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Research Article

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Variations in Nitrogen (NH_4^+, NO_2^-, NO_3^-) and Heavy Metal (Al and Cu) Levels of Water from Swimming Pools in the City Center and Districts of Canakkale, Turkey

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Abstract

In this study, some nitrogen (NO_2^-, NO_3^-, NH_4^+) and metal (Al and Cu) concentrations were analyzed during the summer period by using an ultraviolet-visible (UV-VIS) spectroscopy method in a total of 44 swimming pools located in the province and districts of Canakkale (Turkey). Merck Kits equivalent to EPA, APHA, ISO and DIN standards were used for spectrometric analyses. The monthly quality parameter results in this study were evaluated according to various limit standard values for different countries. As a result, NH_4^+, NO_2^-, NO_3^-, Al and Cu concentrations varied between 0.003 and 0.999 mg L^{-1} (0.073 ± 0.139 mg L^{-1}), 0.042 and 0.277 mg L^{-1} (0.086 ± 0.039 mg L^{-1}), 0.054 and 76.844 mg L^{-1} (12.111 ± 10.487), 0 and 0.652 mg L^{-1} (0.141 ± 0.068 mg L^{-1}) and 0 and 2.216 mg L^{-1} (0.139 ± 0.163 mg L^{-1}), respectively. Although all the maximum values, except for NO_2^-, exceeded the limit values of the Turkish Health Ministry (THM), all the average values were under the limit values of THM.

Keywords: Canakkale; Swimming pool water; Ammonium; Nitrite; Nitrate; Cu; Al

Introduction

Swimming is one of the most popular aquatic activities in industrialized countries. During busy periods, the quality of pool water may be compromised. Indeed, swimmers bring microorganisms and organic substances (saliva, sweat, cosmetics, sunscreen and urine) with them into the water, which strongly contribute to water contamination [1-3].

Ammonia in water is an indicator of bacterial, fecal and animal wastes being mixed into the water [4]. In pool water NH_4^+ may come from the filling water, have human origin (fecal and urine contamination) and be caused by biofilm formation [5]. Ammonium is quickly oxidized to nitrite in an oxygen environment and then transformed into nitrate. Nitrification is carried out by bacteria such as *Nitrosomonas* and *Nitrobacter* group bacteria [6]. Nitrite, nitrate and ammonium are found in lower concentrations compared to water or not often seen, because nitrite is an intermediate nitrogen converted into ammonium during nitrification or de-nitrification [7].

Organic substances which cause water discoloration are usually removed from water by coagulation (clotting), precipitation and filtration [8]. Before filtration, the stability of small particles (electrical) is deteriorated thanks to the addition of flock material into pool water and hence agglomeration of particles is ensured. For flocking, chemical (e.g., addition of colloidal substances such as $Al_2(SO_4)_3$ to water) and electro-physical processes (e.g., addition of copper ions to the water) are used [9]. Flocculation decreases chlorine consumption in the pool and hence eye diseases and BK formation which causes bad smells are avoided [10]. Aluminum (Al) is known to be a non-toxic element. However, excessive amounts of intake have been shown to influence the nervous system and cause anemia. Al is an element also related to Alzheimer and diabetes diseases [11].

Copper (Cu) which is an important metal can be transported into pool water by using cheaper algal drugs and filling water. It creates problems such as green hair and nails [5]. Cu contamination in swimming pools also comes from natural water sources which are used for pool waters [12].

Limit concentration values of the Turkish Health Ministry for NO_2^-, NO_3^-, NH_4^+, Al and Cu are 0.50, 50.0, 0.50, 0.20 and 1.00 mg L^{-1}, respectively [13]. However, there are some differences in limit values of NO_3^- in different countries. For example, while the NO_3^- limit value is 20.0 mg L^{-1} in Italy and Germany according to the Italian standard (2013) and German DIN 19643 (2012) standard, it is 30 mg L^{-1} in Austria according to the Austria Hygiene Regulations. Although the NO_3^- limit value in the Netherlands Standard (1969) is similar to the Turkish Standard value (50.0 mgL^{-1}), there is no limit value in France and England [14].

In order to evaluate some quality standard levels of swimming pools found in the province and the districts of Canakkale (Turkey), this study on variations of nitrogen (NH_4^+, NO_2^-, NO_3^-) and heavy metal (Al and Cu) levels was conducted in the summer period (July, June and August 2013) due to the intense tourism activity in this period.

