Journal of Medicinal Plants Research Vol. 6(19), pp. 3555-3560, 23 May, 2012
Available online at http://www.academicjournals.org/JMPR
DOI: 10.5897/JMPR12.013
ISSN 1996-0875 ©2012 Academic Journals

Full Length Research Paper

Assessment of nitrate and nitrite contamination in herbal tea products

Mohamed Yehia Abouleish* and Naser Abdo

Department of Biology, Chemistry and Environmental Sciences, College of Arts and Sciences, American University of Sharjah, P. O. Box 26666, Sharjah, United Arab Emirates.

Accepted 27 March, 2012

Herbal tea products, are used for medicinal purposes for infants, yet do not fall under strict regulations, like drugs. Herbal tea products, may lead to a cure, but under certain conditions may lead to serious health problems, due to over dosage, heavy metal contamination and presence of other contaminants and active ingredients. This research investigated the nitrate and nitrite contamination of herbal tea products sold in the market, as exceeding the United States Environmental Protection Agency (US EPA) permissible levels, could lead to serious health problems such as methemoglobinemia and cancer. This research indicated that some samples exceeded the permissible US EPA levels (10 mg/L nitrate-nitrogen and 1 mg/L nitrite-nitrogen), while other samples showed high levels but did not exceed the set standard limits, and in others no contamination was detected. This research presented important information about the contamination of herbal tea products and calls for stricter regulations for this industry, as these products are used as drugs but do not fall under strict drug regulations. Accordingly, this will ensure a safer product for our children and better health conditions.

Key words: Herbal tea, nitrates, nitrites, contamination, methemoglobinemia, United States Environmental Protection Agency (US EPA), WHO.

INTRODUCTION

The quality of the herbal tea products is of great concern, as they are used for medicinal purposes without being subjected to extensive clinical studies, and do not require a doctor's prescription (Lanski et al., 2003; Woolf, 2003). Herbal tea products are used by mothers to help with their children digestion, teething problems, gas, respiratory problems, and fussiness, in addition to being used as a beverage (Zhang et al., 2011). Herbal teas were shown to be used as early as one month of age (Zhang et al., 2011).

Herbal tea products do not fall under strict regulations that involve quality control testing and accurate identification of the herbal plants and their concentration, and that control manufacturer processes and labeling claims, and accordingly there is a concern about their purity, effectiveness, level of contaminants, and the interaction with other herbs and drugs that may be experienced (Kemper et al., 2008; Woolf, 2003). Herbal tea products are sold under many brands, and may contain many other ingredients of natural herbs, oils, plants, chemicals, and contaminants, which raises concern over the toxicity and adverse effects of the herbal tea products (Woolf, 2003). If such product is administered at high doses, it may affect children more than adults, due to size difference, body function and ability to detoxify them, and as a result may lead to health problems and death if not treated properly (Bakerink et al., 1996; Ize-Ludlow et al., 2004; Saper et al., 2004; Tomassoni and Simone, 2001; Woolf, 2003).

Herbal tea products may also pose other health problems, were contaminated soils and improper cleaning and manufacturing processes, may lead to the presence of microorganisms, heavy metals such as arsenic and mercury (Rembialkowska, 2007; Saper et al., 2004; Woolf, 2003) and other chemical contaminants such as nitrate (NO_3^-) and nitrite (NO_2^-), in the herbal plants used for the production.Several research showed concern over the presence of nitrate and nitrite in the prepared food and water, as they may lead to serious

*Corresponding author. E-mail: mabouleish@aus.edu. Tel: 971-6-515-2480. Fax: 971-6-515-2450.

