Article

Concentration of Selected Anions in Bottled Water in the United Arab Emirates

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Abstract: Several studies have shown concern over nitrate and nitrite contamination of prepared infant formula used by infants less than six months old, as it may lead to methemoglobinemia and death. One possible source of contamination is through the use of improperly treated drinking water. Contamination of water could result from fertilizers and manure runoff, not fully treated and released human and industrial waste, or from disinfection processes. In the United Arab Emirates (UAE), bottled water is the major source of drinking water and may be used for the preparation of infant formula. Therefore, in this study, several bottled water brands that are sold on the UAE market, and could be used for preparation of infant formula, were tested for nitrate and nitrite and other anions to show their compatibility with the permissible levels of the United States Environmental Protection Agency (U.S. EPA), United States Food and Drug Administration/Code of Federal Regulations (U.S. FDA/CFR), and other international organizations. All the bottled water samples demonstrated nitrate, nitrite, and other anions levels below the permissible levels accepted by U.S. EPA, U.S. FDA/CFR, and other international organizations, except for one sample that showed nitrite levels exceeding the European Commission and Drinking Water Directive (EC/DWD) permissible levels. Such study sheds light on the quality of bottled water sold not only in the UAE and the region, but also in other countries, such as France, since some of them are imported. In addition, the results shed light on the effectiveness of the treatment processes and possible sources of infant formula contamination that can affect the health of infants.

Keywords: nitrate; nitrite; bottled water; infant formula; contamination

1. Introduction

Drinking water is important for survival, and biological and chemical contamination is a serious matter that may have serious health effects. There is a worldwide concern over the quality of tap water, due to pollution, bacterial contamination, and the associated taste and odor [1,2]; therefore, people are turning to bottled water for safety and quality characteristics [3]. As a result, the global market for bottled water is huge and is growing continuously to meet the increase in demand and the search for good quality drinking water [3,4].

Such increase in demand for bottled water is especially noticed in arid areas, where fresh water resources are limited and weather conditions require the consumption of more water. For example, in the United Arab Emirates (UAE), the human population is growing rapidly because of an increase in the Emirati population as well as an increase in migration rate. In the UAE, there are limited fresh water resources, and to meet the increase in demand for water due to increases in both the population and the number of tourists, the country resorts to desalination and the import of bottled water from other countries. Therefore, maintaining good quality water, through regular testing and use of advanced techniques, is important to meeting this increasing demand for water.

Bottled water is used mainly for drinking, but is also being used in the preparation of infant formula and other food [5]. Therefore, contamination of bottled water is a concern for all countries and needs to be continuously monitored to meet the required quality of bottled water [2,3,5–14]. There are different biological (e.g., bacteria and viruses) and chemical (e.g., organic and inorganic, including, nitrate, nitrite, chloride, sulfate, and others) contaminants that are regulated by U.S. and other international organizations to maintain the required quality of bottled water. The presence of nitrate and nitrite in bottled water, for example, is of great concern, as it may affect human health. Bottled water is not only used for drinking, but also used for preparation of food and infant formula [5]; as a result, any contamination of bottled water would eventually result in the presence of nitrate and nitrite in the prepared food and formula. Contamination of bottled water with nitrate and nitrite affects the health of infants (up to 3–6 months) more than adults, as they are more susceptible to serious health conditions such as methemoglobinemia, due to the difference in their body size and chemical conditions of the body [15–17]. Other less serious conditions for children involve diarrhea, which can occur when high nitrate water is ingested [18]. Accordingly, the United States Environmental Protection Agency (U.S. EPA), United States Food and Drug Administration/Code of Federal Regulations (U.S. FDA/CFR), and other regulatory organizations have set a maximum contaminant level for nitrate and nitrite in our drinking water in response to the possible health effects [16,19–22].

In the UAE, bottled water is consumed by 90% of the population [23], which includes infants and children, and this rate of consumption of bottled water is increasing; therefore, it is necessary to continue to deliver bottled water to the consumer that meets the increase in demands and the required standards. Therefore, this research aims to measure the nitrate and nitrite levels in bottled water sold in the UAE that may be used for drinking and for preparing food and infant formula, and to compare the results with U.S. and other international permissible set standard limits. The results of testing the bottled water for nitrate and nitrite contamination will provide insight into the quality of the bottled water sold in the UAE and the region, and the effectiveness of the treatment processes used during production. In addition, the research will shed light on the nitrate and nitrite levels in bottled water and

whether there is a chance that infants who drink infant formula prepared using this bottled water may develop methemoglobinemia if levels exceeded the set standards.

