Evaluation of Changes in Some Physico-Chemical Properties of Bottled Water Exposed to Sunlight in Bauchi, Nigeria

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Abstract. It is common for bottled water and other assorted drinks to be seen displayed outside stores and in the sun in most parts of Nigeria. The country is mostly hot year-round and over the course of the year temperatures can rise to as high as 40°C around March-April in the study area. The leaching effect of chemicals from polyethylene terephthalate (PET) bottled water

10   was investigated for five (5) commercially available bottled water brands. Temperature, pH, Antimony, Bisphenol A and Nitrate levels were measured on day zero, 14 and 28 for control samples and samples exposed to direct sunlight, using destructive sampling technique. Antimony was not detected in brands A, B and E in the baseline measurement at day zero while brands C and D had low values; after 28days all the control samples still had Antimony levels within the United States Environmental Protection Agency (US EPA) standard. Meanwhile all the samples exposed to sunlight exceeded US EPA

15   standard levels at 14 and 28days except brand A which was within limit at 14days with value of 4.59µg/L. All control and exposed samples were below the European Union Drinking Water Directive (EU DWD) total daily intake (TDI) of Bisphenol A (0.05mg/kg body weight/day). Values obtained for Nitrate showed that all control samples did not exceed the US EPA guideline level for Nitrates in drinking water for 0, 14 ad 28days while three (3) of the samples, Brands C, D and E, exceeded the guideline level at 28days. Exposure of bottled water to sunlight was seen to impair the quality of the water for consumption.

20   **Keywords:** Antimony (Sb), Bisphenol A (BPA), Nitrates (NO_3), Polyethylene terephthalate (PET), pH, temperature, sunlight exposure

1 Introduction

Bottled water is classified as a consumer food product and consist of spring, purified, mineral, sparkling, artesian and well water which are to be carefully processed to meet regulatory standards (IBWA, 2020).

25   Historical records show the introduction of for sale bottled water to the 17th century in the United Kingdom (Holy Well Bottling Plant) when water from mineral Springs were thought to possess therapeutic and healing properties and sold as remedies for ailments (Malvern Beacon, n.d.; Mitte Team, 2019). When Johann Jacob Schweppe (1783) discovered how to carbonate water, the 'fizzy' quality of plain water gave a considerable competition to mineral water (Mitte Team, 2019;

Schweppes-Heritage, n.d.). Lowering cost of glass and techniques of bottling increased the popularity of bottled water and

30   carbonated drinks in the United States in the 19th century. Sales were common up till the early part of the 20th century when Chlorination was discovered to make tap water safer, causing a decline in the choice of bottled water (Hurly, 2016; National Center for Environmental Health et al., 1999).

The introduction of polyethylene terephthalate (PET) as a replacement for glass in the 1970-80s caused a reduction in cost of manufacturing bottled drinks (including water) (Leigh, 2011; Mitte Team, 2019; Parker, 2019). PET is a clear, strong and light

35   weight plastic (thermoplastic polyester resin) used in producing packaging materials for food, beverages, cosmetics, water, photographic films; and in making apparels (International Life Sciences Institute, 2000; PETRA, 2015; US EPA, 1995). PET was first synthesized in the 1940s by DuPont chemists (PETRA, 2015). The processing of PET involves mixtures of ethylene glycol (EG) and terephthalic acid (TPA) or dimethyl terephthalate (DMT) to form chain of polymers. The output is extruded, cooled and cut into small pellets and the pellets are moulded into any shape by heating them into a molten liquid (PETRA,

40   2015; US EPA, 1995). PET bottles can be recycled by breaking it down into its constituents and using same to make new PET materials, otherwise they become a nuisance and an environmental hazard when improperly disposed (PETRA, 2015; US EPA, 1995).

Drinking water quality is a major risk factor for diseases and established standards ascertain the suitability of water for intended use. These standards can be broadly categorized as physical, chemical, biological and radiological (Davis, 2010; Gaur, 2008;

45   Spellman, 2003).

The physical properties of water are colour, turbidity, temperature, taste and odour. The chemical properties of water are pH or the presence of chemicals like arsenic, Iron, Lead, Sodium, Zinc or other toxic organic or inorganic substances. Some chemicals are essential to human and animals in trace amounts, but prolonged exposure in higher amounts can be dangerous to human health. Chemicals can occur naturally from water source; whereas others are as a result of human activities

50   (industrial-mining and human dwellings); agricultural activities (fertilizer and pesticide application); water treatment (supply lines, coagulants); pesticides (public health use); or containing vessels where water is stored (plastic bottles) (WHO, 2017).Regulations specifically aim at ensuring the deleterious effects of the chemicals are avoided. Other chemicals like Antimony, Bisphenol A and Nitrates can be present in drinking water as a result of plastic bottle feeding of infants and other plastics including bottled drinking water (US EPA, 2010; WHO, 2017). For example, Antimony found in Antimony trioxide

55   is used as a flame retardant or in polycondensation when PET is produced (Bach et al., 2014; WHO, 2003a).