health problems, such as methemoglobinemia (Adam, 1980; ATSDR, 2007-2010; BruningFann and Kaneene, 1994; Casanova et al., 2006; Egboka, 1984; Eichholzer and Gutzwiller, 2003; Gapper et al., 2004; George et al., 2006, 2007; L'Hirondel and L'Hirondel, 2002; McMullen et al., 2005; Merino et al., 2000; WHO, 1992). Methemoglobinemia "Blue Baby Syndrome" is a condition in infants, where nitrite combines with hemoglobin and decreases the ability of blood to carry oxygen (BruningFann and Kaneene, 1994; Casanova et al., 2006; Eichholzer and Gutzwiller, 2003; L'Hirondel and L'Hirondel, 2002; McMullen et al., 2005; Santamaria, 2006). Methemoglobinemia, if not recognized and treated, even though fatalities caused by it are rare and treatments are available and can reverse the conditions rapidly, it might lead to drastic conditions (L'Hirondel and L'Hirondel, 2002). Nitrate contamination can also lead to the formation of nitrite in the body, which can potentially lead to the formation of carcinogenic N-nitroso compounds, in adults (BruningFann and Kaneene, 1994; Casanova et al., 2006; Eichholzer and Gutzwiller, 2003; McMullen et al., 2005; Merino et al., 2000; Rembialkowska, 2007), and can affect pregnant woman and her fetus (ATSDR, 2007-2010).

Nitrate and nitrite exist naturally at low levels in the environment, yet human activities increase their levels, and can lead to both environmental and health problems. Elevated amounts of nitrate and nitrite can exist in our drinking water, as a result of agricultural activities that involve the excessive use of fertilizers and animal waste, runoff, leaching and release of untreated sewage and industrial waste (Adam, 1980; ATSDR, 2007-2010; Egboka, 1984; George et al., 2006; George et al., 2007; L'Hirondel and L'Hirondel, 2002; Merino et al., 2000; Santamaria, 2006; WHO, 1992). Nitrate and nitrite levels can also exist in food products as a result of food processing, use of preservatives and improper storage, for example, they have been found in grains and vegetables (Eichholzer and Gutzwiller, 2003; Sanchez-Echaniz et al., 2001).

As a result, the United States Environmental Protection Agency (US EPA) has set a Maximum Contaminant Level Goal (MCLG) of nitrate-nitrogen at 10 mg/L and nitrite-nitrogen at 1 mg/L in our drinking water (US EPA, 2009).

Many research studied the nitrate and nitrite levels in the final prepared food or beverage product, that is, after it is prepared with water, and considered the source of nitrate and nitrite contamination to be mainly from the water used, yet few studies were done on the plants or ingredients that were used in the preparation and their contribution to the overall contamination levels in the final product. Nitrates and nitrites not only find their way in our drinking water, but also into our plants and food. Therefore, the goal of this study is to investigate the nitrate and nitrite contamination contributed only from the herbal tea products that are sold in the United Arab Emirates (UAE), for infants and children intake, and as a result demonstrate their contribution to the overall contamination after it is prepared with water, and accordingly reflect on the quality of the herbal teas, and on the need for stricter regulations for the production of such products that are used as an alternative to prescribed drugs.

MATERIALS AND METHODS

Herbal tea samples

Ten brands of herbal tea products, representing all of the brands that are marketed for infant/children intake in the UAE market, were purchased from local markets in Dubai, Abu Dhabi, and Sharjah in UAE (Table 1). The samples varied in ingredients, texture, form, smell and flavor (Table 1). All the samples were in the processed form when purchased and were in the form of small dried pellets, except for sample 7 that was unprocessed (Figure 1). The samples were stored at similar conditions to the local market, and were analyzed after purchase. The herbal tea samples were tested for nitrate and nitrite anions.

The herbal tea samples were prepared by dissolving 1 g of the sample in 10 ml de-ionized water (Millipore "Simplicity" Purification System, USA), and were filtered through a 0.45 μm membrane filter (Schleicher and Schuell, UK), prior to ion chromatography (IC) analysis, without heating. Sample 7 was prepared in the same way as the other samples, but to speed up the dissolving process, it was sonicated for 5 min before filtration.

Analysis of the anions

The concentration of the anions were measured at room temperature using waters IC system, consisting of IC-Pak Anion HR 4.6 × 75 mm column, 616 pump with 600 S controller, 717plus autosampler, 432 conductivity detector and Millennium 32 software. The isocratic mobile phase was prepared by mixing 20 ml of n-butanol (high performance liquid chromatography (HPLC) grade, Hipersolv, UK), 120 ml of acetonitrile (HPLC grade, Hipersolv, UK), 20 ml of sodium borate/gluconate concentrate, then diluted to 1 L by de-ionized water, then homogenized, filtered through 0.2 μm membrane filter (Schleicher & Schuell, Germany) and degassed by sonication. The IC flow rate was set at 1.0 ml/min. The autosampler injection volume was 50 μl for the standards, and 5 to 100 μl for the samples depending on the anion concentration in the samples.