2. Materials and Methods

2.1. Bottled Water Samples

Bottled water samples analyzed in this research were obtained from local markets in the UAE (Table 1). The bottled water samples represented the brands that were available on the shelves at the time of the research. Some of the bottled water samples were manufactured in the UAE while others were imported from other countries. All of the purchased bottled water samples were sold in plastic bottles. The bottled water samples were opened in the laboratory and analyzed after purchase for the following anions, nitrates (NO_3^-), nitrites (NO_2^-), fluorides (F^-), chlorides (Cl^-), bromides (Br^-), sulfates (SO_4^{2-}), and phosphates (PO_4^{3-}).

Table 1. Bottled water samples purchased from the UAE market.

Country of Origin	Water Samples	Water Type [a,b]
UAE	14	Pure Natural Water
		Pure Drinking Water
		Pure Natural Mineral Water
		Pure Distilled Water
		Natural Spring Water
Saudi Arabia	2	Drinking Water
France	2	Natural Mineral Water from Alps
Lebanon	2	Natural Mineral Water

[a] Terms used to describe bottled water or drinking water, *i.e.*, water in sealed bottles, which is safe to be consumed by humans [20]; [b] Terms used by manufactures to describe source of water (e.g., mineral refers to underground water, while spring refers to water flowing naturally to the surface from underground formation) [20] and production process used (e.g., distilled), or as a marketing tool.

2.2. Analysis of the Anions

Bottled water samples were measured for the concentration of anions, using a Metrohm 861 Advanced Compact Ion Chromatography (IC) system (Metrohm AG, Herisau, Switzerland). The Metrohm system includes a Metrosep A Supp 4/5 guard column for contaminant removal, a Metrosep A Supp 5-250/4.0 4 × 250 mm anion column for separation, and an IC732 IC detector. The analysis was performed at 20 °C and 13.2 MPa, with an injection volume of 1 μL at a 0.8 mL·min^{-1} flow rate. The eluent was prepared using 3.2 mmol·L^{-1} Na_2CO_3 (Fisher Scientific, PA, USA) and 1.0 mmol·L^{-1} $NaHCO_3$ (Fisher Scientific, PA, USA).

Different calibration curves (correlation coefficient ≥ 0.99) were developed, using a multi-ion standard (Seven anion standard II standard, Dionex, CA, USA) as the standard stock solution for all of the measured anions. The method applied to analyze the anions was tested for its recovery, by spiking the bottled water samples with aliquots of anion standard, and analyzed. The recovery of the analyzed spiked samples ranged from 92.6 to 98.0 %.

Analysis of the anions in the standards, the bottled water samples, and the spiked water samples was performed at the United Arab Emirates University (UAEU), Central Laboratories Unit (CLU). All analysis above, calibration analysis, system suitability, method validation, and other quality control checks were performed at CLU, in accordance with the International quality system, ISO/ICE 17025, in general requirements for the competence of testing and calibration laboratories.

3. Results and Discussion

Whether we are investigating tap or bottled drinking water, several things need to be examined and maintained to ensure that the drinking water is of good quality and lacks the presence of bacteria, chemicals, and other contaminants [20]. Tap drinking water may be obtained from different sources, such as surface or groundwater, or from seawater through desalination. Similarly, bottled water may be obtained from those resources, and as a result may contain different types of water, as demonstrated from the bottled water samples that were tested in this research (Table 1). For example, mineral water or spring water are terms that reflect different information, such as the source of water, chemical parameters, and production processes [20,21].

