The Effect of Chemical Fertilizers Used in Tea Agriculture on Groundwater in the Region of Rize

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ABSTRACT

In this study, water samples were taken from the drinking water of 12 villages, selected as samples from the villages in Rize province, before (April 2017), and after (August and November 2017) the fertilization period of tea farming in the area. Analysis of the nitrite and nitrate was performed on all samples, and it was seen that the nitrite and nitrate (ppm) values of the drinking waters were statistically significant (P<0.01) in relation to the pre and post-fertilization periods. The highest nitrate and nitrite values occurred in August and April, and the lowest nitrate and nitrite values were in April and August, respectively. Furthermore, there were statistically significant differences in the nitrate (P<0.01) and nitrite values (P<0.05) between all of the different villages. According to the mean results obtained, it was observed that the nitrate levels of water taken from the villages of the Çamlıhemşin (Dikkaya with 49.67 mg/L) and Kalkandere (Geçitli with 41.21 mg/L) districts are very close to the nitrate limits (50 mg/L) set by the World Health Organization, the European Community and the Environmental Protection Agency. When the nitrate values were evaluated separately in terms of periods, it was determined that the nitrate values of water taken from villages belonging to the Çamlıhemşin (59.1 mg/L) and Kalkandere (60.4 mg/L) districts in August were actually above the nitrate limits mentioned above (50 mg/L). According to the Water Pollution Control Regulation, it was determined that water samples in the villages had 3rd, and 2nd, 3rd and 4th classes of water quality in terms of nitrite and nitrate, respectively. When all of the above data is considered, the need to take measures to reduce nitrate levels in drinking water is apparent.

Keywords: Drinking Water, Chemical Fertilizers, Nitrite, Nitrate, Water Quality, Rize

Çay Tarımında Kullanılan Kimyasal Gübrelerin Rize Bölgesindeki Yeraltı Sularına Etkisi

ÖZET

Bu çalışmada; Rize ilindeki köylerden örnekleme olarak seçilen 12 köyün içme sularından çay tarımındaki gübreleme dönemi öncesi ve sonrası alınan numunelerde nitrit ve nitrat değerleri araştırıldı. Bu amaçla, gübreleme öncesi (Nisan) ve sonrası dönemlerde (Ağustos ve Kasım) belirlenen köylerden su numuneleri alındı. Çalışılan köylerin icme sularındaki nitrat (ppm) ve nitrit (ppm) değerleri gübreleme öncesi ve sonrası dönemler dikkate alındığında istatistiksel olarak çok önemli (P<0,01) farklılıklar gösterdi. En yüksek nitrat (ppm) değeri Ağustos, en düşük nitrat değeri ise Nisan aylarında gözlemlendi. Nitrit değerlerinde ise tam tersi bir durum gözlemlendi. Köyler açısından değerlendirildiğinde istatistiksel olarak nitrat ve nitrit değerlerinde sırasıyla çok önemli (P<0,01) ve önemli (P<0,05) seviyede farklılıklar tespit edildi. Elde edilen sonuçlar değerlendirildiğinde; tüm ilçelere ait köylerden alınan suların ortalama nitrit değerlerinin sınır değerlere göre düşük seviyelerde, ortalama nitrat değerlerinin ise Çamlıhemşin (49,67 mg/L ile Dikkaya Köyü) ve Kalkandere (41,21 mg/L ile Geçitli Köyü) ilçelerine ait köylerden alınan sularda Dünya Sağlık Örgütü, Avrupa Birliği ve Çevre Koruma Ajansı tarafından belirtilen sınır değerlere (50 mg/L) çok yakın olduğu gözlemlendi. Ağustos ayında Çamlıhemşin (59,1 mg/L) and Kalkandere (60,4 mg/L) ilçelerine ait köylerden alınan suların nitrat değerlerinin belirtilen sınır değerin (50 mg/L) üzerinde olduğu tespit edildi. Su Kirliliği Yönetmeliği'ne göre köylerdeki su örneklerinin nitrit açısından III. sınıf, nitrat açısından ise II., III. ve IV. sınıf su kalitesine sahip olduğu tespit edildi. Elde edilen veriler dikkate alındığında içme sularındaki nitrat seviyelerinin düşürülmesi için gerekli önlemlerin alınması gereklidir.