Radiological standards deal with water in contact with radioactivity. Parameters like alpha, beta particles, Radium-226, 228 and Uranium have standards set for drinking water to ensure no untoward health effects to human wellbeing when consumed (Davis, 2010). Radiation occurs naturally (accounting for almost 80% of dose from all radiation sources), from medical diagnosis (accounts for 19.6%) and from man-made sources (accounts for 0.4%) (WHO, 2017).

60 A summary of the drinking water and bottled water (shown in parenthesis) quality standards available as issued by major institutions for physical, chemical, biological and radiological characteristics; and suspected health effects are presented in Table 1. As seen in Table 1, in US and Canada as with many other places a number of the regulations cover for both drinking water and bottled water quality while for regions like Nigeria no bottled water regulation is in place and sometimes drinking water quality standards are adopted.

65 There is increasing concern for the leaching of chemicals from containers (including PET) as a result of increasing ambient temperatures (University of Florida Institute of Food and Agricultural Sciences, 2014; Zanolli, 2019). In most parts of Nigeria, including the study area, bottled water and other assorted drinks for sale are displayed outside stores and in the sun for days and weeks or hawked in traffic by the roadside until they are sold. The country is mostly hot year-round. This study seeks to investigate the changes in levels of specific parameters (Antimony, Bisphenol A, Nitrates, pH) in five (5) commercially

70 available market brands of bottled water continually exposed to sunlight for 0 – 28days in Bauchi, Bauchi State and the effects on the quality of the water. The study area is a metropolitan city and capital of Bauchi State in Nigeria. Over the course of the year temperatures can rise to as high as 40ºC around March-April in the study area. The study was carried out between December, 2019 to January, 2020 when maximum temperature was around 31ºC (World Weather Online, 2020).

2 Materials and Methods

75 **2.1 Reagents**

Reagent grade analytical chemicals and deionized water were used for preparing all solutions.

2.2 Sampling of the Bottled Water

Chemical analysis of the water contained in the plastic bottles were carried out in order to investigate whether the bottles leach chemical(s) to the bottled water under heat (source: sun) and the level of the chemical concentration over time (28days) in the

80 bottled water. Five (5) commercially available brands of bottled water (obtained directly from the suppliers) were taken as samples and tested (for pH, Antimony, Bisphenol A, Nitrates and Temperature) immediately afterwards to establish the baseline parameters. Samples (labelled A to E) were exposed under direct sunlight and control samples were kept in storage to replicate non-exposure to direct sunlight. Sample testing was carried out at 14 and 28 days between December 2019 (Ambient: Max temp = 31ºC, Avg. temp = 26ºC and Min Temp = 16ºC) and January 2020 (Ambient: Max temp = 31ºC, Avg.

85 temp = 24ºC and Min temp = 14ºC) (World Weather Online, 2020) using a destructive sampling technique.

2.3 Procedures Used in Measuring Parameters

pH and Temperature were evaluated using standard methods according to (APHA, 2018c) and (APHA, 2018a) respectively. Antimony was measured according to 3110 Metals by Atomic Absorption Spectrometry (APHA, 2018b). The presence of Nitrate was determined using the colorimeter (sulphanilamide method) according to 4500-NO3− Nitrogen (Nitrate) (APHA, 2018d). Bisphenol A was measured using High-performance liquid chromatography (HPLC) according to 6810 Pharmaceuticals and Personal Care Products (APHA, 2018e).

2.4 Method of Data Analysis

The data in this study was statistically analysed using R (3.6.3) (R Core Team, 2020) programming (in RStudio Version 1.2.5033). The method of data presentation of tables and charts were done in comparison to the water quality standards set out by WHO (2006) guidelines for drinking water quality, US EPA (2009) National Primary Drinking Water Regulations and the Nigerian Standard for Drinking Water Quality (NSDWQ).

