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Short Communication

Changes in the content and forms of vitamin K in processed foods

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ABSTRACT

High intake of *trans* fatty acids has been linked to deleterious health effects including increased risk of cardiovascular disease. Since 2006, the Food and Drug Administration requires companies to label the *trans* fatty acid content of foods. This has resulted in an overall decrease of commercially-hydrogenated oils in the food supply. Hydrogenation of vitamin K (VK)-rich plant oils changes the form and content of VK. It is not known if changes in use of hydrogenated oil in the U.S. food supply resulted in a change in the forms and amount of VK in processed foods. To test this, we compared 253 foods for total and individual forms of VK in foods analyzed pre- and post-2006 as part of the U.S. Department of Agriculture (USDA) Nutrient and Food Analysis Program. Overall, foods identified as rich sources of the hydrogenated form of VK pre-2006 (dihydrophylloquinone; dK) had lower amounts of dK post-2006 with a concomitant increase in the parent form of VK, phylloquinone. However, the range of dK was large within foods, suggestive of a wide range of current practices regarding use of hydrogenated oils in the U.S. food supply.
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1. Introduction

In response to the evidence regarding the deleterious health effects associated with the consumption of *trans* fatty acids, (Booyens et al., 1988; Katan et al., 1995; Korver and Katan, 2006) the Food and Drug Administration (FDA) now requires labeling of the amount of *trans* fatty acids contained per serving of food. *Trans* fatty acids are primarily produced when vegetable oils are commercially hydrogenated (Davidson et al., 1996). Since this FDA policy went into place in 2006, the presence of *trans* fatty acids in the food supply has been diminishing with a decrease in the use of partially hydrogenated oils and the increased use of *trans* fat-free spreads (Korver and Katan, 2006; Vesper et al., 2012).

Whereas the focus of the health consequences of hydrogenated oils has been on the formation and subsequent consumption of *trans* fatty acids, other nutrients, including vitamin K (VK), are similarly transformed during the process of hydrogenation of plant oils. The most common dietary form of VK is phylloquinone (PK), which is found primarily in green leafy vegetables and plant oils. When PK-rich plant oils are hydrogenated, overall VK content is reduced, with the majority of remaining PK being chemically transformed into dihydrophylloquinone (dK) (Davidson et al., 1996). Unlike *trans* fatty acids, which can be naturally occurring, dK is the exclusive product of commercial hydrogenation of plant oils, with no known natural sources. As such, dK is limited to those foods containing hydrogenated oils, such as commercially fried and baked foods (Dumont et al., 2003; Elder et al., 2006; Ferreira et al., 2006; Peterson et al., 2002). The recommended adequate intake (AI) for VK is 120 and 90 μg/d for men and women respectively. It has been demonstrated that dK is absorbed less than equimolar amounts of PK (Booth et al., 1999), and its intake is associated with lower bone mass. Since the presence of dK reflects the amount of hydrogenated oils in these food sources, it has also been used as a marker of an unhealthy diet (Troy et al., 2007). Therefore, reduction of *trans* fatty acids in the food supply should also reduce the presence of dK.

The purpose of this study was to analyze the PK and dK contents of commonly consumed foods that had previously been identified as rich in hydrogenated oils and compare their concentrations pre- and post-2006, since this was the implementation date of the FDA policy on *trans* fatty acid labeling.

2. Methods

The analysis of PK and dK in U.S. food samples is an ongoing collaborative agreement of the Vitamin K Laboratory at Tufts University with the U.S. Department of Agriculture (USDA). Food samples were obtained from the USDA Nutrient Data Laboratory as part of the National Food and Nutrient Analysis Program (Haytowitz et al., 2008). Samples were collected in 12 cities in the U.S. and combined to form either brand-specific or subnational composites as appropriate to the food item. This food sampling plan provides

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aliquots of homogenized foods that are representative of key foods consumed in the U.S. (Pehrsson et al., 2000). Food samples were shipped to the Food Analysis Laboratory Control Center at Virginia Polytechnic Institute and State University in Blacksburg, Virginia, for the preparation of aliquots and quality-control materials. Aliquots of the foods were then shipped on dry ice to the Vitamin K Laboratory at Tufts University, Boston, Massachusetts, and stored at −80 °C until analyzed. Vitamin K is stable under these conditions for a minimum of five years (unpublished data).

