Analysis of Fluoride Content in Alternative Milk Beverages

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Background: *The dentist has a responsibility to provide nutritional counseling and fluoride consumption recommendations. The purpose of this study was to measure and compare the concentrations of fluoride in a large number of alternative milk beverages and bovine milk.* ***Study design:*** *Thirty-three milk alternatives, including 9 diverse types and 11 different brands, were analyzed using a fluoride ion-selective electrode (ISE) and an ISE meter. Fluoride concentrations were then compared among different types and between different brands.* ***Results:*** *Fluoride concentration ranged from 0.01 ppm (Malk® Pure Cashew Milk) to 0.80 ppm (Almond Breeze® Original Unsweetened Almond Milk) with a mean concentration of 0.32 ppm. When compared, bovine whole milk (0.03*±**0.00** *ppm) was found to be significantly lower in fluoride than all samples analyzed except Malk Pure Cashew Milk, Soy Milk Vanilla, Rice Milk, and Pecan Milk. Major differences also existed between the same milk alternative types of different brands.* ***Conclusion:*** *The amount of fluoride varies among different types of milk alternatives and different brands. To ensure that the dental team can provide proper recommendations regarding fluoride use, manufacturers should consider placing fluoride concentrations on nutrition labels.*

Keywords: Fluoride, milk beverages, fluorosis, pediatric dentistry

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INTRODUCTION

Due to its role in dental caries prevention, fluoride can be credited for the vast improvement in oral health. However, when ingested in critical amounts, fluoride can also cause enamel fluorosis [1]. Dental fluorosis is a condition in which the structural makeup of enamel is weakened due to an increased amount of ingestion of fluoride while teeth are in the developmental phase [2]. Fluoride has a detrimental effect on ameloblasts, the cells that deposit enamel [3]. Most of the effects of fluorosis are minor in severity and limited only to esthetic concerns, such as white spots [4], although such effects can be psychologically distressful and difficult to treat. In the most severe forms, which is rare in occurrence, hypomineralization can lead to the development of phenomenon termed mottling, which is featured with porosity and extensive loss of surface enamel [1]. Severe mottling of the enamel can result in loss of integrity in which physiological functions can be affected [5]. A linear relationship exists between fluoride dose and enamel fluorosis in human populations [1].

The rise in dental fluorosis has been well documented since the mid-19980s and this increase is commonly attributed to ingestion of fluoridated toothpaste, the misuse of fluoride supplements, and the ever-increasing levels of fluoride in foods and drinks processed with fluoridated water [4]. The latter is the phenomenon known as the "halo" effect [6], and is the cause of the upward trend in fluorosis both in fluoridated and nonfluoridated regions [4]. The processed beverages being consumed include a variety of juices and milk alternatives.

At one time milk was the most widely consumed beverage for children in the United States; however, juice is now consumed more than milk [7]. In recent years, the American Academy of Pediatrics placed recommendations on the intake of juices in infants, due to the increased consumption of these beverages [7]. These recommendations state that children one to three years of age should have no more than four ounces per day, yet consumption is over twice the recommended amount in 10% of toddlers aged 19 to 24 months [8]. Using National Health and Nutrition Examination Survey 1994-2004 data, Wang *et al.* concluded that nearly half of children between the ages of two to five consume 100 percent juice at an average of 11.1 ounces per day [9].

Bovine milk consumption in children has decreased due to the surge in dairy alternatives such as soy, flax, rice and nut milks, which include almond, cashew, macadamia, pecan and coconut milk. Plant-based food sales have grown from $5.1 billion in 2013 to a projected $10.9 billion in 2019 [10]. This market is dominated by soy milk followed by almond milk and rice milk [10]. Total volume sales of bovine milk beverages have fallen more than 8% from 2013-2018 [11]. The switch to these alternatives can be a result of concerns about high fat and lactose content in bovine milk, lactose intolerance, implementation of plant-based diets, genetic engineering, antibiotics, hormones, and a greater awareness of animal abuse in factory farming [10,12-14]. Little research is available on the impact of these beverages on oral health although in vitro data suggests these milks have cariogenic potential [15].

The fluoride content of these beverages has not been reported, complicating the role of the dentist in providing dietary counseling and recommendations about fluoride consumption. The "optimal" daily suggested intake of fluoride for caries prevention is 0.05 to 0.07 mg per kilogram of body weight (mg/kg bw) [16], but, for fluorosis prevention, should not exceed 0.10 mg/kg bw [17]. With increasing availability of fluoride and undocumented levels in fluoride-containing products in our society today, keeping childhood intake within the suggested range becomes increasingly difficult. Therefore, the purpose of this study was to provide information about the fluoride content of these popular, milk-alternative beverages.