A multi ion standard (Seven Anion Standard II Standard, Dionex, USA) was used as the standard stock solution, from which the different calibration curves (correlation coefficient ≥ 0.99), for all of the measured anions were developed. The calibration curves were prepared with a concentration range of 0 to 50 mg/L nitrate and nitrite. The detection limits of nitrate-nitrogen and nitrite-nitrogen were 0.12 and 0.05 mg/L, respectively.

Recovery of the applied method of analysis of the anions, was accomplished by spiking the prepared herbal tea samples with aliquots of the anion standard, and then was analyzed. Recovery of nitrate and nitrite in the analyzed spiked samples ranged from 98.62 to 106.7% for nitrate, and 98.69 to102.6% for nitrite, with a standard deviation ranging from +/- 0.004 to 0.05.

RESULTS AND DISCUSSION

Nitrate and nitrite contamination of the herbal tea products was the main concern of this research. In the literature, many research and sources report the nitrate

Table 1. Description of herbal tea samples: ingredients and age intake recommendation.

Herbal sample	Herbal sample description				
	Herbal ingredients	**Other ingredients**	**Recommended for month**	**Processed (P) or non-processed (N)**	**Origin**
1	Fennel	Glucose, sucrose	Birth	P	Germany
2	Chamomile	Glucose, sucrose	2	P	Germany
3	Chamomile, balm, peppermint, fennel, anise, thyme	Dextrose, maltodextrin	6	P	Germany
4	Fennel	Dextrose, sucrose, maltodextrin	6	P	Germany
5	Herbal extract	Glucose, sucrose	4	P	Germany
6	Hibiscus, rose hip, raspberry, orange, peach	Dextrose, sugar, maltodextrin, citric acid, ascorbic acid, vegetable oil	6	P	Germany
7	Fennel	None	Birth	N	Germany
8	Fennel	Dextrose, sucrose, maltodextrin	6	P	Germany
9	Chamomile, balm, peppermint, fennel, anise, thyme	Dextrose, maltodextrin	6	P	Germany
10	Chamomile	Glucose, sucrose	2	P	Germany

and nitrite levels in water and food, without mentioning, whether it is the nitrate-nitrate or nitrate-nitrogen parameter that is used for example, similarly for nitrite levels. The US EPA states that the amount of permissible nitrate and nitrite levels (10 and 1 mg/L, respectively), correspond to the nitrogen forms of these anions, for example nitrate-nitrogen (US EPA, 2009). In Table 2, we report the results in both parameters for comparison, first to show that there is a difference between the values of the corresponding parameters and second to emphasis the necessity to mention which form of nitrate-nitrate or nitrate-nitrogen parameter is reported, for example, in the literature, as this could lead to a misunderstanding, which could lead to serious health problems, if high levels exist. In this research, the nitrate- nitrogen and nitrite-nitrogen parameters will be discussed, to be in agreement with the US EPA guidelines and the literature.

Nitrate-nitrogen was detected in six of the ten analyzed samples, with contamination levels that ranged from 2.8 to 12.0 mg/L (Table 2). Only two samples demonstrated nitrite-nitrogen contamination, 0.7 and 4.3 mg/L (Table 2).

Results showed that all the samples which contained nitrate-nitrogen and nitrite-nitrogen, did not exceed the US EPA allowed MCLG of 10 mg/L for nitrate-nitrogen, and 1 mg/L for nitrite-nitrogen, except for sample 5 for nitrate-nitrogen, and sample 1 for nitrite-nitrogen (Table 2 and Figure 2). Samples 1, 2, 3 and 9 contained high levels of nitrate-nitrogen, and also sample 3 contained high level of nitrite-nitrogen, but were all still below the allowed US EPA MCLG (Table 2 and Figure 2). High levels of nitrate-nitrogen and nitrite-nitrogen in the herbal tea products could be attributed to the use of fertilizers and other sources as mentioned earlier, or from the production process which may involve the use of preservatives. Such high levels may lead to a serious problem, especially, when such products are prepared with drinking water that already have some nitrate and nitrite contamination, which will lead to even higher levels and as a result exceed the US EPA set MCLG. This is especially important when such products are prepared and given to infants under 4 to 6 months of age (Knobeloch et al., 2000; McMullen et al., 2005). Infants under this age have a stomach pH higher than 4, which can allow more nitrate to change to nitrite, presenting more danger (WHO, 2007). Nitrite is more dangerous as it has the ability to form carcinogenic compounds by more danger (WHO, 2007). Nitrite is more dangerous as it has the ability to form carcinogenic compounds by

A B

Figure 1. Herbal samples obtained from the UAE market. Herbal sample in the processed pellet form (A), and natural unprocessed form (B).