The PK and dK contents of the food samples were determined using a high-performance liquid chromatography (HPLC) procedure described elsewhere (Ferreira et al., 2006). The assay used to generate data pre-2006 is the same as used for current analysis and has remained stable, as confirmed through use of in-house control samples.

All samples were analyzed in duplicate. If the CV of duplicates was greater than 15% (for samples with PK concentrations >5 μg/100 g) the assay was repeated. For samples containing <5 μg/100 g of PK, the assay was repeated if the duplicate differed by greater than 0.75 μg/100 g. A control sample (consisting of an aliquot of baby food chicken vegetable dinner, Beechnut®, Amsterdam, NY) was run with each batch of foods. If the determined concentrations of the control sample varied by more than 2.5 standard deviations, the entire sample batch was rejected and rerun. Quantification was achieved by direct comparison of peak area ratios (PK or dK) to the assay standard K1(25) generated from the calibration standard to those generated by the sample. Peak integration and sample concentration calculations were performed using Waters Millennium32 software, version 3.20.

Food composition data for PK and dK obtained prior to 2006 have been previously reported (Dumont et al., 2003; Elder et al., 2006; Ferreira et al., 2006; Peterson et al., 2002) and incorporated into USDA National Nutrient Database for Standard Reference (http://www.ars.usda.gov/nutrientdata/sr). For the purpose of this study, data were selected for those foods analyzed prior to 2006 in which dK was present and for which we had repeated VK analysis in samples collected post-2006. We did not conduct further laboratory analysis on these samples for which data were available. Food samples were not matched for brand or sampling location in the pre- and post-2006 comparisons, and therefore can only be used to monitor overall trends.

Table 1
Changes in the PK and dK contents of selected foods pre and post *trans* fatty acid labeling.