MATERIALS AND METHOD

Milk beverages

A total of 33 milk alternative beverages were analyzed in this study. The brand names (and abbreviations) and manufacturers of these milk beverages are listed in Table 1. They included seven soymilks, twelve almond milks, three coconut milks, four cashew milks, two flax milks, two macadamia milks, one pecan milk, one rice milk, and one whole bovine milk used as a control. These thirty-three alternative milk beverages include products from the following eleven brands: 365® (Whole Foods Market, Austin, TX), Almond Breeze® (Blue Diamond Growers, Sacramento, CA), Califia Farms® (Califia Farms, Los Angeles, CA), Forager Project® (Forager Project, San Francisco, CA), Good Karma® (Good Karma Foods, Inc., Boulder, CO), Horizon® (control) (The WhiteWave Food Company, Denver, CO), Malk® (Malk Organics, LLC, Houston, TX), Rice Dream® (Hain Celestial, Lake Success, NY), Royal Hawaiian Orchards® (MacFarms, LLC, Dana Point, CA), Silk® (The WhiteWave Food Company, Denver, CO), and So Delicious® (So Delicious Dairy Free, Springfield, OR).

Table 1. Milk beverages analyzed and the fluoride content*

Milk type and brand name#	Abbreviation	Fluoride in ppm Mean (±SD)
Forager Project® Unsweetened Plain Cashew-milk	FUC	0.54 (0.03)
Forager Project® Chocolate Cashew-milk	FCC	0.44 (0.01)
Forager Projectv Original Cashew-milk	FOC	0.46 (0.03)
Malk® Pure Cashew Milk	MPC	0.01 (0.00)
So Delicious® Coconut milk Vanilla	SCV	0.26 (0.02)
So Delicious® Coconut milk Unsweetened	SCU	0.29 (0.01)
So Delicious® Coconut milk Original	SCO	0.18 (0.02)
Good Karma® Flax milk Unsweetened	KFU	0.23 (0.01)
Good Karma® Flax milk Vanilla	KFV	0.14 (0.04)
Califia Farms® Toasted Coconut Almond Milk	CCA	0.22 (0.06)
Califia Farms® Unsweetened Almond Milk	CUA	0.30 (0.04)
Califia Farms® Vanilla Almond Milk	CVA	0.17 (0.02)
Califia Farms® Creamy Original Almond Milk	CCO	0.17 (0.05)
Almond Breeze® Unsweetened Vanilla Almond Milk	AUV	0.25 (0.04)
Almond Breeze® Unsweetened Chocolate Almond Milk	AUC	0.17 (0.00)
Almond Breeze® Original Unsweetened Almond Milk	AU	0.80 (0.04)
Almond Breeze® Original Almond Milk	AO	0.61 (0.04)
Silk® Chocolate Almond Milk	CHOC	0.66 (0.02)
Silk® Original Unsweetened Almond Milk	SU	0.70 (0.04)
Silk® Original Almond Milk	SO	0.74 (0.03)
Silk® Vanilla Almond Milk	VAN	0.71 (0.03)
Silk® Soymilk	SOY	0.19 (0.01)
Silk® Soy Milk Vanilla	SSV	0.21 (0.01)
Silk® Soy Milk Vanilla Lite	SLV	0.37 (0.01)
Silk® Soy Milk Light Chocolate	SLC	0.65 (0.05)
365® Soy Milk Original Light	3OL	0.15 (0.01)
365® Soy Milk Unsweetened	3SU	0.17 (0.01)
365® Light Soy Milk Vanilla	3LV	0.11 (0.00)
Rice Dream® Rice Milk	RDR	0.04 (0.00)
Royal Hawaiian Orchards® Original Macadamia Milk	OOM	0.23 (0.02)
Royal Hawaiian Orchards® Vanilla Macadamia Milk	OVM	0.20 (0.02)
Malk® Maple Pecan Milk	MMP	0.03 (0.02)
Horizon® Whole Milk	COW	0.03 (0.00)

*The fluoride content in parts per million (ppm) is expressed as mean (±standard deviation) of three separate assays.

#, Forager Project®, Forager Project, San Francisco, CA; Malk®, Malk Organics, LLC, Houston, TX; So Delicious®, So Delicious Dairy Free, Springfield, OR; Good Karma®, Good Karma Foods, Inc., Boulder, CO; Califia Farms®, Califia Farms, Los Angeles, CA; Almond Breeze®, Blue Diamond Growers, Sacramento, CA; Silk®, The WhiteWave Food Company, Denver, CO; 365®, Whole Foods Market, Austin, TX; Rice Dream®, Hain Celestial, Lake Success, NY; Royal Hawaiian Orchards®, MacFarms, LLC, Dana Point, CA); and Horizon® (control) (The WhiteWave Food Company, Denver, CO.