3 Results and Discussion

3.1 pH

The pH values obtained from the tests carried out for the five brands of bottled water are as given in Fig 1. The baseline (day zero) pH values for all brands ranged between 5.25 and 7.4. For the control sample at 14 day and 28 day, there was a general decrease of pH implying increase in acidity in the 5 brands. Brand A showed approximately 3.8% and 5.8% reduction in pH in day 14 and 28; Brand B-3.6% and 7.3%; Brand C-5.4% and 9.2%; Brand D-13% and 20% (highest reduction); and Brand E- 2.8% and 7% respectively. The final pH values at the 28 day for Brands A, B, C, D and E were 6.97, 5.93, 6.45, 4.17 and 6.07 respectively (Fig. 1). Samples exposed to direct sunlight had temperatures of 47°C and 56°C on day 14 and day 28 respectively with Brand A showing 15% and 20% reduction in pH at day 14 and day 28; Brand B-23.4% and 46.7%; Brand C-16% and 39%; Brand D-19.8% and 49%; and Brand E- 27% and 54.8% respectively. The final pH values at the 28 day for Brands A, B, C, D and E were 5.92, 3.41, 4.32, 2.67 and 2.95 respectively.

The baseline pH values all fall within range of standard (NSDWQ and US-EPA: 6.5-8.5) except for Brands B and D. For the control samples at the 28 day only Brand A had pH at regulatory level (pH of 6.97). The sources of water may be one of the reasons for the pH values obtained for the control samples. Sample A is sourced directly from a spring aquifer which may explain why it has the lowest pH (Fisher et al., 2017), while samples B-E source water are from deep boreholes which might account for the changes in the values of pH noticed (Wright, 2015). The geology of the locations of water source may also have influence on the pH values.

The sample exposed to sunlight had a lower pH (higher acidity) for each Brand and also as the days of exposure to sunlight increased. This is similar to research by (Muhamad et al., 2011), where samples were exposed to sunlight for 5 days (there was a decrease in pH with mean max temperature between 41ºC to 47ºC). However, the result from this study varies from investigation by (Akharame et al., 2018), where the pH values after the 28 day were all within the WHO and NSDWQ regulation levels. WHO has highlighted health effects of pH<4 to include eye redness and irritation and for pH<2.5 damage to epithelium (WHO, 2003b). The result suggests that sunlight exposure (temperature) affects pH of bottled water.

3.2 Antimony

The values for antimony obtained from the tests carried out for the 5 Brands of bottled water are as given in Fig 2. In the baseline measurement (day zero), Antimony was not detected in Brands A, B and E while Brands C and D had values of 0.095 µg/L and 0.35 µg/L respectively. At day 14, the control samples had a range of Antimony values between 1.53µg/L and 4.35µg/L respectively (Fig. 2). On day 28, Antimony levels for the control samples had a range of 2.87µg/L and 5.41µg/L. This corresponded to an 88%, 45%, 21%, 24% and 37% increase from day 14 to day 28 for the control samples. The samples exposed to sunlight had a range of 4.59µg/L and 11.41 µg/L on day 14; and 8.17 µg/L and 14.49 µg/L on day 28. This corresponded to about 78%, 58%, 34%, 27% and 8% increase from day 14 to day 28 of being exposed to sunlight.

The increase in level of Antimony in samples exposed to sunlight after the 28 day was higher than in the experiments carried out by Bach et al. (2014). In that research, water samples had increase in Antimony levels from 0.7µg/L to 0.98µg/L (about 27% or 1.4times the initial value) and 0µg/L to 0.5µg/L after the 10th day of exposing mineral (non-carbonated) and ultrapure water samples to sunlight respectively. The increase in levels of Antimony could be attributed to effects of pH, temperature and UV radiation (Fig. 2) from the sun, supported by researchers which identified the significance of pH, temperature and days of exposure in the amount of Antimony leached into bottled water (Al-Khatim et al., 2019; Chapa-Martínez et al., 2016; Dogan and Cebi, 2019; Zmit and Belhaneche-Bensemra, 2019). The high level of the Antimony values can also be attributed to the type of plastics used by the different brands as identified by (Koyuncu and Alwazeer, 2019) who suspected reason for high Antimony levels in milk contained in PET bottles results from type and production processes employed in making containers. Qiao et al. (2018) also recorded higher values of Antimony leached in drinking water bottles exposed to high temperatures. Notably, all control samples did not exceed the US-EPA level of 6µg/L (Fig. 2), while in the samples exposed to sunlight all exceeded the US-EPA values after the 14th day though they did not exceed the WHO regulation of 20µg/L.