Anahtar Kelimeler: İçme Suyu, Kimyasal Gübreler, Nitrit, Nitrat, Su Kalitesi, Rize

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1. Introduction

The issue of water pollution has become a significant part of environmental pollution recently, which, in itself, is one of the most important factors that will determine humanity's future quality of life. The main cause of water pollution stems from the activities of industry and agriculture, of which the chemical fertilizers used in agricultural production play an important part. Much of the pollution of water due to fertilizers comes from nitrate. This is because increasing amounts of nitrate are being used in fertilizers, and this nitrate is accumulating in varying amounts, depending on conditions, both within the soil due to washing, as well as in the ground and surface waters. Numerous studies (Omurtag, 1992; Kaplan et al., 1999; Durmaz et al., 2007; Sönmez et al., 2008; Ardıç, 2013; Gemici et al., 2015; Yener and Ongun, 2017) have been conducted around the world in regard to the water pollution caused by the use of such nitrogenous fertilizers.

In fact, nitrate and nitrite are commonly found in nature in the form of additives or contaminants, and these compounds are known to have significant effects on human health. More specifically, nitrite reacts with secondary amines and other nitrogenous substances to form N-nitrosamines, which are known to cause cancer of the liver, esophagus, kidney, stomach, intestine, central nervous system and lymphoid system (Ağaoğlu et al., 2007).

Industrial wastewater, artificial fertilizers and degraded organic materials, all of which contain nitrogen, are the primary sources of nitrate and nitrite (Abercrombie and Caskey, 1972; Scorer, 1974). Nitrate enters the bodies of humans and animals through food, after which it is reduced to nitrite and ammonia due to the effect of microorganisms in the stomach. The excess nitrate is absorbed together with this nitrite and ammonia in the stomach to enter the bloodstream, thus converting the oxyhemoglobin in the blood to methemoglobin. As methemoglobin cannot carry oxygen, nitrite

poisoning is the result. Nitrite is oxidized to nitrate and eliminated by urine (Parsons, 1978). While nitrate at certain levels can always be found in water and is harmless to adults, continuous drinking of water containing more than 20 mg of nitrate per liter leads to acute and chronic poisoning in humans, and methemoglobinemia causes bruising in infants under 6 months (Stahr, 1977).

Nitrate and nitrite are formed by the decomposition of organic substances of human and animal origin. The presence of these compounds in water is indicative of bacterial contamination. In recent years, due to population growth and industrialization, the likelihood and amount of these substances in water have increased. Industrial wastewater, artificial fertilizers and degraded organic materials, all of which contain nitrogen, are the main sources of nitrate and nitrite (Abercrombie and Caskey, 1972; Scorer, 1974).

It is known that much of the potable water in Turkey is supplied by groundwater. It is also reported that this water is exposed to pollution as a result of leaks in dams and solid waste storage areas, as well as from industrial, agricultural and livestock waste (Tuncay, 1994).

The aim of this study is to determine whether the nitrite and nitrate values of drinking water in selected villages in Rize province are a threat to human health and whether chemical fertilizers used in tea farming are the cause of nitrite and nitrate pollution in groundwater during the pre and post-fertilization period.

2. Materials and Methods

2.1. Site Description, Sampling and Analysis

This research was conducted in Rize, which is located in the Eastern Black Sea Region. Twelve different locations were chosen as part of the study of the impact of chemical fertilizers on water quality and to determine the quality of drinking water. Based on the results of previous annual analyzes, the villages with the highest value were selected. The geographic coordinates of all localities were recorded using a portable global positioning system (Garmin GPSmap 62sc, Garmin International, Olathe, KS). Details of these localities and sampling dates are provided in Table 1.