Table 2. Nitrate and nitrite levels (mg/L) in the analyzed herbal tea samples. Nitrate and nitrite levels are expressed in two different parameters with a standard deviation (SD) that ranged from ± 0.0 to 2.9.

Herbal sample	Nitrate-Nitrate	Nitrite-Nitrite	Nitrate-Nitrogen	Nitrite-Nitrogen
1	35.4	14.3	7.9	4.3
2	31.5	0.0	7.0	0.0
3	44.2	2.4	9.8	0.7
4	0.0	0.0	0.0	0.0
5	54.2	0.0	12.0	0.0
6	0.0	0.0	0.0	0.0
7	0.0	0.0	0.0	0.0
8	0.0	0.0	0.0	0.0
9	38.0	0.0	8.4	0.0
10	12.7	0.0	2.8	0.0

reacting with the naturally present amines in the body, under the right conditions and concentrations (McMullen et al., 2005). In addition, nitrate and nitrite contribute directly and indirectly to methemoglobinemia in infants, which can cause other serious health problems (Knobeloch et al., 2000).

Comparing the results of the analyzed samples, it was shown that, when detected, the nitrate-nitrogen levels exceeded the nitrite-nitrogen levels in each sample, which is consistent with several thoughts (Table 2). Nitrates are more stable than nitrites (Okafor and Ogbonna, 2003). Nitrates usually exist in small amounts in plants, but under certain conditions, such as when inadequate amount of water is available, and when there are changes in temperature and energy, and when excessive amounts of fertilizers and animal waste are used, nitrate levels can increase and become stored in the stalk and parts of the leaves of the plants, and accordingly, contaminate the food (Undersander et al., (www.uwex.edu)).

The label of the product act as an important source of information for the consumer, as it reflects on the contents and quality of the product. For all the analyzed products, the labels included information on the ingredients, calorie content and only one product reported the calcium content and another reported the sodium content. Results, in this research demonstrated the importance of testing for nitrate and nitrite in the herbal tea products and as a result, call for the reporting of the nitrate and nitrite levels on the labels of the products, as they may lead to serious health problems, if the permissible levels are exceeded.

In conclusion, this research presented the need for reporting the nitrate and nitrite contamination levels in the literature, using unified parameters, for example, using the nitrate-nitrogen parameter that is required by the US EPA versus nitrate-nitrate parameter, and not using them interchangeably without reporting which parameter is used, as this could lead to misinterpretation of the results. This research demonstrated that nitrate and nitrite