3.3 Bisphenol A (BPA)

The values of Bisphenol A obtained from the tests carried out for the 5 brands of bottled water are as given in Fig 3. The baseline results for both control and sunlight exposed samples were below limit of detection as seen in Fig. 3. For the control sample on day 14 there was increase in level of BPA to 3.61µg/L, 3.92 µg/L, 4.1 µg/L, 5.4 µg/L and 3.46 µg/L for Brands A,

B, C, D and E respectively. At the 28th day, there were 14.7%, 14.5%, 20.7%, 25.7% and 24.6% increase respectively from day 14. The samples exposed to sunlight showed BPA levels of 8.64 µg/L, 9.34 µg/L, 10.1 µg/L, 13.42 µg/L and 8.59 µg/L for Brands A, B, C, D and E respectively at day 14. There were also corresponding increases on day 28 to 10.34 µg/L, 11.95 µg/L, 13.59 µg/L, 15.48 µg/L and 9.72 µg/L showing 19.7%, 28%, 34.6%, 15.4% and 13.2% increase respectively.

Restriction on intake per body weight exists such as tolerable daily intake (TDI) of 0.05mg/kg body weight (bw)/day by European Food Safety Authority (EFSA, 2008). In this study, the highest increase in BPA level for control and sunlight exposed sample was 6.97µg/L and 15.48 µg/L respectively for Brand D. Using an average (crude) weight of men and women in the US for 2015/2016 as 89.7kg and 77.3kg (Fryar et al., 2018) and an average daily intake of 4.5litres for a physically active person (WHO, 2005), the daily intake for this study was calculated as 0.772µg/kg (bw)/day and 0.901µg/kg (bw)/day for men and women respectively using Eq. (1). For a child aged 3-6months assuming the same amount of BPA is transferred through water intake of 1.171L/day (95th Percentile) (US EPA, 2011), the child will have a BPA level of 18.112µg/day (without dividing by average weight) or 3.012µg/kg (bw)/day for a child weighing 6kg Eq. (1).

$$\text{Daily intake} = A * \frac{B}{C} \qquad\qquad (1)$$

[Where: Daily intake $of\ Bisphenol\ A\ for\ this\ study\ (\mu g/kg(bw)/day)$, A = Max Bisphenol A level in this study $(\mu g/L)$, B = Water standard daily intake (L/day), C = Average body weight $(kg\ (bw)w)$]

The TDI stipulated by EFSA (2008) is more than those calculated for men, women and children (0.772µg/kg (bw)/day, 0.901µg/kg (bw)/day and 3.012µg/kg (bw)/day respectively), implying that there is no significant risk to a person based on crude average weight (male, female, children) and recommended average daily water intake (for physical activity).

3.4 Nitrate

The values for Nitrate obtained from the tests carried out for the 5 brands of bottled water are as given in Fig 4. The baseline (day zero) Nitrate levels were 0.22mg/L, 1mg/L and 0.05mg/L for Brands B, C and D respectively; while Brands A and E were not detected as seen in Fig. 4. The control samples on day 14 had Nitrate level of 0.54mg/L, 1.21mg/L, 2.41mg/L 0.93mg/L and 0.62mg/L for Brands A, B, C, D and E respectively. After the 28th day, the Nitrate level increased by 142.5%, 100.8%, 60.2%, 101.1% and 151.6% for Brands A, B, C, D and E respectively corresponding to 1.31mg/L, 2.43mg/L, 3.86mg/L, 1.87mg/L and 1.56mg/L. The samples exposed to sunlight showed levels of Nitrate after the 14th day as 4.54mg/L, 6.12mg/L, 7.71mg/L, 9.38mg/L and 6.79mg/L for Brands A, B, C, D and E respectively. An increase was noticed after the 28th day of 66.1%, 56.2%, 51%, 63.2% and 56.6% corresponding to 7.54mg/L, 9.56mg/L, 11.64mg/L, 15.31mg/L and 10.63mg/L for Brands A, B, C, D and E respectively. The increase in Nitrate can be attributed to the thermal degradation of the polymers in the PET bottle as a result of temperature increase. This is evidenced by research on temperature effects on bottled water by Muhamad et al. (2011), they found that from 35°C 'molecular degradation as a result of overheating' causes

chemical parameters (including Nitrate) to be altered. Fig 4 shows temperature of 35°C was exceeded on day 14 and day 28 for samples exposed to sunlight.