Bottled water is produced in large quantities to meet the increase in demand for fresh and clean drinking water. The bottled water may be sold in the manufacturing country or imported to other parts of the world. Accordingly, it might sometimes have to meet not only the local set standards, but also other U.S. and international standards, such as those set by the U.S. EPA, the World Health Organization (WHO), the European Commission/Drinking Water Directive (EC/DWD), and the U.S. FDA/CFR. The U.S. EPA sets regulations for tap water and the U.S. FDA/CFR sets regulations for bottled water in the U.S. [20], yet in some countries, the U.S. EPA and U.S. FDA/CFR regulations are referred to interchangeably without any consideration whether it is tap or bottled water. In this research, some of the bottled water samples that were purchased from the UAE market were manufactured in countries other than the UAE, such as France, Saudi Arabia, and Lebanon (Table 1). Therefore, the results obtained in this research are compared to the set standard limits of the different U.S. and international organizations (Tables 2 and 3).

Table 2. Nitrite and nitrate (mg·L^{-1}) levels measured in the bottled water samples, compared to the U.S. EPA [19], WHO [16], EC/DWD [22], and U.S. FDA/CFR [20,21] standard limits.

U.S. EPA	-	-	1.00	10.00
WHO	3.00	50.00	0.90	11.00
EC/DWD	0.50 (0.10)	50.00	-	-
U.S. FDA/CFR	-	-	1.00	10.00
Water Sample	**Nitrite-Nitrite**	**Nitrate-Nitrate**	**Nitrite-Nitrogen**	**Nitrate-Nitrogen**
1	ND	ND	ND	ND
2	ND	2.51	ND	0.57
3	ND	2.81	ND	0.64
4	ND	ND	ND	ND
5	ND	2.77	ND	0.63

Table 2. *Cont.*

Water Sample	Nitrite-Nitrite	Nitrate-Nitrate	Nitrite-Nitrogen	Nitrate-Nitrogen
6	ND	ND	ND	ND
7	ND	ND	ND	ND
8	ND	2.78	ND	0.63
9	ND	5.67	ND	1.29
10	ND	2.94	ND	0.67
11	ND	2.50	ND	0.57
12	ND	ND	ND	ND
13	ND	2.80	ND	0.64
14	ND	ND	ND	ND
15	ND	2.78	ND	0.63
16	ND	ND	ND	ND
17	ND	2.50	ND	0.57
18	ND	2.90	ND	0.66
19	ND	2.99	ND	0.68
20	1.29	2.92	0.39	0.66

ND = Not detected.

Table 3. Anion contaminant level $(mg \cdot L^{-1})$ measured in the bottled water samples, compared to the U.S. EPA [19], WHO [16], EC/DWD [22], and U.S. FDA/CFR [20,21] standard limits.

	Fluoride	Chloride	Bromide	Phosphate	Sulfate
U.S. EPA (MCL)[a]	**4.00**	-	-	-	-
U.S. EPA (SMCL)[a]	2.00	250.00	-	-	250.00
WHO	1.50	-	-	-	-
EC/DWD	1.50	250.00	-	-	250.00
U.S. FDA/CFR	1.70	250.00	-	-	250.00
Water Sample	**Fluoride**	**Chloride**	**Bromide**	**Phosphate**	**Sulfate**
1	ND	44.90	1.15	ND	4.18
2	0.12	39.00	1.21	ND	5.68
3	ND	19.04	ND	ND	50.20
4	0.20	5.56	1.31	ND	31.40
5	ND	56.30	1.16	ND	16.70
6	0.61	147.90	1.37	ND	4.38
7	0.20	5.53	1.31	ND	31.10
8	ND	55.50	1.15	ND	16.50
9	ND	2.47	1.36	ND	1.75
10	1.05	67.18	1.21	4.56	16.23
11	0.12	45.66	1.16	ND	3.65
12	ND	39.00	1.19	ND	5.27
13	ND	19.13	ND	ND	50.40
14	0.20	5.53	1.31	ND	31.30
15	ND	55.50	1.15	ND	16.30
16	0.64	145.70	1.45	ND	3.85

Table 3. *Cont.*

Water Sample	Fluoride	Chloride	Bromide	Phosphate	Sulfate
17	0.20	5.50	1.31	ND	31.00
18	ND	47.72	ND	4.58	ND
19	ND	46.50	1.05	4.55	ND
20	1.07	69.60	1.20	4.55	15.00

[a] MCL (Primary standard, maximum contaminant level) and SMCL (Secondary standard, maximum contaminant level). ND = Not detected.