Fluoride analysis

For fluoride analysis, the milk containers were shaken briefly and gently, and a 3.0 ml aliquot was taken from each container. The fluoride concentration of the aliquot was then determined using a fluoride ion-selective electrode (ISE, 9609 BNWP, ThermoFisher Scientific, Fort Collins, CO) in conjunction with a pH/ISE meter (model 710, Thermo Orion, Beverly, MA). Five fluoride standards ranging from 0.1 ppm, 1 ppm, 5, ppm, 10 ppm, and 25 ppm were used to produce the calibration line [18,19]. To test effects of adjusting and stabilizing the total ion strength on the results, a set of experiments were also carried out by mixing 2.7 ml of milk samples with 0.3 ml of total ionic strength adjusting buffer (TISAB) III (Orion Ionplus, Chemlsford, MA) or TISAB I as recommended by the manufacturer to adjust the total ion strength and pH of the alternative milk beverages. All samples were analyzed three separate times to ensure the reliability and consistency.

Statistical analysis

Analysis of Variance (ANOVA) was first used to determine if significant differences exist among different milk products, and then pairwise Tukey's Studentized Range test was used to analyze the differences between different milk products. All statistical analyses were conducted using SAS 9.0 (SAS Institute Inc., Cary, NC). A difference at $P \leq 0.05$ is considered statistically significant.

RESULTS

Fluoride in nature exists in ionized, ionizable (relatively weakly bound) and strongly bound form. The bound fluoride can be released when the environmental pH is adjusted at or below 5.3 [20]. To investigate if adjustment of the beverages pH and ion strength have any major impact on the fluoride content, samples were treated with TISAB I and /or III before measurement. However, the results showed that such adjustments had no significant differences between the ones treated and those without receiving the treatment (data not shown), indicating that the bound fluoride is limited in these alternative milk beverages. Therefore, no additional treatment was carried out with the samples and only the fluoride ions were measured and recorded in this study.

The fluoride concentrations of all 33 samples were summarized in Table 1 and Figure 1. As a whole, the fluoride concentrations ranged from 0.01ppm (MPC) to 0.80 ppm (AU), with an average of 0.32 ppm. As a control, bovine whole milk (COW) contained 0.03 (±0.00) ppm of fluoride. When compared to COW and the other types, the highest fluoride concentration was found in almond milks with an average of 0.46 (±0.26) ppm (Figure 2). It was followed by cashew milks, soy milks, coconut milks, macadamia milks and flax milks, averaging 0.36 (±0.24) ppm, 0.26 (±0.22) ppm, 0.24 (±0.05) ppm, 0.21 (±0.00) ppm, 0.19 (±0.01), respectively. The least fluoride was measured with pecan and rice milks with an average of 0.03 (±0.02) ppm and 0.04 (±0.00) ppm, respectively.

Of the almond milks analyzed, AU contained the highest amount of fluoride, averaging 0.80 (±0.04) ppm, while AUC contained the lowest with an average of 0.17 (±0.00) ppm. Differences existed between the original almond milks (AO), the original unsweetened almond milks (AU) and those with addition of chocolate (AUC) and vanilla flavor (AUV). Relatively, addition of chocolate and vanilla seemed to lead to significant reduction of fluoride, such as CUA vs CVA. On the other hand, addition of chocolate to soymilk (SLC) or the processing needed to make this type of milk seemed to increase fluoride content (SLC vs SOY)

Cashew milks contained an average of 0.360 (±0.24) ppm of fluoride with the highest being measured in FUC at 0.54 (±0.03) ppm and the lowest measured with MPC at 0.01 (±0.00) ppm. Of the two different brands analyzed, Forager Project products were

Figure 1. Analysis of fluoride content in milk beverages. Data represent average (±standard deviation in error bars) of three independent assays. *The highest fluoride content was measured in AU, with *P*<0.05 vs all others except SO, VAN and SU. **The lowest fluoride was measured with MPC, *P*<0.05 vs all others except MMP, RDR and COW.