Water samples were taken from the drinking water of 12 selected villages (1 village representing each district) in Rize province before (April) and after (August and November) the tea farming fertilization period. 36 drinking water samples were collected from 12 different villages (Karasu, Gürgen, Ihlamur, Geçitli, Çakmakçılar, Büyükçiftlik, İncesırt, Boğazlı, Yaltkaya, Dikkaya, Ortaalan and Ihlamurlu) in April, August and November 2017. The samples were taken in a 500 ml sterile bottle and were brought to the laboratory in the cold chain. Nitrate and nitrite analyses were performed on all samples. Test kits specific (Hach LCK 340 test kit for nitrate and Hach LCK 341 test kit for nitrite) to each of the nitrate and nitrite measurements were used and, the protocol written in each test kit was applied. Nitrate and nitrite concentrations were measured using a spectrophotometer (Hach DR 3900) at the wavelengths (340 nm for nitrate and 535 nm for nitrite) as written in the protocol.

Table 1. The geographic coordinates and sampling dates of all localities

District	Village	Latitude	Longitude	1 st Sampling Date	1 st Sampling Date	1 st Sampling Date
Merkez	Karasu Village	40° 53' 37"	40° 34' 10"	28.04.2017	17.08.2017	22.11.2017
Güneysu	Gürgen Village	40° 57' 12"	40° 39' 23"	28.04.2017	17.08.2017	20.11.2017
İkizdere	Ihlamur Village	40° 47' 47"	40° 31' 55"	28.04.2017	16.08.2017	20.11.2017
Kalkandere	Geçitli Village	40° 57' 19"	40° 25' 34"	28.04.2017	17.08.2017	20.11.2017
Derepazari	Çakmakçılar Village	40° 0' 18"	40° 27' 0"	28.04.2017	16.08.2017	22.11.2017
İyidere	Büyükçiftlik Village	40° 59' 13"	40° 23' 32"	28.04.2017	16.08.2017	22.11.2017
Pazar	Boğazlı Village	41° 8' 51"	40° 55' 36"	28.04.2017	17.08.2017	21.11.2017
Hemşin	Yaltkaya Village	41° 4' 18"	40° 53' 49"	28.04.2017	17.08.2017	21.11.2017
Çamlıhemşin	Dikkaya Village	41° 5' 0"	41° 1' 46"	28.04.2017	17.08.2017	21.11.2017
Ardeşen	Ortaalan Village	41° 10' 56"	41° 5' 52"	28.04.2017	17.08.2017	20.11.2017
Fındıklı	Ihlamurlu Village	41° 12' 45"	41° 13' 31"	28.04.2017	17.08.2017	21.11.2017
Çayeli	İncesırt Village	41° 5' 38"	40° 44' 22"	28.04.2017	16.08.2017	22.11.2017

2.2. Statistical analysis

Statistical analysis was performed with SPSS version 21 (IBM SPSS Statistics for Windows, Armonk, NY). One-way analysis of variance (ANOVA) was performed to reveal the differences in the nitrate and nitrite values of water samples, according to localities and months, and Tukey's HSD Test was performed on significant results using the SPSS software. All samples were measured in triplicate.

3. Results

3.1. Nitrite and Nitrate (ppm) Concentrations according to Pre and Post-Fertilization Periods

The nitrite and nitrate (ppm) values in the drinking water of the studied villages showed statistically significant differences (P<0.01) in terms of pre and post-fertilization periods (Table 2). The highest and lowest nitrite (ppm) values were observed in April and August, respectively, whereas the opposite situation occurred in nitrate values (Figure 1).



Figure 1. Nitrite and nitrate (ppm) concentrations according to pre and post-fertilization periods

Table 2. Results obtained with Tukey's HSE)
test that reveal nitrite and nitrate concentrations	3
according to pre and post-fertilization periods	

Month	Nitrite (ppm)	Nitrate (ppm)
April	0.047±0.0027 a	12.77±1.86 b
August	0.029±0.0020 b	29.22±3.17 a
November	0.032±0.0020 b	21.55±2.53 a

The different letters denote the difference between the groups, according to Tukey's HSD test (rejection level 0.05).

3.2. Nitrite and Nitrate (ppm) Concentrations of Water in Villages

The nitrate and nitrite (ppm) content of the drinking water obtained from the villages studied showed statistically significant differences (P<0.01) and (P<0.05), respectively (Table 3). The highest nitrite (ppm) values were observed in Yaltkaya village, while the lowest nitrite values were in İncesırt and Geçitli villages. The highest and lowest nitrate concentrations were in Dikkaya and Ihlamur villages, respectively (Figure 2).