The U.S. EPA and other organizations, such as the WHO, EC/DWD, and U.S. FDA/CFR, have set standard limits (Table 2 and 3) for several drinking water (tap or bottled) contaminants, beyond which health problems may be experienced. Some of those contaminants, which are the main focus of this research, are nitrate and nitrite, in addition, other anions were measured for chromatographic purposes to identify the nitrate and nitrite anions, and for reflecting the quality of the bottled water. Contamination of the bottled water is not only a reflection on the quality of the source water that was used for the production, but could also reflect the effectiveness of the water treatment processes as well as the storing conditions [24].

3.1. Bottled Water Container

Bottled water is sometimes sold in glass or plastic bottles. All of the bottled water that was sampled in this research was marketed in plastic bottles constructed from polyethylene terephthalate (PET or PETE; both are abbreviations for the same type of plastic). Earlier research showed no effect of such a container (plastic) on the anions analyzed in this research [9]. Therefore, leaching from the plastic bottles did not contribute to the level of anions reported in this research.

3.2. Nitrate and Nitrite Anions

Nitrate and nitrite are natural forms of nitrogen that are present and needed in the environment. Human activities increase their levels through different point and non-point sources, which eventually cause them to be present in our drinking water. Such activities include the excessive use of fertilizers that leach into ground and surface waters, and the release of untreated industrial and sewage waste [15,16,25–32]. Drinking water treatment plants use different technologies—for example, ion exchange, reverse osmosis, biological processes and others—to reduce or remove the nitrate present in water used to prepare drinking water, yet due to human error and lack of frequency of monitoring (testing), sometimes high nitrate levels in drinking water may be experienced. Therefore, in addition to improving the methods used for the preparation of the drinking water, it is important to prevent the original water used for the production of drinking water from being contaminated with nitrate and nitrite, through better management of the wastewater before release into the environment, and also by reducing input from other sources [16].

As a result, the U.S. EPA considers the nitrate and nitrite permissible levels as primary standards under the National Primary Drinking Water Regulations (NPDWRs) that are required to be met by drinking water suppliers [19].

The nitrite and nitrate levels in the tested bottle water will be expressed in terms of nitrite-nitrogen and nitrate-nitrogen (mg·L^{-1}) levels (U.S. EPA and U.S. FDA/CFR parameters) to meet the uniformity in reporting parameters used for such contaminants in the literature. Other parameters, such as nitrate-nitrate and nitrite-nitrite (mg·L^{-1}), are also used in the literature and will be referred to when needed. Both parameters are expressed in Table 2. In comparison, to the U.S. EPA and U.S. FDA/CFR permissible levels for nitrite-nitrogen (1 mg·L^{-1}), and the WHO-set level of 0.9 mg·L^{-1} nitrite-nitrogen, all of the samples exhibited levels below the set standard limits, as no nitrite contamination was detected in any of the bottled water samples, except for sample 20, at 0.39 mg·L^{-1} nitrite-nitrogen (1.29 mg·L^{-1} nitrite-nitrite) (Table 2, Figure 1) [16,19–21]. Although this contamination level is below the U.S. EPA, U.S. FDA/CFR, and WHO set standard limits, it should be given careful attention. In addition, this level of nitrite contamination (the equivalent mg·L^{-1} nitrite-nitrite measured parameter) is above the EC/DWD permissible levels, and as a result may be a source of danger and may cause health problems for infants, considering this set standard limit (Table 2).

Figure 1. Nitrate-nitrogen and nitrite-nitrogen levels measured in the bottled water samples.
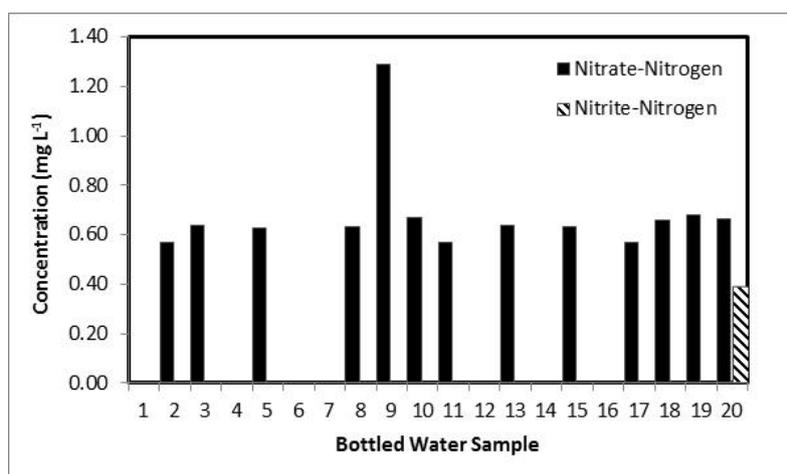
Thirteen of the 20 bottled water samples contained nitrate-nitrogen (Table 2, Figure 1). The nitrate-nitrogen levels in the bottled water samples are below the U.S. EPA set standard limit of 10 mg L^{-1} and the set standard limits of all the other organizations (Table 2, Figure 1) [19].