Material and Methods

Study area and period

In this study, 44 swimming pools (Figure 1) were selected as the study area. Pool water samples were collected monthly from sampling locations (swimming pools) in the summer of 2013 (June, July and August, 2013) in Canakkale province and districts (Figure 1). The sample numbers 1-13 are from Ayvacik-Kucukkuyu, 14-16: Bozcaada, 17: Ezine, 18: Eceabat, 19: Gelibolu, 20-21: Çan, 22-23: Yenice, 24-25: Biga, 26-32: Çanakkale, 33-36: Ayvacik-Assos, 37-44: Ayvacık-Kadırga (Figure 1).

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Figure 1: Water sampling points for swimming pools in the summer period in the provinces and the districts of Canakkale, Turkey (Sample numbers 1-13: Ayvacik-Kucukkuyu, 14-16: Bozcaada, 17: Ezine, 18: Eceabat, 19: Gelibolu, 20-21: Çan, 22-23: Yenice, 24-25: Biga, 26-32: Çanakkale, 33-36: Ayvacik-Assos, 37-44: Ayvacık-Kadırga).

Pool water samples were collected from special points where the water flow is lowest or swimmers are the most crowded. Moreover, special sampling depths and points were just under surface (0.20 m) and about one meter away from the edge line of the pools. The collected samples were put in clean polythene sample bottles (1.00 L) and stored in deep freezer at -21.0°C until analysis. In this study, a total of 132 samples from the 44 swimming pools were analyzed for each parameter.

Analytical methods

In this study, some nitrogen (NO_2^-, NO_3^-, NH_4^+) and metal (Al and Cu) concentrations were analyzed in the summer period by using ultraviolet-visible (UV-VIS) spectroscopy method in a total of 44 swimming pools found in the province and districts of Canakkale (Turkey). Thermo Aquamate Brand Spectrophotometer was used for analyses. For analyses, Merck Kits equivalent to EPA, APHA, ISO and DIN standards were used.

Ammonium nitrogen (NH_4^+-N) occurs partly in the form of ammonium ions and partly as ammonia. A pH-dependent equilibrium exists between the two forms. In strongly alkaline solutions ammonium nitrogen is present almost entirely as ammonia, which reacts with hypochlorite ions to form monochloramine. This, in turn, reacts with a substituted phenol to form a blue indophenol derivative that is determined photometrically. The method is analogous to EPA 350.1, APHA 4500-NH3 F, ISO 7150-1, and DIN 38406-5. Analysis measuring range is from 0.01 to 2.00 mg L^{-1} NH_4^+ -N and 0.01 to 2.58 mg L^{-1} NH_4^+. In the production control, the following data were determined in accordance with ISO 8466-1 and DIN 38402 A51: standard deviation of the method ± 0.0146 (mg L^{-1} NH_4^+-N), coefficient of variation of the method ± 1.40 (%), confidence interval ± 0.035 (mg L^{-1} NH_4^+-N), sensitivity: absorbance 0.010 A corresponds to 0.009 (mg L^{-1} NH_4^+-N), accuracy of a measurement value max ± 0.052 (mg L^{-1} NH_4^+-N) [15].

In acidic solutions nitrite ions react with sulfanilic acid to form a diazonium salt, which in turn reacts with N-(1-naphthyl) ethylenediamine dihydrochloride to form a red-violet azo dye. This dye is determined photometrically. The method is analogous to EPA 354.1, APHA 4500-NO_2^- B, and DIN EN 26777. Analysis measuring range was 0.010 to 0.700 mg L^{-1} NO_2^--N and 0.03 to 2.30 mg L^{-1} NO_2^- unless otherwise stated. In the production control, the following data were determined in accordance with ISO 8466-1 and DIN 38402 A51: standard deviation of the method ± 0.0028 (mg L^{-1} NO_2^--N), coefficient of variation of the method ± 0.84 (%), confidence interval ± 0.008 (mg L^{-1} NO_2^--N), sensitivity: absorbance 0.010 A corresponds to 0.003 (mg L^{-1} NO_2^--N), accuracy of a measurement value max. ± 0.0010 (mg L^{-1} NO_2^--N) [15].