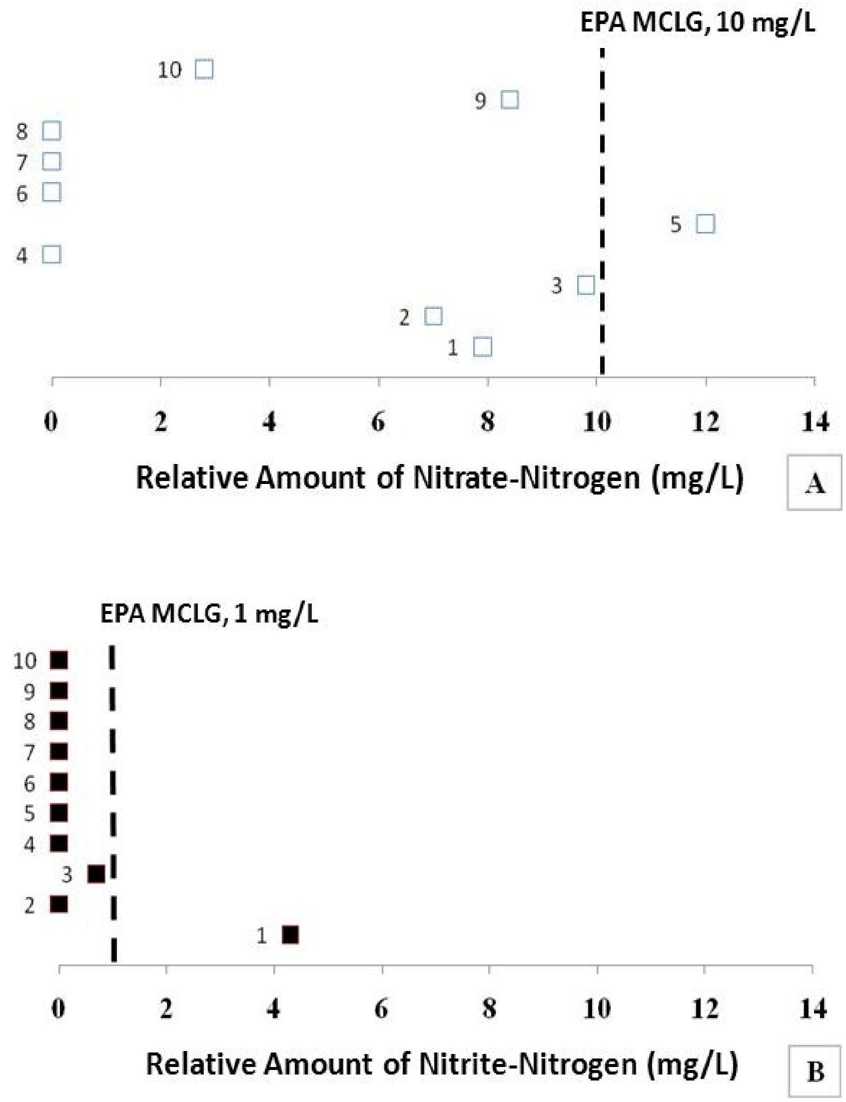

Figure 2. Nitrate-Nitrogen (A) and Nitrite-Nitrogen (B) levels in herbal tea samples, compared to the EPA MCLG (Dashed vertical line). Herbal tea samples are represented by (□) and (■).

contamination of the herbal tea products, after they are prepared with water, could result from the herbs themselves, and not only contributed from the drinking water used for the preparation. In other words, such contamination levels could have resulted from the contamination of the plants themselves, due to excessive use of fertilizers and harsh environmental conditions, and or they could have resulted from the manufacturing process itself.

Accordingly, special attention should be given to their use, manufacturing processes, information reporting and labeling and contamination. Herbal tea products do not fall under strict regulations like drugs, but since nitrate and nitrite contamination of the samples was detected and herbal tea products are used for medicinal purposes, then this research calls for stricter testing and regulations for this industry, in order to make sure that the products are safe for consumption by our children.

ACKNOWLEDGEMENT

The authors wish to thank the American University of Sharjah, for supporting this work.

REFERENCES

Adam JWH (1980). Health aspects of nitrate in drinking-water and possible means of denitrification (Literature Review). Water SA, 6: 79-84.

ATSDR, Agency for Toxic Substances and Disease Registry (2007-2010). Case studies in environmental medicine (CSEM): Nitrate/Nitrite Toxicity. Atlanta: US Department of Health and Human