The WHO, EC/DWD, and U.S. FDA/CFR, in addition to having individual set limits for the nitrate and nitrite levels, also have other set conditions for contamination levels, in terms of considering the total concentration of both nitrate and nitrite anions and other criteria [16,20–22].

3.3. Other Anions

Other anions such as fluoride, chloride, bromide, phosphate, and sulfate were measured in this research. Set standard limits are reported for those anions, based on the probable danger and effect that they may pose to health (Table 3). Under the U.S. EPA regulations, some of those anions are considered as primary standards (Table 3), while others are considered as secondary standards under the National Secondary Drinking Water Regulations (NSDWRs) (Table 3) [19]. Primary standards (e.g., fluoride only) are required to be followed and met by the water manufacturer/system, while secondary standards (e.g., fluoride, chloride, and sulfate only) are recommended, but not required, to

be met by the water manufacturer/system, which presents an idea about their effect on human health, as one causes health problems, while the effects of the other are more cosmetic and esthetic, respectively (Table 3) [19]. Fluoride is considered under both the primary and secondary standards, depending on the level of fluoride present in the water, as different effects on the body may be experienced at different levels [19].

Fluoride is added to drinking water for dental care, although when the level exceeds the allowed limit, it may lead to bone disease and teeth problems [19]. Bottled water that contains fluoride may contribute either positively or negatively to dental care, depending on the fluoride level and whether it is within the permissible levels or not. Fluoride levels measured in all the bottled water samples were below the permissible levels (Table 3, Figure 2), yet samples 10 and 20 were close to the WHO (1.5 mg·L^{-1}), EC/DWD (1.5 mg·L^{-1}), and U.S. FDA/CFR (1.7 mg L^{-1}, as maximum level in bottled water to which fluoride is added) required levels [16,20–22]. The U.S. FDA/CFR, under the 21CFR 165.110, sets the permissible fluoride levels based on the annual average maximum daily air temperatures at the location where the bottled water is to be sold, and also reports other fluoride permissible levels that are based on whether it is imported or not, and whether the fluoride is added (0.8 mg·L^{-1}, maximum level for imported bottled water) or not added (1.4 mg·L^{-1}, maximum level for imported bottled water) [20,21].