In strongly sulfuric solutions, in the presence of chloride nitrate ions react with resorcinol to form a red-violet indophenol dye that is determined photometrically. Analysis measuring range is 0.10 to 3.00 mg L^{-1} NO_3^--N and 0.40 to 13.3 mg L^{-1} NO_3^- unless otherwise stated. In the production control, the following data were determined in accordance with ISO 8466-1 and DIN 38402 A51: standard deviation of the method ± 0.028 (mg L^{-1} NO_3^--N), coefficient of variation of the method ± 1.90 (%), confidence interval ± 0.007 (mg L^{-1} NO_3^--N), sensitivity: absorbance 0.010 A corresponds to 0.01 (mg L^{-1} NO_3^--N), accuracy of a measurement value max ± 0.09 (mg L^{-1} NO_3^--N) [15].

In weakly acidic, acetate-buffered solutions aluminum ions react with chromazurol S to form a blue-violet compound that is determined photometrically. The method is analogous to APHA 3500-Al-B and DIN ISO 10566 E30. Analysis measuring range is 0.02 to 2.00 mg L^{-1} Al unless otherwise stated. In the production control, the following data were determined in accordance with ISO 8466-1 and DIN 38402 A51 (10-mm cell): standard deviation of the method ± 0.012 (mg L^{-1} Al), coefficient of variation of the method ± 1.70 (%), confidence interval ± 0.03 (mg L^{-1} Al), sensitivity: absorbance 0.010 A corresponds to 0.001 (mg L^{-1} Al), accuracy of a measurement value max. ± 0.008 (mg L^{-1} Al) [15].

In an ammoniacal medium, copper (II) ions react with cuprizone to form a blue complex that is determined photometrically. Analysis measuring range is 0.05 to 8.00 mg L^{-1} Cu unless otherwise stated. In the production control, the following data were determined in accordance with ISO 8466-1 and DIN 38402 A51: standard deviation of the method ± 0.034 (mg L^{-1} Cu), coefficient of variation of the method ± 0.84 (%), confidence interval ± 0.08 (mg L^{-1} Cu), sensitivity: absorbance 0.010 A corresponds to 0.04 (mgL^{-1} Cu), accuracy of a measurement value max ± 0.14 (mgL^{-1} Cu) [15].

Results and Discussion

Nitrogen (NH_4^+, NO_2^- and NO_3^-) variations

Nitrogen (NH_4^+, NO_2^- and NO_3^-) variations in swimming pool waters in the summer period in the province and districts of Canakkale, Turkey are given in Table 1 and Figures 2-4.

Average summer results showed that NH_4^+, NO_2^- and NO_3^- concentrations varied between 0.003 and 0.999 mg L^{-1} (0.073 ± 0.139 mgL^{-1}), 0.042 and 0.277 mg L^{-1} (0.086 ± 0.039 mgL^{-1}) and 0.054 and 76.844 mgL^{-1} (12.111 ± 10.487), respectively. Based on both maximum and average values, nitrogen (NH_4^+, NO_2^- and NO_3^-) concentrations, except for NO_3^-, increased from June to August probably due to the increasing tourist population. However, NO_3^- reached a maximum in July based on maximum and average concentrations (Table 1). However, although all the maximum nutrient values, except for NO_2^-, were over the limit values for pool water standards of the Turkish Health Ministry and the standards of some European countries, all the average values were under these limit values (Table 1).

Figure 2 shows that NH_4^+ concentrations varied between 0.0102 and 0.9992 mg L^{-1} during the sampling period. Based on the limit values for pool water standards of the Turkish Health Ministry and the standards of some European countries, concentration values such as 0.7563 (in