The results showed that all levels of Nitrate in the control did not exceed the levels for US EPA (10mg/L) or that of the NSDWQ, WHO or EU (50mg/L). For samples exposed to sunlight, significant increase was noticed in the level of Nitrate as the study progressed leading (after the 28th day) to Brands C, D and E exceeding the US EPA (2009) regulation for Nitrates in drinking water. Another study (Muhamad et al., 2011), also recorded increase in Nitrate as temperature increased, though, the Nitrate levels were not as high as in this study. Lastly, all Nitrate values (100%) in this study still fell below 50mg/L standards of WHO (2006) and NSDWQ (2007).

4 Conclusion

Leaching of chemicals from PET bottles have been investigated by (Abboudi et al., 2018; Al-Khatim et al., 2019; Zmit and Belhaneche-Bensemra, 2019). This study found that for each of the parameters the following conclusions apply:

1. The pH for commercially available brands of bottled water is affected by sunlight exposure and based on the conditions of storage and sale will be a potential source of ill health when consumed.
2. Sunlight exposure accelerates the leaching of Antimony and Nitrates overtime as reported by similar studies. This is in addition to the characteristic of plastic bottled containers used for storing bottle water.
3. Though sunlight exposure increases the leaching of Bisphenol A, it does not translate to any significant threat to health when consumed in line with current legislation by European Food and Safety Agency (EFSA).
4. The level of Nitrate leached as a result of sunlight exposure is within the limit set by NSDWQ and WHO.

Author contribution

Rose Edwin Daffi conceptualized the study, developed the methodology and carried out the investigation. Fwangmun Benard Wamyil carried out the analysis or results. Both authors contributed significantly to writing the draft, proof reading and editing the report.

Competing interests

The authors declare that they have no conflict of interest

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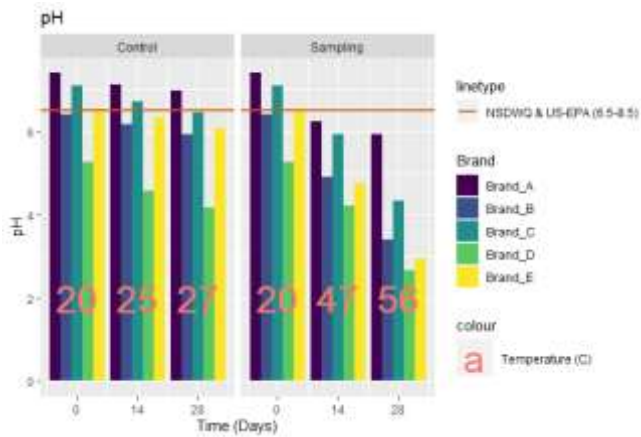
Table 1: Regulated Standards of Selected Physical, Chemical, Biological and Radiological Parameters

Parameter	NSDWQ[a]	WHO[b]	US EPA[c] [FDA]	EU-DWD[d]	Government of Canada[e]	Suspected Health Effects[a,b,c,d,e]
Turbidity	5NTU		NA [5NTU]	4NTU	0.3-1NTU	Indicates inefficient water treatment process/distribution system.
Temperature	Ambient	-	-		<15°C	High temperature enhances microbial growth and problems associated with taste, odor, color and corrosion[a,e]
Color	15TCU	-	[15 color units]	20mg/L Pt/Co	<15TCU	Indicates unsafe water/presence of chemical or biological impurities.
pH	6.5 – 8.5	-	6-8.5		7-10.5	Acidic pH corrodes conduits & leach chemicals into water, irritate eyes.
Antimony (Sb)	-	20µg/L	6µg/L [6µg/L]	5.0µg/L	6µg/L	Affects blood (increase in cholesterol, decrease in sugar)
Arsenic (As)	0.2mg/L	10µg/L	[10µg/L]	10µg/L	10µg/L [10µg/L]	Affects Skin & nervous system, increases cancer risk
Lead (Pb)	0.01mg/L	10µg/L	15µg/L [5µg/L]	25µg/L	5µg/L [10µg/L]	Affects central and peripheral nervous system, cancer & kidney damage
E.coli	0cfu/100mL		0/100mL [0/100mL]	0/100mL	0/100mL [0/100mL]	Gastroenteric disease
Total coliforms	10cfu/100mL		0/100mL [0/100mL or <5% incidence/month]	0/100mL	0/100mL [10/100mL]	Gastroenteric disease
Alpha particles (α)(activity)	0.1Bq/L	0.5Bq/L	15pCi/L [15pCi/L]			Increases Cancer risks
Beta particles (β) (activity)	0.1Bq/L	1Bq/L	4 millirems per year [4 millirems per year]			Increases Cancer risks
Radium (Ra)-226 + Radium (Ra)-228	-	1Bq/L	5pCi/L [5pCi/L]		0.5Bq/L	Increases Cancer risks
Uranium	-	30µg/L	30µg/L [30µg/L]			Increases Cancer risks