Figure 2. Nitrite and nitrate (ppm) concentrations according to villages

Village	Nitrite (ppm)	Nitrate (ppm)
Karasu Village	0.03556 a	6.8178 d
Gürgen Village	0.03078 a	25.4711 bc
Ihlamur Village	0.02633 a	5.8489 d
Geçitli Village	0.02456 a	41.2111 ab
Çakmakçılar Village	0.03933 a	22.1267 cd
Büyükçiftlik Village	0.04311 a	17.6333 cd
Boğazlı Village	0.04300 a	27.7067 bc
Yaltkaya Village	0.04600 a	6.5356 d
Dikkaya Village	0.04222 a	49.6667 a
Ortaalan Village	0.03367 a	11.5333 cd
Ihlamurlu Village	0.03944 a	11.8200 cd
İncesırt Village	0.02456 a	27.8122 bc

 Table 3. Results obtained with Tukey's HSD test that determine nitrite and nitrate concentrations according to villages

The different letters denote the significant differences between the groups, according to Tukey's HSD test (rejection level 0.05). Means followed by the same letter are not significantly different at the 0.05 level, as per the results obtained using Tukey's HSD test.

4. Discussion

This study includes the results of the observed changes of nitrite and nitrate parameters in the water samples taken from the drinking water of 12 villages in the districts of Rize before (April), and after (August and November) the fertilization period. According to the results of the study, the highest and lowest mean nitrite values were 0.046 ppm recorded in Yaltkaya village, and 0.02456 ppm recorded in Incesirt and Gecitli villages, respectively. When the nitrite values were analyzed periodically, the highest and lowest mean nitrite values were 0.047 ppm recorded in April, and 0.029 ppm recorded in August, respectively. Taş (2011) reported that the average amount of nitrite in Gaga Lake was 0.03 mg/L. Similarly, Bulut and (2019) found that the nitrite Kubilay concentration of Eğirdir lake was 0.021-0.074 mg/L (mean: 0.032 mg/L). Seasonal average nitrite levels in Karagöl were found to be 0.001 mg/L, 0.002 mg/L, 0.005 mg/L, and 0.004 mg/L in winter, spring, summer and fall, respectively (Mutlu et al., 2013). Atea (2017) pointed out that the lowest nitrite level was 0, and the highest was 0.008 mg/L recoded in the waters of Germectepe dam. The lowest concentration of nitrite in the Coruh River was determined to be 0.00 mg/L in the spring season of 2013 at the

Bayburt exit point, and highest the concentration was measured to be 0.045 mg/L in the summer season of 2013 at the same point by Birici et al. (2017). In addition to this, Dede and Sezer (2017) found that the lowest nitrite level in the samples taken to determine the water quality of the Aksu Stream, which is one of the most important sources of drinking water in Giresun province, was 0 mg/L and the highest level was 0.4 mg/L. The nitrite concentration in Cip Dam Lake varied between 0.02 mg/L and 0.14 mg/L. The amounts analyzed in spring were higher than those in winter. The highest value (0.14 mg/L) was recorded in May. This indicates that the amount of organic matter in the lake increases towards the summer season (Gültürk, 2018). Özel et al. (2019) stated that the average annual amount of nitrite in Isparta Stream varied from 0.035 mg/L to 4.168 mg/L. Verep et al. (2017) also found that the nitrite values in Derepazarı Stream (Rize) were between 0.002 mg/L and 0.02 mg/L (mean: 0.006 mg/L). Verep et al. (2019) reported that the nitrite values in Salarha basin streams were 0.033 mg/L (0-0.91 mg/L). Tepe and Kutlu (2019) pointed out that the nitrite nitrogen values in Karkamış Dam Lake were recorded to be 0.001-0.053 µg/L in surface water, 0.001- $0.049 \ \mu g/L$ at a depth of 4m, and 0.001-0.029

 μ g/L at a depth of 8m. Nitrite values obtained from the study were mostly similar to ones in the literature. Although increases and decreases have been observed during some months and in some villages, nitrite data has generally been stable in the villages. Although there were statistical differences between the villages in the nitrite values of the water samples, the nitrite values of each village were statistically gathered in a single group since they were close to each other. Nitrogen compounds are expected to turn into nitrite in surface waters and nitrite, an unstable compound, to eventually nitrate into oxide (Teksoy et al., 2019). In addition, nitrite is an intermediate product of the nitrogen cycle, it does not accumulate in the environment; it immediately turns into nitrate (Taş, 2011).