Food	PK (μg/100 g)						dK (μg/100 g)*			
	Pre-2006			Post-2006			Pre-2006		Post-2006	
	N	Mean (SD)	Range	*N*	Mean (SD)	Range	Mean (SD)	Range	Mean (SD)	Range
Baked goods										
Cake										
Yellow cake, with chocolate icing	NA[a]	NA (NA)	NA	3	25.1 (3.6)	21.1–28.3	NA (NA)	NA	8.5 (8.1)	ND[b]–16.2
Chocolate cake, with chocolate icing	NA	NA (NA)	NA	3	28.9 (3.3)	25.3–31.7	NA (NA)	NA	5.8 (8.5)	ND–15.6
Cinnamon buns, packaged	4	14.0 (0.9)	12.8–14.9	6	10.1 (7.6)	1.7–20.4	22.8 (2.4)	19.4–24.5	2.5 (2.1)	ND–5.0
Cookies										
Oatmeal	NA	NA (NA)	NA	3	25.1 (16.3)	7.7–40.0	NA (NA)	NA	7.6 (13.2)	ND–22.9
Sugar Wafer	NA	NA (NA)	NA	4	2.1 (0.1)	2.0–2.2	NA (NA)	NA	2.0 (1.2)	2.0–2.1
Chocolate Chip	6	11.9 (9.0)	4.3–29.1	8	22.5 (17.1)	1.8–41.3	48.2 (38.5)	ND–79.7	0.3 (0.6)	ND–1.5
Chocolate Sandwich	2	22.8 (9.0)	16.4–29.2	7	27.4 (11.1)	5.6–40.9	0.0 (–[c])	NA	0.1 (0.2)	ND–0.5
Muffins, blueberry	2	39.3 (–)	32.6–45.9	6	30.7 (11.6)	16.3–44.8	1.0 (–)	ND–1.9	1.2 (1.3)	ND–2.6
Bread, rice and pasta										
Bread, specialty										
Sweet Bread	NA	NA (NA)	NA	2	1.7 (–)	1.6–1.8	NA (NA)	NA	10.5 (–)	3.1–17.8
Corn Bread	NA	NA (NA)	NA	2	7.8 (–)	6.2–9.3	NA (NA)	NA	9.7 (–)	7.9–11.5
Crackers										
Crackers, cheese	NA	NA (NA)	NA	2	9.4 (–)	8.1–10.6	NA (NA)	NA	ND (–)	–
Crackers, no cheese	20	24.3 (14.8)	7.1–52.0	13	19.5 (17.3)	2.2–69.55	17.3 (33.2)	ND–141.4	7.9 (9.7)	ND–24.7
Tortillas, fried	NA	NA (NA)	NA	3	25.8 (20.7)	11.9–49.7	NA (NA)	NA	21.9 (25.3)	ND–49.7
Lasagna with meat sauce	4	4.4 (3.1)	2.1–8.8	12	7.1 (1.7)	5.3–10.8	1.1 (0.5)	0.4–1.5	0.1 (0.2)	ND–0.4
Rice										
Spanish Rice Mix, prepared	NA	NA (NA)	NA	2	2.7 (–)	2.3–3.1	NA (NA)	NA	ND (–)	–
Spanish Rice Mix, unprepared	NA	NA (NA)	NA	2	3.1 (–)	2.5–3.7	NA (NA)	NA	0.8 (–)	ND–1.6
Fats and oils										
Margarine, 80% fat	8	93 (46)	50.8–163.0	3	53.2 (5.0)	47.4–56.5	111 (48)	68.9–182.0	67.4 (1.2)	66.6–68.7
Meat										
Chicken, breaded, fried	8	7.9 (6.4)	3.8–23.5	18	29.8 (8.9)	17.3–53.3	20.1 (16.7)	ND–35.3	0.1 (0.2)	ND–0.9
Chicken Pot Pie	7	3.9 (6.4)	0.5–18.2	2	14.0 (–)	1.5–26.5	1.8 (3.4)	ND–9.0	0.4 (–)	ND–0.8
Hush Puppies	NA	NA (NA)	NA	2	3.8 (–)	2.4–5.3	NA (NA)	NA	16.0 (–)	12.6–19.5
Corn dogs	NA	NA (NA)	NA	4	4.0 (4.7)	1.55–11.0	NA (NA)	NA	0.8 (0.7)	ND–1.5
Shrimp, breaded, fried	NA	NA (NA)	NA	2	6.4 (–)	1.9–11.0	NA (NA)	NA	32.3 (–)	39.9–24.8
Fast food										
French Fries	12	11.2 (4.5)	5.3–17.0	13	32.9 (7.7)	20.6–48.1	42.8 (18.1)	14.5–64.0	ND (0.1)	ND–0.2
Onion Rings, fried	NA	NA (NA)	NA	8	43.3 (9.6)	26.1–53.8	NA (NA)	NA	0.1 (0.1)	ND–0.4
Hamburger	14	11.2 (7.7)	1.1–23.4	3	6.2 (1.5)	4.7–7.7	0.3 (0.5)	ND–1.5	ND (0.0)	–
Miscellaneous										
Toaster Pastry	10	6.1 (1.4)	4.7–8.3	2	19.75 (–)	19.0–20.5	23.0 (5.0)	15.7–29.1	ND (–)	–
Popcorn, microwave, butter flavor, prepared	4	4.2 (2.0)	2.6–7.0	8	4.3 (1.7)	1.3–6.8	68.9 (16.3)	49.0–88.3	30.2 (34.9)	ND–80.1
Ramen noodle soup mix, unprepared	4	2.6 (0.3)	2.2–2.9	2	2.7 (–)	1.9–3.5	0.9 (0.4)	0.5–1.4	ND (–)	–
Tamale, corn	NA	NA (NA)	NA	3	5.4 (5.8)	1.1–12.0	NA (NA)	NA	1.1 (1.8)	ND–3.2

* Data entries for dK are the same as the corresponding PK values and can be found in the PK columns.
[a] NA = not available.
[b] ND = below the lower limit of detection (0.2 μg/100 g).
[c] For food items with 2 or less samples, no standard deviation was reported.