Figure 2. Anion concentrations (**a**–**e**) measured in the bottled water samples.
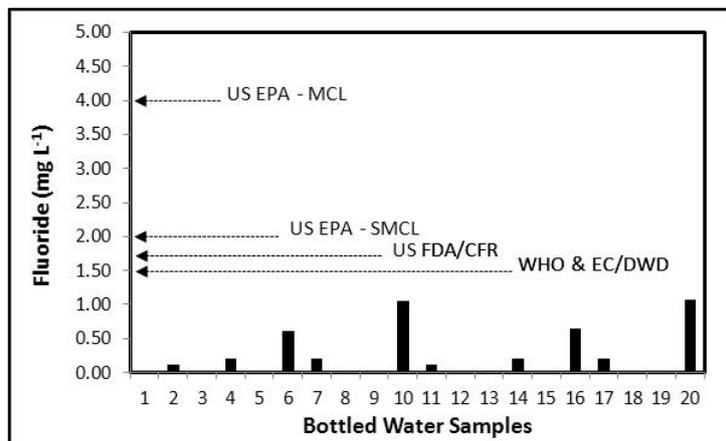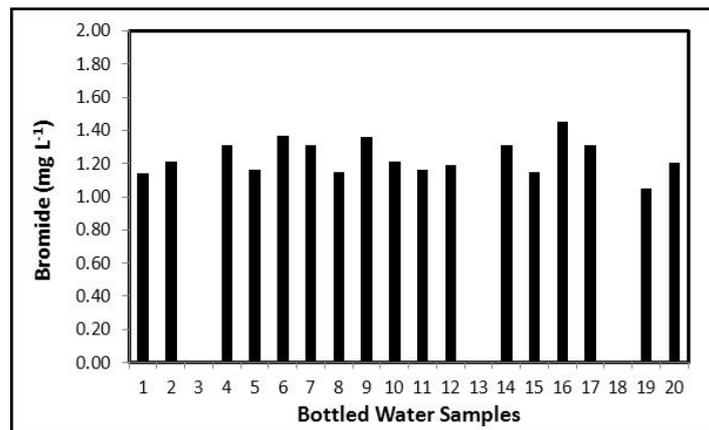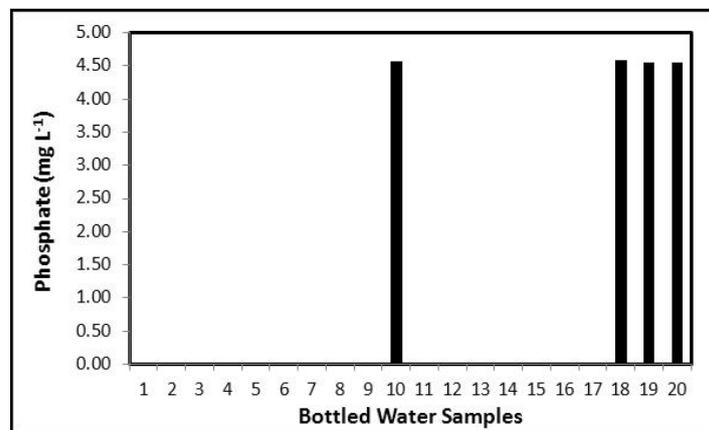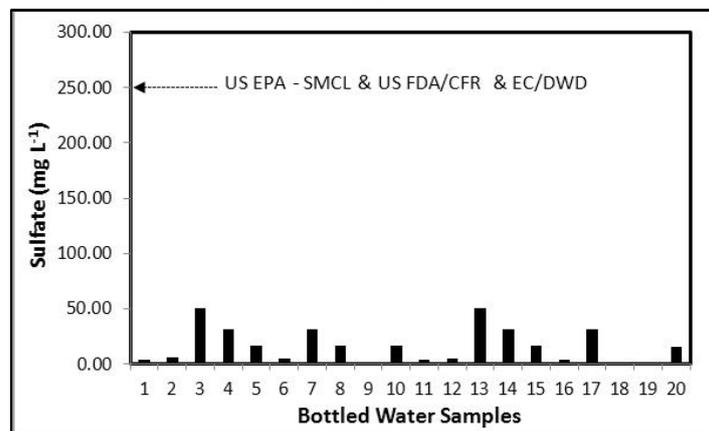

(**a**)
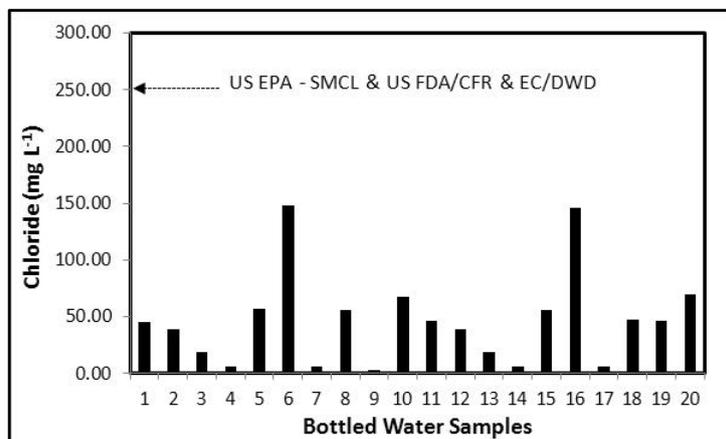
(**b**)

Figure 2. *Cont.*

(c)

(d)

(e)

Permissible levels for chlorides and sulfates are provided by the U.S. EPA, EC/DWD, and the U.S. FDA/CFR (Table 3). The U.S. EPA considers chlorides and sulfates as secondary standards, *i.e.*, they may cause cosmetic or esthetic effects [19]. The WHO does not provide a guideline value for either chloride or sulfate, since the levels existing in the drinking water industry pose no health concerns [16]. The WHO also states that chloride concentration exceeding 250 mg·L^{-1} can change the taste of water, and that sulfate levels exceeding 500 mg·L^{-1} may cause gastrointestinal problems (laxative effect) in sensitive people [16], and therefore may have an effect on infants and children.