The highest mean nitrate value of drinking water was determined to be 49.67 ppm in Dikkaya village, and the lowest nitrate mean of 5.85 ppm was found in Ihlamur village. When the nitrate values were examined periodically, the highest mean nitrate value of 29.22 ppm was observed in August, and the lowest nitrate mean of 12.77 ppm was observed in April. Similarly, Kaplan et al. (1999) reported that the nitrate concentration of 16-20 mg/L in a well drilled in the Bursa Plain increased up to 110-150 mg/L during the fertilization season. The findings of Bulut and Kubilay (2019) indicated that nitrate concentration in Eğirdir Lake ranged from 0.4-2.9 mg/L (mean: 1.2 mg/L). Gültürk (2018) stated that the nitrate concentration varied between 1.2 mg/L and 2.15 mg L, and that it was higher in spring than in other months. Verep et al. (2017) found that the nitrate values in the Derepazarı Stream (Rize) were between 1.63 mg/L and 7.08 mg/L (mean: 4.66 mg/L). Verep et al. (2019) reported that the nitrate values in Salarha basin streams were 1.73 mg/L (0.1-8.3 mg/L). Tepe and Kutlu (2019) pointed out that the nitrate nitrogen values in Karkamış

Dam Lake were determined to be between 1.54-3.47 mg/L in surface water, 1.54-3.36 mg/L at a depth of 4 m, and 1.15-3.19 mg/L at a depth of 8 m. Nitrate, the most important of nitrogen compounds bound in surface waters, emerges as a result of the complete oxidation of nitrogen compounds (Gültürk, 2018). Indeed, our results were found to confirm the view that nitrite and nitrate pollution might be related to fertilization. Eryurt and Sekin (2011), Aslan and Akkaya (2001), Aras and Findik (2018), Minareci and Sungur (2019) also reported that fertilization and agricultural activities are the main sources of nitrate pollution in waters. Supporting this notion, the uncontrolled use of high amounts of chemical fertilizers and pesticides used in agricultural activities may result in nitrate accumulation (Keskin, 2009). It can be clearly seen from these studies that excessive fertilizer use and uncontrolled use of agricultural chemicals pose an important risk to surface and groundwater. According to Chapman and Kimstach (1996), if the nitrate-nitrogen content in water is above 5 mg/L, this is due to agricultural activities or domestic wastes (Özel et al., 2019).

to Water Pollution Control According Regulations, nitrite levels in the surface water of the continent are classified as Class 1 (highquality waters) if the level is 0.002 ppm, Class 2 (slightly contaminated waters) if it is 0.01 ppm, Class 3 (contaminated waters) if it is 0.05 ppm, and Class 4 (highly contaminated waters) if it is above 0.05 ppm. According to this classification, the water of the villages (mean nitrite 0.0357 ppm) were of fourth class in terms of the nitrite level (Table 4). On the other hand, these results are below the nitrite limits determined by the World Health Organization, the European Community and the US Environmental Protection Agency (Table 5).

	Village	Ν	Mean	Class*
	Karasu Village	9	0.035	3 rd
	Gürgen Village	9	0.030	3 rd
	Ihlamur Village	9	0.026	3 rd
	Geçitli Village	9	0.024	3 rd
	Çakmakçılar Village	9	0.039	3 rd
	Büyükçiftlik Village	9	0.043	3 rd
Nitrite	Boğazlı Village	9	0.043	3 rd
	Yaltkaya Village	9	0.046	3 rd
	Dikkaya Village	9	0.042	3 rd
	Ortaalan Village	9	0.033	3 rd
	Ihlamurlu Village	9	0.039	3 rd
	İncesırt Village	9	0.024	3 rd
	Total	108	0.035	3 rd
	Karasu Village	9	6.817	2^{nd}
	Gürgen Village	9	25.471	4^{th}
	Ihlamur Village	9	5.848	2^{nd}
	Geçitli Village	9	41.211	4^{th}
	Çakmakçılar Village	9	22.126	4^{th}
	Büyükçiftlik Village	9	17.633	3 rd
Nitrate	Boğazlı Village	9	27.706	4^{th}
	Yaltkaya Village	9	6.535	2^{nd}
	Dikkaya Village	9	49.666	4^{th}
	Ortaalan Village	9	11.533	3 rd
	Ihlamurlu Village	9	11.820	3 rd
	İncesırt Village	9	27.812	$4^{ ext{th}}$
	Total	108	21.181	4^{th}