Month 2013	Parameters (mg L^{-1})	*n*	Maximum Values	Minimum Values	Average Values	Standard Deviation	Analysis Accuracy	Limit Values	*l*
June	NH_4^+	44	0.4425	0.0102	0.0460	0.0658	± 0.059	0.50	0
	NO_2^-	44	0.2162	0.0417	0.0742	0.0315	± 0.040	0.50	0
	NO_3^-	44	36.206	0.4554	10.869	8.6847	± 0.039	50.0	0
July	NH_4^+	44	0.9639	0.0032	0.0756	0.1541	± 0.059	0.50	1
	NO_2^-	44	0.2381	0.0468	0.0814	0.0349	± 0.040	0.50	0
	NO_3^-	44	76.844	0.4192	12.973	13.815	± 0.039	50.0	2
August	NH_4^+	44	0.9992	0.0123	0.0967	0.1983	± 0.059	0.50	3
	NO_2^-	44	0.2767	0.0520	0.1025	0.0496	± 0.040	0.50	0
	NO_3^-	44	50.786	0.0536	12.492	8.9622	± 0.039	50.0	1
Summer Average	NH_4^+	44	0.9992	0.0032	0.0728	0.1394	± 0.059	0.50	4
	NO_2^-	44	0.2767	0.0417	0.0860	0.0387	± 0.040	0.50	0
	NO_3^-	44	76.844	0.0536	12.111	10.487	± 0.039	50.0	3

Table 1: Descriptive statistical results for NH_4^+, NO_2^- and NO_3^- concentrations in swimming pools in the summer period in province and districts of Canakkale, Turkey (*n*: Number of observations; *l*: number of samples exceeding the limit concentration values specified by Turkish Health Ministry).

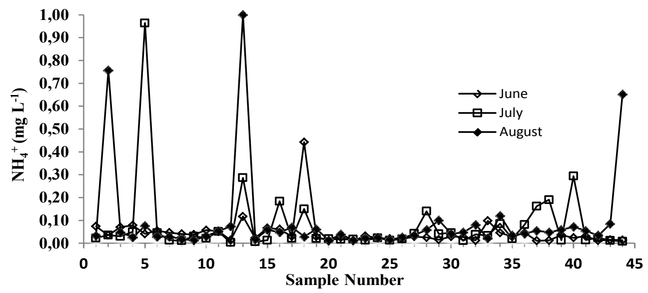

Figure 2: Variations of NH_4^+ in swimming pool waters in the summer period in province and districts of Canakkale, Turkey (Sample numbers 1-13: Ayvacik-Kucukkuyu, 14-16: Bozcaada, 17: Ezine, 18: Eceabat, 19: Gelibolu, 20-21: Çan, 22-23: Yenice, 24-25: Biga, 26-32: Çanakkale, 33-36: Ayvacik-Assos, 37-44: Ayvacık-Kadırga).

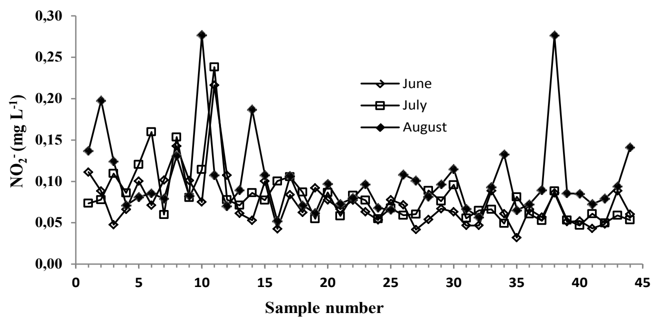

Figure 3: Variations of NO_2^- in swimming pool waters in the summer period in province and districts of Canakkale, Turkey (Sample numbers 1-13: Ayvacik-Kucukkuyu, 14-16: Bozcaada, 17: Ezine, 18: Eceabat, 19: Gelibolu, 20-21: Çan, 22-23: Yenice, 24-25: Biga, 26-32: Çanakkale, 33-36: Ayvacik-Assos, 37-44: Ayvacık-Kadırga).

Ayvacik-Kucukkuyu), 0.9639 (in Ayvacik-Kucukkuyu), 0.9992 (in Ayvacik-Kucukkuyu), and 0.6513 mgL^{-1} (in Ayvacik-Assos-Kadirga) for NH_4^+ were considerably higher than limit values. The percentage exceeding standard limit values (unhealthy pool water ratio) of Turkish Health Ministry for NH_4^+ was calculated as 3.03%. All the concentration values of NH_4^+ over or exceeding the limit values were found in pool waters from Ayvacik, Canakkale, Turkey [16,17].

