cancers and other health effects and the risk is higher when waters are bacterially contaminated. Nitrates in Groundwater of artesian wells in six villages, located in an intensive agriculture land in Akkar plain, were analyzed during one year. We found that Nitrates concentrations are below the drinking water standard (50 mg/L) in almost all locations but they are still higher than concentration in natural unpolluted groundwater, which indicates that Nitrates sources are probably surface soil fertilizers, wastewaters and their accumulation in groundwater is quite possible. Groundwater pollution normally appears long-delayed in wells, springs and streams resulting in a very slow process of recovery of aquifer's quality once it is polluted, often during a few decades. Since groundwater moves slowly through the subsurface, the impact of anthropogenic activities may last for a relatively long time. From this point and regarding the deep depth of artesian wells (at least 100 m, see [Table 1](#)), the environmental measures should be mainly focused on the prevention of the pollution.

The associated non-carcinogenic health risks are assessed for groundwater from the studied wells based on the reference dose and hazard index. The methemoglobinemia, called blue-baby syndrome in addition to gastro-intestinal disturbance are the main non-carcinogenic risk of Nitrates. Literature review shows that elevated Nitrates concentrations are generally associated with bacterial contamination in water and form together with the principal exposure risks of methemoglobinemia. However, in Akkar plain, the sources of Nitrates are for the most part from fertilizers, while fecal coliforms come from landfills and non-treated wastewater. This is why we find that the concentrations of fecal coliforms and Nitrates are not correlated except in Autumn season because of rain water filtration into groundwater.

Carcinogenic health risk is assessed based on a combined probabilistic/fuzzy-set framework giving the cancer probability interval. The approximate cancer probability gives an indication of the role of Nitrates in gastro-intestinal cancer. These indicative values take their importance from the fact that gastro-intestinal cancer is the second most common cancer type in Syria where Tartus governorate has the highest cancer rates.

In conclusion, our study provided a characterization of potential health risk associated with groundwater Nitrates in Akkar plain in Tartus governorate in Syria. Further investigations are required to evaluate the cancer cases in Tartus governorate and associate them with environmental factors and/or other factors like diet, life style and genetic factors.

## Acknowledgements

The authors gratefully acknowledge the Environment Directorate in Tartus, for help in field measurement and laboratory analysis.

## References

- [1] Elliott, P., Hills, M., Beresford, J., *et al.* (1992) Incidence of Cancers of the Larynx and Lung near Incinerators of Waste Solvents and Oils in Great Britain. *The Lancet*, **339**, 854-858. [https://doi.org/10.1016/0140-6736\(92\)90290-J](https://doi.org/10.1016/0140-6736(92)90290-J)
- [2] Elliott, P., Shaddick, G., Kleinschmidt, I., *et al.* (1996) Cancer Incidence near Municipal Solid Waste Incinerators in Great Britain. *British Journal of Cancer*, **73**, 702-710. <https://doi.org/10.1038/bjc.1996.122>
- [3] Bhopal, R.S., Moffatt, S., Pless-Mulloli, T., *et al.* (1998) Does Living near a Constellation of Petrochemical, Steel, and Other Industries Impair Health? *Occupational & Environmental Medicine*, **55**, 812-822. <https://doi.org/10.1136/oem.55.12.812>
- [4] Rushton, L. (2003) Health Hazards and Waste Management. *British Medical Bulletin*, **68**, 183-197. <https://doi.org/10.1093/bmb/ldg034>
- [5] Tsai, S.-S., Tiao, M.-M., Kuo, H.-W., *et al.* (2008) Association of Bladder Cancer with Residential Exposure to Petrochemical Air Pollutant Emissions in Taiwan. *Journal of Toxicology and Environmental Health, Part A*, **72**, 53-59. <https://doi.org/10.1080/15287390802476934>
- [6] Porta, D., Milani, S., Lazzarino, A., Perucci, C.A. and Forastiere, F. (2009) Systematic Review of Epidemiological Studies on Health Effects Associated with Management of Solid Waste. *Environmental Health*, **8**, 60. <https://doi.org/10.1186/1476-069X-8-60>
- [7] Ranzi, A., Fano, V., Erspamer, L., Lauriola, P., Perucci, C.A. and Forastiere, F. (2011) Mortality and Morbidity among People Living Close to Incinerators: A Cohort Study Based on Dispersion Modelling for Exposure Assessment. *Environmental Health*, **10**, 22. <https://doi.org/10.1186/1476-069X-10-22>
- [8] Smargiassi, A., Goldberg, M.S., Wheeler, A.J., *et al.* (2014) Associations between Personal Exposure to Air Pollutants and Lung Function Tests and Cardiovascular Indices among Children with Asthma Living near an Industrial Complex and Petroleum Refineries. *Environmental Research*, **132**, 38-45. <https://doi.org/10.1016/j.envres.2014.03.030>
- [9] Ancona, C., Badaloni, C., Mataloni, F., Bolignano, A., Bucci, S., Cesaroni, G., Sozzi, R., Davoli, M. and Forastiere, F. (2015) Mortality and Morbidity in a Population Exposed to Multiple Sources of Air Pollution: A Retrospective Cohort Study Using Air Dispersion Models. *Environmental Research*, **137**, 467-474. <https://doi.org/10.1016/j.envres.2014.10.036>
- [10] Khan, K., Lu, Y., Saeed, M.A., *et al.* (2017) Prevalent Fecal Contamination in Drinking Water Resources and potential Health Risks in Swat, Pakistan. *Journal of Environmental Sciences*.
- [11] Mousavi, S.F., Amiri, M.J., Gohari, A.R. and Afyuni, M. (2011) Estimation of Nitrates Concentration Using Fuzzy Regression Method and Support Vector Machines. *World Applied Sciences Journal*, **12**, 774-782.
- [12] United Nations (2006) Council Directive of 12 December 2006 on the Protection of Groundwater against Pollution and Deterioration. Directive 2006/118/EC, *Official Journal of the European Union*, **L372**, 19-31.
- [13] United Nations (1991) Council Directive of 12 December 1991 Concerning the