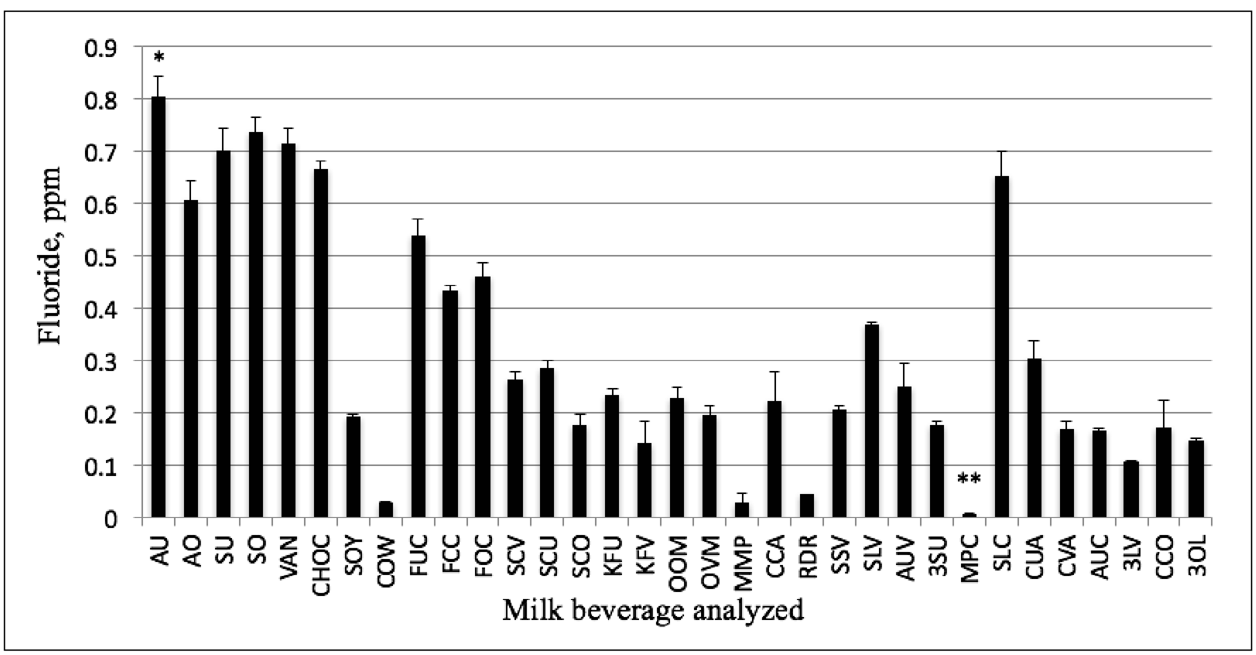
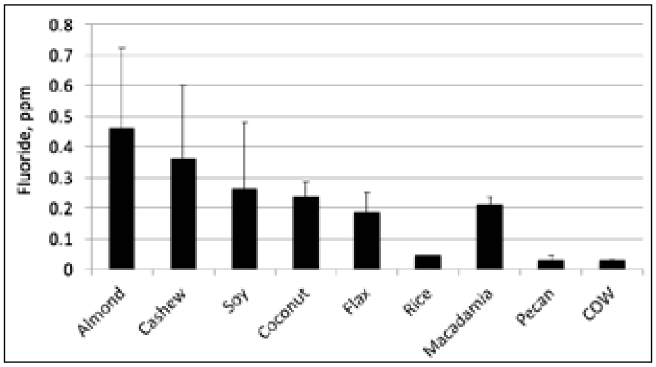


Figure 2. Comparison of fluoride content between different types of milk beverages specified. Data is expressed as mean (±standard deviation).



67-fold higher in fluoride content than the Malk product (MPC) (Figure 1). Major differences also found between the different manufacturers of the almond and soy milks (Table 1), which is at least part of the contributing factors to the deviations shown in Figure 2. Of the almond milks analyzed, Almond Breeze and Silk products possessed >2-fold higher fluoride than those by Califia Farms, such as the Breeze's SU and AU vs Califia Farms' CUA and the Breeze's VAN vs Califia Farms' CVA (Table 1). A similar phenomenon was also present in soymilk between Silk and 365, such as Silk's SSV and SLV vs 365's 3LV. On average, the Silk products had >2-fold higher fluoride than those of the 365 brand with an average of 0.36 (±0.22) ppm for the Silk brand and 0.14 (±0.03) for the 37 brand.

DISCUSSION

All milk beverages analyzed in this study contained fluoride and major differences exist between the different types of the alternative milk beverages. The highest fluoride concentrations were found in almond milks and the cashew milks (with the exception of MPC) and the least fluoride concentrations were found in rice milk (RDR) and pecan milk (MMP). Bovine whole milk (COW) possesses relatively low amount of fluoride which is well documented in the literature [21] and consistent with some of the recent studies [22]. The results also demonstrated that major differences exist between different brands and between products with or without addition of other ingredients and/or flavors.