Findings revealed that NO_2^- concentrations varied between 0.0417 and 0.2767 mg L^{-1} during the sampling period. Although in Ayvacik, some NO_2^- concentrations in July and August were high (>0.20), all the concentrations were under limit values for pool water standards of

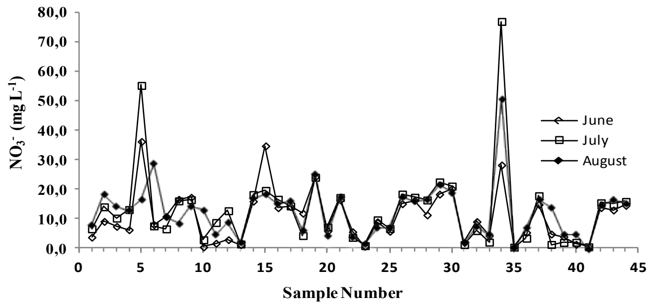

Figure 4: Variations of NO_3^- in swimming pool waters in the summer period in the provinces and the districts of Canakkale, Turkey (Sample numbers 1-13: Ayvacik-Kucukkuyu, 14-16: Bozcaada, 17: Ezine, 18: Eceabat, 19: Gelibolu, 20-21: Çan, 22-23: Yenice, 24-25: Biga, 26-32: Çanakkale, 33-36: Ayvacik-Assos, 37-44: Ayvacık-Kadırga).

Turkish Health Ministry and the standards of some European countries. Unhealthy pool water ratio for NO_2^- was calculated as 0.00%.

Results revealed that NO_3^- concentrations varied between 0.0536 and 76.844 mgL^{-1} during the sampling period. The study findings for NO_3^- also showed that although there was a few NO_3^- concentration values (3 samples, 2.27% of the total) exceeding limit standard values of Turkish Health Ministry and the Netherlands (50.0 mg L^{-1}), there were a number of NO_3^- values (14 samples, 10.61% of the total) exceeding limit standard values (20.0 mg L^{-1}) in Italy and Germany according to the Italian standard (2013) and German DIN 19643(2012) standard (Figure 3). NO_3^- concentrations over 20.0 mg L^{-1} were found in Canakkale provincial centre (4 samples, 3.03% of the total), Gelibolu (3 samples, 2.27% of the total), Bozcaada (1 samples, 0.75% of the total), Ayvacik (6 samples, 4.55% of the total), Ayvacık-Kucukkuyu (3 samples, 2.27% of the total) and Ayvacık-Assos-Kadırga (3 samples, 2.27% of the total). As with ammonium (NH_4^+) concentrations, the highest concentration values for nitrate (NO_3^-) were from the Ayvacik district more than any other district. Unhealthy pool water ratio for NO_3^- was calculated as 10.61%.

Metal (Al and Cu) variations

Metal (Al and Cu) variations in swimming pool waters in the summer period in the province and districts of Canakkale, Turkey are given in Table 2 and Figures 5 and 6.

Average summer results showed that Al and Cu concentrations varied between 0 and 0.652 mg L^{-1} (0.141 ± 0.068 mg L^{-1}) and 0 and

Month 2013	Parameters (mg L⁻¹)	*n*	Maximum Values	Minimum Values	Average Values	Standard Deviation	Analysis Accuracy	Limit Values	*l*
June	Al	44	0.3596	0	0.0325	0.0529	± 0.008	0.20	1
	Cu	44	0.2974	0.0513	0.1127	0.0690	± 0.140	1.00	0
July	Al	44	0.6522	0	0.0500	0.0956	± 0.008	0.20	1
	Cu	44	0.5236	0	0.1165	0.0933	± 0.140	1.00	0
August	Al	44	0.3736	0	0.3390	0.0543	± 0.008	0.20	1
	Cu	44	2.2162	0.0549	0.1884	0.3279	± 0.140	1.00	1
Summer Average	Al	44	0.6522	0	0.1405	0.0676	± 0.008	0.20	3
	Cu	44	2.2162	0	0.1392	0.1634	± 0.140	1.00	1

Table 2: Descriptive statistical results for Al and Cu concentrations in swimming pools in the summer period in province and districts of Canakkale, Turkey (*n*: Number of observation; *l*: number of samples exceeding the limit concentration values specified by Turkish Health Ministry).