- Protection of Waters against Pollution Caused by Nitrates from Agricultural Sources. Directive 91/676/EEC, *Official Journal of the European Union*, **L375**, 1-13.
- [14] Zimoch, I. and Paciej, J. (2016) Spatial Risk Assessment of Drinking Water Contamination by Nitrates from Agricultural Areas in the Silesia Province. *Desalination and Water Treatment*, **57**, 1084-1097.  
<https://doi.org/10.1080/19443994.2015.1043488>
- [15] Fewtrell, L. (2004) Drinking-Water Nitrate, Methemoglobinemia, and Global Burden of Disease: A Discussion. *Environmental Health Perspectives*, **112**, 1371-1374.  
<https://doi.org/10.1289/ehp.7216>
- [16] CDC (Centers for Disease Control and Prevention) (1996) Spontaneous Abortions Possibly Related to Ingestion of Nitrate-Contaminated Well Water—LaGrange County, Indiana, 1991-1994. *Morbidity and Mortality Weekly Report (MMWR)*, **45**, 569-572.
- [17] Sadler, R. Maetam, B., Edokpolo, B., et al. (2016) Health Risk Assessment for Exposure to Nitrates in Drinking Water from Village Wells in Semarang, Indonesia. *Environmental Pollution*, **216**, 738-745.
- [18] Schullehner, J., Jensen, L.L., Thygesen, M., et al. (2017) Drinking Water Nitrates Estimation at Household-Level in Danish Population-Based Long-Term Epidemiologic Studies. *Journal of Geochemical Exploration*, **183**, 178-186.  
<https://doi.org/10.1016/j.gexplo.2017.03.006>
- [19] Espejo-Herrera, N., Cantor, K.P. and Malats, N. (2015) Nitrates in Drinking Water and Bladder Cancer Risk in Spain. *Environmental Research*, **137**, 299-307.  
<https://doi.org/10.1016/j.envres.2014.10.034>
- [20] Stayner, L.T., Almberg, K., Jones, R., et al. (2017) Atrazine and Nitrate in Drinking Water and the Risk of Preterm Delivery and Low Birth Weight in Four Midwestern States. *Environmental Research*, **152**, 294-303.  
<https://doi.org/10.1016/j.envres.2016.10.022>
- [21] Winton, E.F., Tardiff, R.G. and McCabe, L.J. (1971) Nitrates in Drinking Water. *Journal American Water Works Association*, **63**, 95-98.  
<https://doi.org/10.1002/j.1551-8833.1971.tb04035.x>
- [22] USEPA (2008) Final Report: Dose-Response of Nitrates and Other Methemoglobin Inducers on Methemoglobin Levels of Infants. Washington State Department of Health, Washington DC.
- [23] Dourson, M., Stern, B., Griffin, S. and Bailey, K. (1991) Impact of Risk-Related Concerns on the U.S. Environmental Protection Agency Programs. In: Bogárdi, I., Kuzelka, R.D. and Ennenga, W.G., Eds., *Nitrates Contamination: Exposure, Consequence and Control*, Springer, Berlin, Heidelberg, 477-487.  
[https://doi.org/10.1007/978-3-642-76040-2\\_35](https://doi.org/10.1007/978-3-642-76040-2_35)
- [24] Dourson, M. and Stara, F.F. (1983) Regulatory History and Experimental Support of Uncertainty (Safety) Factor. *Regulatory Toxicology and Pharmacology*, **3**, 224-238.  
[https://doi.org/10.1016/0273-2300\(83\)90030-2](https://doi.org/10.1016/0273-2300(83)90030-2)
- [25] Lee, Y.W. (1992) Risk Assessment and Risk Management for Nitrate-Contaminated Groundwater Supplies. Ph.D. Thesis, University of Nebraska, Lincoln.
- [26] Terracini, B., Magee, P.N. and Barnes, J.M. (1967) Hepatic Pathology in Rats on Low Dietary Levels of Demethylnitrosamine. *British Journal of Cancer*, **21**, 559-565.  
<https://doi.org/10.1038/bjc.1967.65>
- [27] Dubrovsky, N.M., Burow, K.R., Clark, G.M., et al. (2010) The Quality of Our Nation's Waters—Nutrients in the Nation's Streams and Groundwater, 1992-2004. US Geological Survey, Washington DC.

- [28] Lee, Y.W., Dahab, M.F. and Bogard, I. (1995) Nitrate-Risk Assessment Using Fuzzy-Set Approach. *Journal of Environmental Engineering*, **21**, 245-256.  
[https://doi.org/10.1061/\(ASCE\)0733-9372\(1995\)121:3\(245\)](https://doi.org/10.1061/(ASCE)0733-9372(1995)121:3(245))
- [29] Shaheen, H., Al Ali, Y. and Harfoush, S. (2013) Study of Some Groundwater Pollution Indicators in the Neighbouring of Wady Alhedda Center for Solid Wastes Treatment in Tartus City. *Tishreen University Journal for Research and Scientific Studies—Engineering Sciences Series*, **35**, 284-301. (In Arabic)
- [30] Syrian National Cancer Registry SNCR (2007) National Cancer Registry. Ministry of Health.