[a](SON, 2007), [b](WHO, 2017), [c](FDA, 2018; US EPA, 2001, 2009), [d](Drinking Water Inspectorate, 2017), [e](Government of Canada, 1981, 2020a, 2020b; Health Canada, 2020)

330 [NSDWQ = Nigerian Standard for Drinking Water Quality, WHO = World Health Organization, US EPA = United States Environmental Protection Agency, EU-DWD = European Union Drinking Water Directive, TCU = True Color Units, NTU = Nephelometric Turbidity Units, Bq/L = Becquerel/Liter, pCi/L = picocuries per Liter, µg/L=micrograms/Liter, mg/L=milligrams/Liter, mL=milliliters, Pt/Co = Platinum-Cobalt scale]

335 **Figure 1: pH Values for Control Group and Sunlight Exposure Group.**

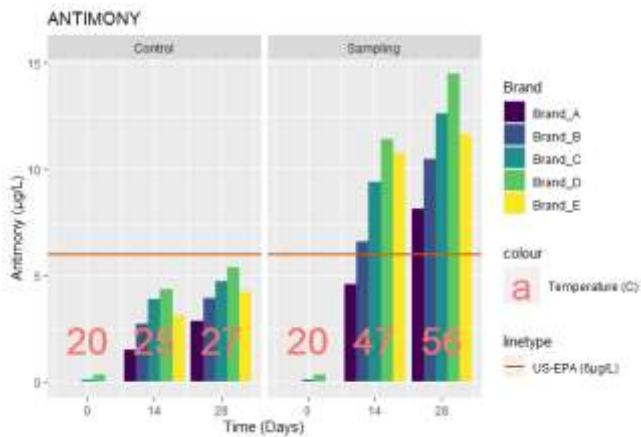
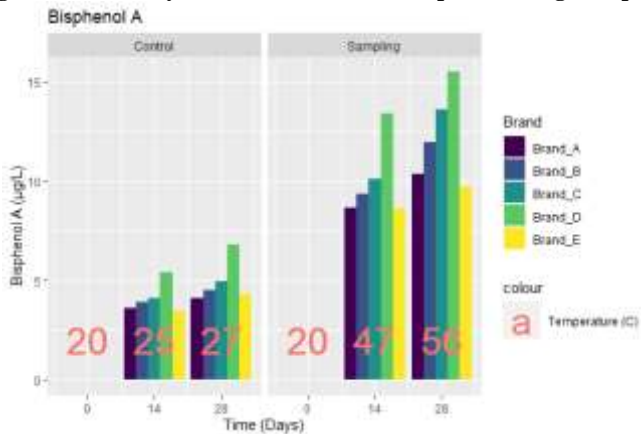



Figure 2: Antimony Values for Control Group and Sunlight Exposure Group.

340 **Figure 3: Bisphenol A Values for Control Group and Sunlight Exposure Group.**

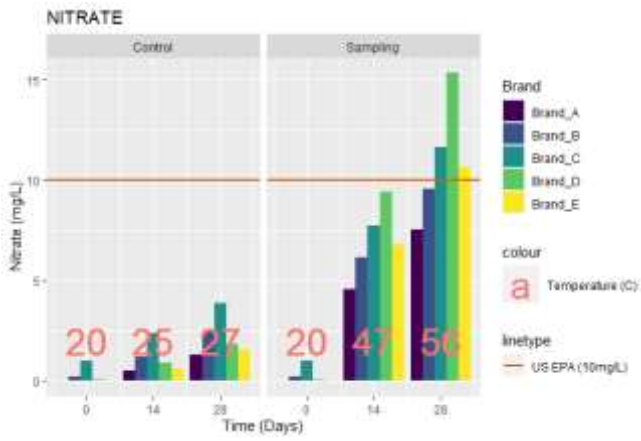



Figure 4: Nitrate Values for Control Group and Sunlight Exposure Group.