Determining the fluoride content of commonly consumed beverages is a challenge for the dentist. Fluoride concentrations in water varies widely, as shown by Winkle SV *et al.* and Jackson RD *et al.* [23,24] The following average fluoride levels have been reported: unfiltered wells (0.45 ppm), filtered wells (0.32 ppm), filtered public supply (0.67 ppm), and bottled water (0.18 ppm) [23]. In comparison, the average fluoride concentration of the milk alternatives (0.33 ppm) presented here are very similar to those of the filtered wells reported by S. V. Winkle and coworkers.[23], but are lower than those of the unfiltered wells and the public water supply. However, the bottled water appears to have even lower levels of fluoride than the milk alternatives analyzed in this study. Still, traditional bovine milks appear to contain the least amounts of fluoride, on average.

Similar results have been reported in studies of fluoride content in other products such as fruit juices and soft drinks [7]. In the infant apple juices, for example, the fluoride level was found at 1.07(±0.11) ppm in Gerber products, 0.25 (±0.02) in Earth Best and 0.43±0.42 in Beech-Nut [7]. According to the United States Department of Agriculture [25], the most fluoride was found in grape juices at the level of 1.13 (±0.91) ppm, followed by apple juices and orange juices at 0.44 (±0.15) ppm and 0.43 (±0.15) ppm, respectively. Similar results were also found in infant juices with the most fluoride measured in grape juices at 1.21 (±0.34) ppm, followed by pear juices at 0.78 (±0.34) ppm and apple juices at 0.57 (±0.45) ppm [7]. According to analysis of the 1999-2004 National Health and Nutrition Examination Survey, approximately 30 percent of adolescents consume fruit juices and nearly 50% of children ages two to five consumed an average 11.1 oz per day [9]. According to Omar *et al.* [7], the average fluoride concentration in commonly consumed infant juices was 0.74 ppm, which is comparable to most of the almond and the cashew milks in this study. When compared to the combined average of all the alternative milks studied here, however, juices appear to contain approximately twice as much fluoride as the milk alternatives. Nevertheless, the commonly consumed infant juices as well as the alternative milk beverages contain approximately >27- and 11-times more fluoride than the traditional bovine milk.

Some groups have performed preliminary studies on the fluoride content of the soy milk beverages. In a recent study by Nagata *et al.* [22], soy milks available in Brazil were found to have an average fluoride concentration of 2.52 ppm, which is significantly higher than concentrations found in the present study (0.18 ppm). Conversely, fluoride concentrations of soy milks available in the United Kingdom have an average fluoride concentration of 0.29 ppm [26], which is similar to the findings of the current study. Likely, variations in fluoride content between different brands can be attributed to the differences in processing and fluoride content of ingredients. The use of fluoridated water or not in the production certainly can be a major contributor, so does the fluoride level in the soil where the almonds, the soybeans and other raw materials were grown. This is the first comprehensive report on the fluoride analysis of alternative milk beverages that include nut and rice milks.

Our study and literature review have shown that wide variations in fluoride content exist between the popular drinks. A general trend of fluoride concentrations, from the least to the greatest, is as follows: bovine milk, bottled water, infant formulas, milk alternatives analyzed (with the exception of rice and pecan milk beverages), filtered wells, unfiltered wells, filtered public water system, and juices. These wide variations make it challenging for dentists to advise families on fluoride use. Currently manufacturers are not required to report the fluoride levels on their products, but with the rising level of fluorosis, this information would be educational for consumers and dentists.

Strengths of the study include the wide variety of brands and flavors tested. To ensure reliability, the tests were conducted in triplicate. A limitation of this study involved the use of different number of samples for each brand and flavor. For example, twelve almond milks were tested, but only one pecan milk was selected due to market limitations. In addition, only bovine whole milk was tested as a control. Also, there is a possibility that variation exists between batches, and only one batch was tested in this study. Finally, the location of processing could not be determined by the packaging of the products. Follow-up studies could track ingestion patterns of these beverages in children through observation or market based research.

CONCLUSION

This study provides further evidence that the fluoride concentration varies between different alternative milk beverages, but with few exceptions, these alternative milk beverages contain significantly more fluoride than bovine milk. These results may guide dentists in dietary recommendations. Public health advocates should support initiatives to encourage beverage manufacturers to include fluoride concentrations per serving on the nutrition labels. This information could allow individuals to optimally use fluoride for caries reduction, while reducing the risk of fluorosis.

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