Data are presented as the mean ± standard deviation (SD) for each food product. Ranges are also given if there were greater than 2 samples per food product.

3. Results and discussion

Overall, foods that contained high amounts of dK prior to the 2006 *trans* fatty acid labeling requirement had lower amounts after the labeling requirement in 2006 (Table 1). In foods where dK was reduced post-2006, a concomitant increase in PK concentrations was often observed. This was most obvious in commercially-baked goods, such as chocolate chip cookies, which had almost a 100% decrease in average dK concentration and a concomitant increase in average PK concentration post-2006. This was an expected change because the hydrogenation process reduces the overall VK content of the plant oils during the conversion of PK to dK, such that hydrogenated oils contain up to 50–80% less VK (PK + dK) than the parent form of the plant oil (Davidson et al., 1996). Use of non-hydrogenated plant oils would predictably contain PK in higher amounts. However in some foods in which the dK was reduced post-2006, the PK content either did not change or was overall reduced. This may reflect a change in the use, or lack thereof, of hydrogenated oils in addition to an overall reduction in oils used or a shift to the use of different plant-based oils that do not contain high amounts of PK, such as peanut or cottonseed oils (Peterson et al., 2002). The bioavailability of PK obtained from plant-based oils is greater than PK obtained from vegetables (Booth et al., 2002), so these trends in changes in PK content of processed foods may influence the stability of oral anticoagulant therapy if these food items are consumed in large quantities.

An unexpected finding was the large variation in the dK content of foods analyzed post-2006. Whereas for some foods, dK was present in all samples analyzed pre-2006, the range of dK in samples analyzed post-2006 indicated a wide range of practices in terms of use of hydrogenated plant oils. For example, butter-flavored popcorn contained large amounts of dK in all samples analyzed pre-2006 but post-2006, dk content ranged from non-detectable to 80 μg/100 g of popcorn. This large range emphasizes the need for clear labeling for consumers to identify ingredients such as hydrogenated plant oils in their foods.

There were some food items analyzed in this study for which we do not have comparative data prior to 2006. However we chose to include these foods that contained dK, and hence hydrogenated plant oils. For example, fried tortillas contained high concentrations of both PK and dK, yet are not food items usually identified as a good dietary source of VK.

This analysis contains some limitations. This study addressed trends in VK content in a limited number of processed foods available in the U.S. food supply, and no attempt was made to identify changes at the regional or specific brand level. In addition, the list of foods reported here are not exhaustive nor did we have concomitant *trans* fatty acid contents for the same food samples as a direct comparison. However, given that dK is exclusively formed from commercial hydrogenation of plant oils, it is likely that the dK concentrations reflect similar trends in *trans* fatty acids in these same foods.

Since dK tracks hydrogenation of foods containing *trans* fatty acids (Troy et al., 2007), its reduction in the foods analyzed is consistent with reports by others that *trans* fatty acids in the U.S. food supply have decreased following the *trans* fatty acid labeling requirement (Angell et al., 2012; Downs et al., 2013; Vesper et al., 2012). Indeed, multiple studies have observed a decrease in the concentration of *trans* fatty acids in both food and human consumption and circulation (Angell et al., 2012; Downs et al., 2013; Vesper et al., 2012). Recently, the FDA has proposed removing partially hydrogenated oils from the generally recognized as safe (GRAS) list. If this were to occur, we would expect a further decrease in the dK content of processed foods (FDA, 2013).

4. Conclusion

Intakes of VK may be inadvertently variable in terms of form and absolute amounts due to the wide range in amounts and types of oils used in food manufacturing. These changes may in part be due to decreased amounts of hydrogenated oils used in processed foods in response to labeling requirements for *trans* fatty acids that have been in effect since 2006.

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