Table 4. The nitrite and nitrate	(ppm)) concentration	of the	villages
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*Classes Water pollution control regulation related tables were taken into consideration (URL-1).

Table 5. The upper limits of nitrite and nitrate parameters according to international standards (Atea et al., 2017)

	WHO (2008)	EPA (2009)	EC (1998)
Nitrite (mg/L)	0.5	0.5	0.5
Nitrate (mg/L)	50	50	50

According to the Water Pollution Control Regulation, nitrate levels in the surface waters of the continent were classified as Class 1 if the level is 5 ppm, Class 2 if the level is 10 ppm, Class 3 if the level is 20 ppm and Class 4 if the level is above 20 ppm. According to this

of both localities and fertilization periods. It was determined that the water samples in the villages had 3rd, and 2nd, 3rd, and 4th classes of

classification, the water of the villages (mean nitrate 21.18 ppm) were of fourth class in terms of the nitrate level (Table 4). On the other hand, these results are below the nitrate limits determined by the World Health Organization, the European Community and the US Environmental Protection Agency (Table 5).

When all the mean results are evaluated, it was

found that there was a statistically significant

difference in nitrite and nitrate values in terms

water quality in terms of nitrite and nitrate, respectively. Similarly, the water of the Ayvali Creek was determined as being 3rd and 4th Class according to the class and limit values of the Water Quality Regulation and Regulation Concerning Water Intended for Human Consumption (TS-266) (Akay et al., 2019). The average concentrations of water nitrite were determined to be of 4th class (Aras and İpek, 2019). The reason of this pollution may be caused by the release of fertilizer packaging waste, by the contamination of rainwater and groundwater by fertilizers, and due to the use of excess fertilizer (Ataseven, 2011). The most important agricultural activities that lead to the pollution of the groundwater were concluded to be the use of pesticides and fertilizers and the direct disposal of animal wastes into the soil. These substances can easily be spread by water into the soil and be the cause of significant pollution (Özdemir, 2006). Generally, the presence of more than 5-10 mg/L of nitrate in indicates contamination water bv an anthropogenic source (Davraz and Ünver, 2014). It can be said that there may be pollution in the villages determined in Rize province as a result of domestic and agricultural activities, and that this pollution may be at a level that threatens the water supply.

5. Conclusion

In conclusion, when the mean results were evaluated, it was observed that the nitrate values of the water samples taken from the villages of Çamlıhemşin (49.67 mg/L along with Dikkaya village) and Kalkandere (41.21 mg/L along with Geçitli village) districts were very close to the nitrate limits (50 mg/L) reported by the World Health Organization, the European Community and the US Environmental Protection Agency. However, when the nitrate values of the studied villages were evaluated separately in terms of periods, the nitrate values of the waters taken from the villages of Çamlıhemşin (59.1 mg/L with Dikkaya village) and Kalkandere (60.4 mg/L with Gecitli village) districts in August (the period after fertilization) were above the

nitrate limits (50 mg/L) reported by the World Health Organization, the European Community and the US Environmental Protection Agency. Similarly, Demirarslan and Başak (2019) recorded a nitrate value of above 50 ppm, which is the limit, in the City Campus.

Suggestion

Since the nitrate values in Çamlıhemşin and Kalkandere districts were above the limit values, in order to reach a more precise and accurate result, the boundaries of the study should be expanded, and different villages in these districts should be included. Furthermore, the number and frequency of analyses should be increased by taking samples from different points. This would allow more detailed conclusions to be drawn about drinking water in the villages of these two districts. Given the fact that water quality is an important issue worldwide, it is of vital importance to take precautions in order to reduce the nitrate and nitrite levels in drinking water.

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