Some of the samples, 6 and 16, experienced high chloride levels, yet all the chloride levels measured in all the bottled water samples were below the permissible levels for all the organizations that report set standard limits for chloride (Table 3, Figure 2). All the bottled water samples demonstrated sulfate levels below all the organizational set standard limits discussed in this research (Table 3, Figure 2).

No set limits are provided for bromide and phosphate from any of the organizations (Table 3). According to the WHO, no guideline values have been established for bromides and phosphates since they exist in drinking water at levels below those of health concern [16]. Most of the bottled water samples demonstrated bromide levels, with a maximum of 1.45 mg·L^{-1} (Table 3). Samples 10, 18, 19, and 20 are the only bottled water samples that contained phosphate (Table 3).

3.4. Bottled Water Labels

Results obtained in this research for nitrate, nitrite, and the other anions cannot be compared to the information of the anions reported on the labels of bottled water samples, as not all the bottled water samples reported levels for those anions on their labels. Studying and comparing together the information provided on the labels (only) of the different bottled water samples that were tested in this research shows no consistency for most of the samples, in terms of the type of anions reported and the levels for some of the reported anions (Figure 3). For example, some of the bottled water samples did not report the nitrate levels on their labels (similarly for fluoride, chloride, and sulfate), and only two samples of bottled water reported the nitrite levels. None of the bottled water samples reported any values for bromide and phosphate, probably because there are no set standard limits that are provided from any of the organizations, as they usually exist at levels below that of health concern in the drinking water [16]. Consistency of information on the labels is important for allowing the consumer to compare the products, as well as for providing a full picture about the quality of the bottled water. Reporting of nitrates and nitrites, for example, is of equal importance, as this reflects the health effects that infants may endure if the permissible levels are exceeded.

Figure 3. Anion concentrations (**a-d**) reported on the labels of bottled water samples. Only bottled water samples with bars and ⬇ (representing zero value) reported values for the anions.
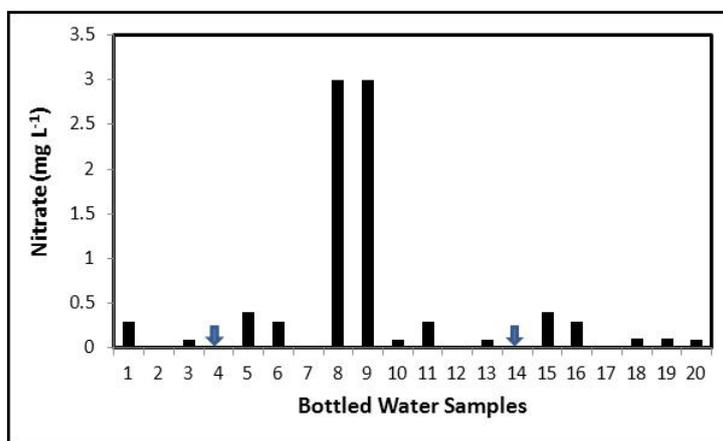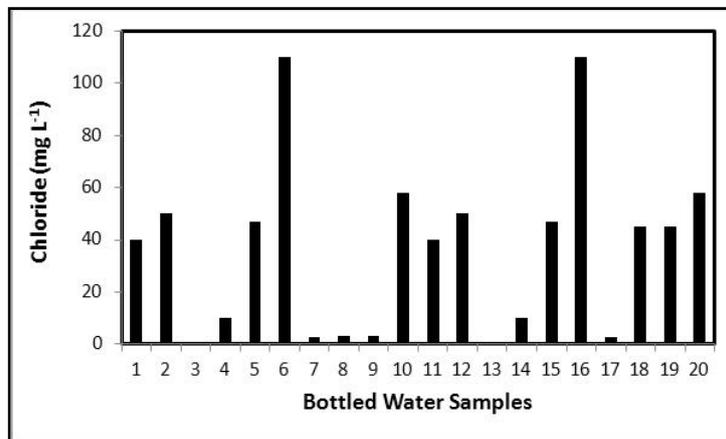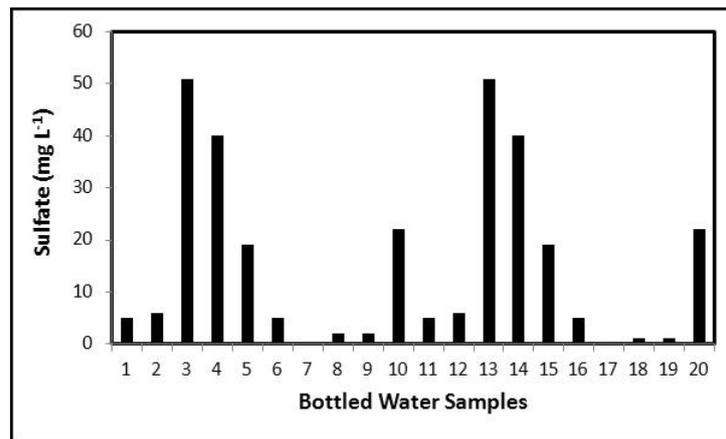