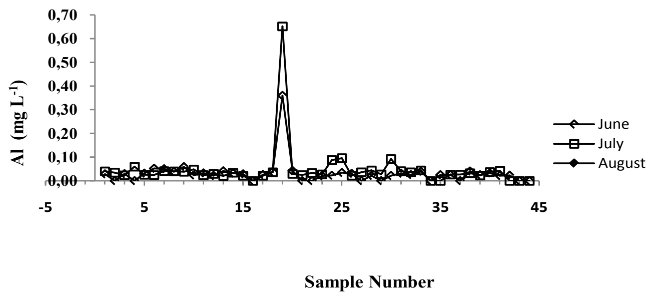

Figure 5: Variations of Al in swimming pool waters in the summer period in the province and districts of Canakkale, Turkey (Sample numbers 1-13: Ayvacik-Kucukkuyu, 14-16: Bozcaada, 17: Ezine, 18: Eceabat, 19: Gelibolu, 20-21: Çan, 22-23: Yenice, 24-25: Biga, 26-32: Çanakkale, 33-36: Ayvacik-Assos, 37-44: Ayvacık-Kadırga).

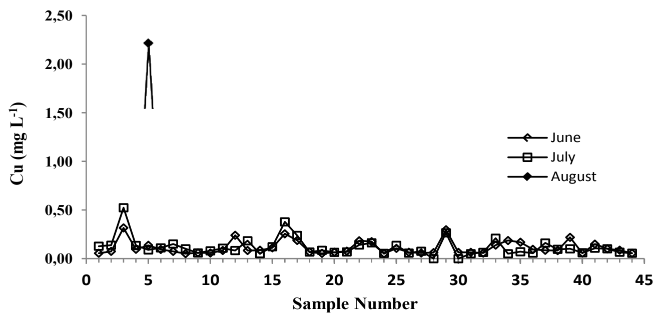

Figure 6: Variations of Cu in swimming pool waters in the summer period in the province and districts of Canakkale, Turkey (Sample numbers 1-13: Ayvacik-Kucukkuyu, 14-16: Bozcaada, 17: Ezine, 18: Eceabat, 19: Gelibolu, 20-21: Çan, 22-23: Yenice, 24-25: Biga, 26-32: Çanakkale, 33-36: Ayvacik-Assos, 37-44: Ayvacık-Kadırga).

2.216 mg L⁻¹ (0.139 ± 0.163 mg L⁻¹), respectively. Although average Al and Cu concentrations increased from June to August probably due to the increasing swimming population based on both average values, maximum values of Al (0.6522 mg L⁻¹) and Cu (2.2162 mg L⁻¹) were found in July and August, respectively (Table 2). However, although all the maximum Al and Cu values were over the limit values for pool water standards of the Turkish Health Ministry and the standards of some European countries, all the average values were considerably under these limit values (Table 2).

Results revealed that Al concentrations varied between 0 and 0.6522 mg L⁻¹ during the sampling period (Figure 5). There were 3 Al measurements exceeding limit standard values of Turkish Health Ministry (0.20 mgL⁻¹) in the same pool (Sample number: 19) in Gelibolu district during the sampling period. These values over the limit values in

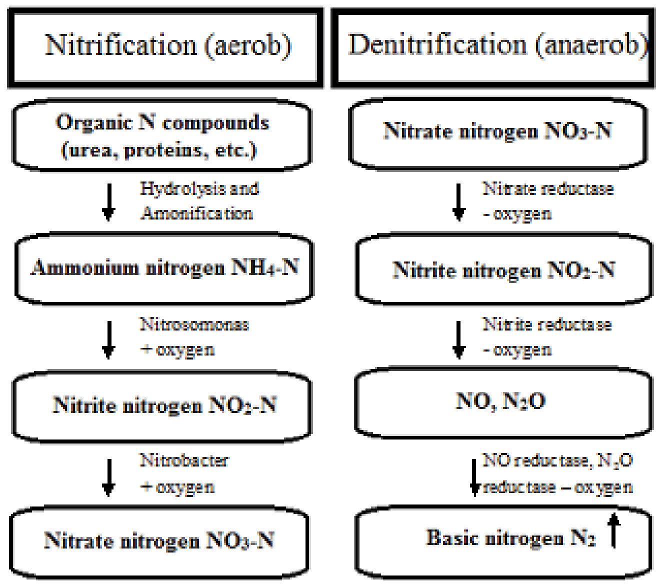

Figure 7: Nitrification and denitrification.