Services. [Online] Available: www.atsdr.cdc.gov/csem/csem.html.
Bakerink JA, Gospe Jr SM, Dimand RJ, Eldridge MW (1996). Multiple organ failure after ingestion of pennyroyal oil from herbal tea in two infants. Pediatr., 98: 944-947.
BruningFann CS, Kaneene JB (1994). FDA poisonous plant database: the effect of nitrate, nitrite and n-nitroso compounds on human health, a Review. Vet. Hum. Toxicol., 35: 521-538.
Casanova JA, Gross LK, McMullen SE, Schenck FJ (2006). Use of griess reagent containing vanadium (iii) for post-column derivatization and simultaneous determination of nitrite and nitrate in baby food. J. AOAC Int., 89: 447-451.
Egboka BC (1984). Nitrate contamination of shallow groundwaters in Ontario, Canada. Sci. Total Environ., 35: 53-70.
Eichholzer M, Gutzwiller F (2003). Dietary nitrates, nitrites and n-nitroso compounds and cancer risk with special emphasis on the epidemiological evidence. In: D'Mello JPF (ed) Food Safety: Contaminants and Toxins. Massachusetts: CABI Publishing, pp. 217-234.
Gapper LW, Fong BY, Otter DE, Indyk HE, Woollard DC (2004). Determination of nitrite and nitrate in dairy products by ion exchange LC with spectrophotometric detection. Int. Dairy J., 14: 881-887.
George S, Rasmussen W, Ofitserova M, Pickering M (2006). Nitrite and Nitrate Analysis: Simultaneous Determination of Nitrite and Nitrate in Baby Foods. California: Pickering Laboratories, Inc. [Online]
Available: www.pickeringlabs.com.
George S, Ofitserova M, Pickering M (2007). Nitrite and Nitrate Analysis: Simultaneous Determination of Residual Nitrite and Nitrate in Processed Meats. California: Pickering Laboratories, Inc. [Online]
Available: www.pickeringlabs.com.
Ize-Ludlow D, Ragone S, Bernstein J, Bruck I, Duchowny M, Pena B (2004). Chemical composition of Chinese star anise (Illiciumverum) and neurotoxicity in infants. JAMA, 291: 562-563.
Kemper KJ, Vohra S, Walls R (2008). The Use of Complementary and Alternative Medicine in Pediatrics. Pediatr., 122: 1374-1386.
Knobeloch L, Salna B, Hogan A, Postle J, Anderson H (2000). Blue babies and nitrate-contaminated well water. Environ. Health, Perspect., 108: 675-678.
L'Hirondel J, L'Hirondel J-L (2002). Nitrate and Man: Toxic, Harmless or Beneficial? New York: CABI Publishing, pp. 1-94.
Lanski S, Greenwald M, Perkins A, Simon H (2003). Herbal therapy use in a pediatric emergency department population: expect the unexpected. Pediatric, 111: 981-985.
McMullen SE, Casanova JA, Gross LK, Schenck FJ (2005). Ion chromatographic determination of nitrate and nitrite in vegetable and fruit baby foods. J. AOAC Int., 88: 1793-1796.
Merino L, Edberg U, Fuchs G, Aman P (2000). Liquid chromatographic determination of residual nitrite/nitrate in foods: NMKL collaborative study. J. AOAC Int., 83: 365-376.
Okafor P, Ogbonna U (2003). Nitrate and nitrite contamination of water sources and fruit juices marketed in south-eastern Nigeria. J. Food Compos. Anal., 16: 213-218.
Rembialkowska E (2007). Quality of plant products from organic agriculture, J. Sci. Food, Agric,. 87: 2757-2762.
Sanchez-Echaniz J, Benito-Fernández J, Mintegui-Raso S (2001). Methemoglobinemia and consumption of vegetables in infants. Pediatric, 107: 1024-1028.
Santamaria, P (2006). Nitrate in vegetables: toxicity, content, intake and EC regulation. J. Sci. Food Agric., 86:10-17.
Saper RB, Kales SN, Paquin J, Burns MJ, Eisenberg DM, Davis RB, Phillips RS (2004). Heavy metal content of ayurvedic herbal medicine products. JAMA, 292: 2868-2873.
Tomassoni A, Simone K (2001). Herbal medicines for children: an illusion of safety? Curr. Opin. Pediatr., 13: 162–169.
US EPA, United States Environmental Protection Agency (2009). National Primary Drinking Water Regulations, EPA 816-F-09-004. [Online]
Available: water.epa.gov/drink/contaminants/upload/mcl-2.pdf.
WHO, World Health Organization (1992). Analytical and Water Treatment Methods for Health-Based Guideline Values. Report on a WHO consultation, Medmenham, UK, 27-29 January. Copenhagen: WHO Regional Office for Europe.
WHO, World Health Organization (2007). Nitrate and Nitrite in Drinking-Water: Background Document for Development of WHO Guidelines for Drinking-Water Quality, WHO/SDE/WSH/07.01/16. [Online].
Available:www.who.int/water_sanitation_health/dwq/chemicals/nitratesnitrite/en/index.html
Woolf AD (2003). Herbal remedies and children: do they work? are they harmful? Pediatric, 112: 240 –246.
Zhang Y, Fein EB, Fein SB (2011). Feeding of dietary botanical supplements and teas to infants in the United States. Pediatric., 127: 1060-1066.