(**a**)

Figure 3. *Cont.*
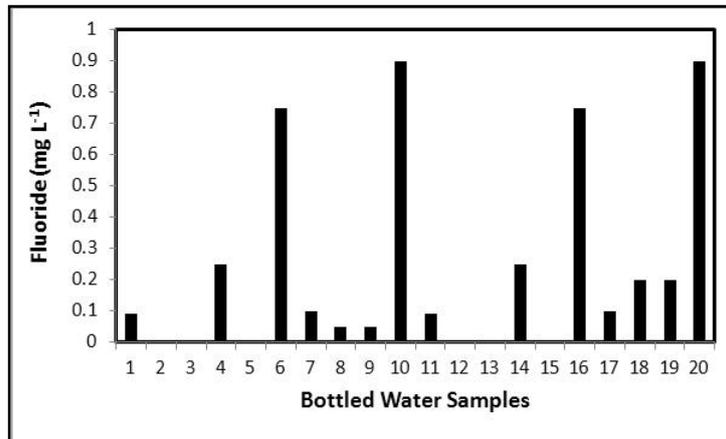
(**b**)

(**c**)

(**d**)

4. Conclusions

Bottled water is an important source of drinking water in the UAE, which has limited resources of fresh water. Bottled water may be used for infant formula and food preparation for infants. Contamination of bottled water would mean contamination of the infant formula, for example, and a

higher contamination level can exist if the infant formula is already contaminated. As a result, this may act as a source for serious diseases, such as methemoglobinemia.

In this research, the nitrate and nitrite and other anion levels (fluoride, chloride, and sulfate) in the tested bottled water were, in general, below the permissible standards set by U.S. and international organizations. This reflects the quality of the bottled water, yet several things should be addressed:

- In one sample, the nitrite level exceeded the permissible level set by the EC/DWD organization. Nitrite as well as nitrate may cause serious problems, and therefore, the nitrite level should be reduced so it does not pose any health risk, such as methemoglobinemia in infants.
- Some samples experienced high anion concentration levels, yet they did not exceed the set standard limits. Such results suggest that more attention should be given to the monitoring and treatment processes of the drinking water, so exceeding the permissible levels would not be experienced in the future, leading even to increased contamination if the infant formula or food used is also contaminated.
- Labeling of bottled water is an important tool that consumers may use to find information about the quality of the drinking water. Therefore, labeling information, in regard to which type of anions are reported, should be consistent from one brand to another. In addition, nitrite levels should be reported on the labels of the bottled water, since elevated levels of this anion could lead to serious health conditions, as is the case with nitrate and other anions.

In conclusion, continuous monitoring of water quality and effectiveness of the treatment processes, and obeying regulations, are required to ensure that the water quality meets the set standards and to meet the increasing demand for good quality bottled water.

Acknowledgements

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