June, July and August were 0.3596, 0.6522 and 0.3736 mg L⁻¹, respectively (Figure 5). Unhealthy pool water ratio for Al was calculated as 2.27%.

Findings showed that Cu concentrations varied between 0.0517 and 2.2162 mg L⁻¹ during the sampling period (Figure 6). There was only one Cu concentration value in Assos Kadırga area in Ayvacik-Kucukkuyu (2.2162 mg L⁻¹) exceeding the standard limit value for pool waters of Turkish Health Ministry (1.00 mg L⁻¹) (Figure 6). Unhealthy pool water ratio for Cu was calculated as 0.76%.

Conclusion

At the end of the study, NO_2^-, NO_3^- and NH_4^+ findings showed that there were suitable aerobic nitrification conditions due to the high dissolved oxygen concentrations in the pool waters. Therefore, NO_3^- contents in the pool waters, especially in the Ayvacik district, were higher than any other nutrient parameter. It is known that NH_4^+ and NO_2^- very quickly convert to NO_3^- due to nitrification in aerobic conditions (Figure 7) and so NO_2^- completely and NH_4^+ partly were under standard limit values (Table 1). In a study on microbiological and chemical qualities of 144 swimming pool waters (69 indoor, 75 outdoor) over a three-year period (2010-2012) in Bologna (Italy), Dallolio et al. using the same methodology (spectrometric method) as this study, it was revealed that noncompliance rates to standard limit values (unhealthy pool water ratio) were 1.20 and 0.00% in the indoor swimming pool waters and outdoor pool waters, respectively. Unhealthy pool water ratio for NO_3^-

in this study was higher (10.61%) than the levels (1.20%) in the study by Dallolio et al. This shows that in Turkey we have more serious problems with pool water pollution than Italy in terms of NO_3^-. In another study on nitrate, chlorate and THM in outdoor swimming pools located in 4 different groups (1: hotel-motel pools; 2: site pools; 3: building pools; 4: municipal swimming pools) in Miami (USA), the average NO_3^- concentrations in the 4 different groups were between 2.70 (in hotel-motel pools) and 16.5 mg L^{-1} (in building pools).

In general, high NO_3^- concentrations in pool waters may have human origin (urine, faces and sweat). However, non-human factors such as birds and plants may also contribute. It is known that feces of birds contain higher nutrient values than feces of humans. To solve such problems, it is important to ensure sanctions and obligations for hygiene checks and periodic changes of pool water in a legal manner. Although NO_3^- was maximum in July, maximum and average concentrations of nutrients and metals increased from June to August, probably due to the increasing tourist population. In Turkey, June and August periods have peak tourist population and hence pool use increases in these periods.

According to our findings, there were 3 Al measurements (0.3596 mg L^{-1} in June 0.6522 in July and 0.3736 in August) in the same pool (sample number: 19) in Gelibolu district exceeding the limit standard values of Turkish Health Ministry (0.20 mg L^{-1}) (Figure 5). However, noncompliance rate (unhealthy pool water ratio) for Al (2.27%) was in normal range according to the range values of other European countries. Al showed high concentration values in some pool waters and periods due to the high use of flock agents which contain Al and insufficient pool water changes.

Although Cu concentrations varied between 0.0517 and 2.2162 mg L^{-1}, there was only one peak value during the study. The peak value exceeding the standard limit value for pool waters of Turkish Health Ministry (1.00 mg L^{-1}) was in Assos Kadırga area of Ayvacik-Kucukkuyu (2.2162 mg L^{-1}) (Figure 6). The lowest noncompliance rate to standard limit value (unhealthy pool water ratio) between pool water quality parameters (NH_4^+, NO_2^-, NO_3^-, Al and Cu) was for Cu (0.76%). In other words, the smallest problem was with Cu values during the study except for sample number 5 in Assos-Kadırga (2.2162 Cu mg L^{-1}). The peak value of Cu is estimated to be caused by overuse of algaecide chemicals.

As a result, due to the fact that Canakkale province and some districts such as Ayvacık, Kucukkuyu, Assos and Bozcaada host important tourism activity in Turkey, pool waters in such tourism regions should be monitored in view of not only chemical (Cu, Al, NO_3 and NH_4) and physical quality parameters (temperature, pH and DO), but also microbiological (*Salmonella* and *Vibrio,* etc.) quality parameters.

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