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Tocopherol and Tocotrienol Levels of Foods Consumed in Hawaii

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Because of the individual biological effects and the uncertain or missing information on levels of tocopherols (T) and tocotrienols (T3) in foods frequently consumed in Hawaii, 79 food items (50 in duplicate) were analyzed for α-, β-, γ-, and δ-tocopherol (αT, βT, γT, and δT) and α-, β-, γ-, and δ-tocotrienol (αT3, βT3, γT3, and δT3) in addition to α-tocopheryl acetate (αTac). Foods from local markets were stored according to usual household habits, freeze-dried, homogenized, and extracted three times with hexane containing butylated hydroxytoluene as a preservative and tocol as an internal standard. A normal-phase high-pressure liquid chromatography system was applied with fluorescence and photodiode array detection that resulted in baseline separation of all eight analytes and the internal standard tocol (To). The sum of all E vitamer concentrations, or total E vitamers (TEV), in all foods analyzed ranged an average from 0.6 to 828 mg/kg (T ≤ 542 mg/kg and T3 ≤ 432 mg/kg) and showed the following ranges: oils, 497−828 mg/kg (mainly αT and γT); margarines, 359−457 mg/kg (mainly γT); salad dressings, 20−291 mg/kg (mainly γT, except αT when soy oil was the main ingredient); cookies, 54−138 mg/kg (mainly γT); snacks, 101−220 mg/kg (mainly γT); nuts, 22−201 mg/kg (mainly αT); vegetables, 2−152 mg/kg (mainly αT); pasta, 24−90 mg/kg; cereals, 4−56 mg/kg (mainly βT3 followed by αT); fish, 2−39 mg/kg (mainly αT); fried tofu, 64 mg/kg (mainly γT); breads, 20−22 mg/kg (mainly βT3); fat-free mayonnaise, 5 mg/kg (mainly αT); poi (fermented taro root), 2 mg/kg (mostly αT); and fruits, 2 (papaya) to 13 mg/kg (canned pumpkin) with αT predominating. Cereals fortified with αTac ranked third and eighth among all foods assayed regarding αT and TEV levels, respectively. As compared to the few data available in the literature, our values agreed with some (corn flakes, mango fruit, fat-free mayonnaise, dry-roasted macadamia nuts, dry-roasted peanuts, mixed nuts, spaghetti/marinara pasta sauce, oils, and red bell pepper) but differed for many other items. Our results provide new information on the E vitamer content in foods, emphasize the vast differences of bioactivities of individual E vitamers, and confirm the need for analyses of foods consumed in specific study populations.

KEYWORDS: Vitamin E; tocopherols; tocotrienols; α-tocopheryl acetate; food levels

INTRODUCTION

Vitamin E is a fat-soluble vitamin that generally functions as a potent antioxidant via chain-breaking reactions during peroxidation of unsaturated lipids (*1*). It is particularly important in maintaining the integrity of cell membranes (*2*). The term "vitamin E" has traditionally encompassed several tocopherols (Ts) and tocotrienols (T3s), but because of differing vitamin activities, it is preferable to refer to these compounds as E vitamers. The Ts contain a saturated phytyl residue at C-2, and the T3s contain a triply unsaturated phytyl chain (**Figure 1**); T and T3 can each occur as four different configurations denoted by an α, β, γ, and δ prefix to describe the methylation pattern in the phenolic moiety of the molecule with none, one (C-7), one (C-5), and two (C-7 and C-5) unmethylated sites in the phenol ring, respectively (**Figure 1**). These eight individual vitamers of T and T3 occur in nature exclusively in the R conformation of all asymmetrical C atoms (C-2,-4′,8′ in T and C-2 in T3; **Figure 1**). Of all of the natural forms of T and T3, α-tocopherol (αT) has the greatest biological activity as measured by the traditional rat sterility (fetal resorption) test. The other forms have a lower activity in this test [β-tocopherol (βT) = 50%, α-tocotrienol (αT3) = 33%, γ-tocopherol (γT) = 10%, and δ-tocopherol (δT) = 3%] or none [β-tocotrienol (βT3), γ-tocotrienol (γT3), and δ-tocotrienol (δT3)] (*3*), leading

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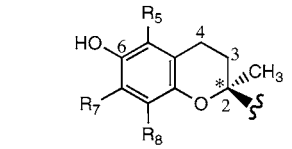
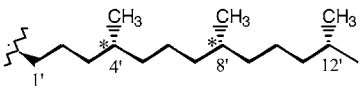
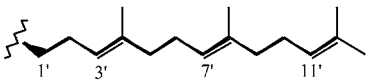




R_5	R_7	R_8	vitamer	code
CH_3	CH_3	CH_3	alpha-tocopherol	aT
CH_3	H	CH_3	beta-tocopherol	bT
H	CH_3	CH_3	gamma-tocopherol	gT
H	H	CH_3	delta-tocopherol	dT
H	H	H	tocol	To



R_5	R_7	R_8	vitamer	code
CH_3	CH_3	CH_3	alpha-tocotrienol	aT3
CH_3	H	CH_3	beta-tocotrienol	bT3
H	CH_3	CH_3	gamma-tocotrienol	gT3
H	H	CH_3	delta-tocotrienol	dT3

Figure 1. Structures of the eight naturally occurring E vitamers analyzed and their internal standard tocol. *Asymmetrical C atom (chiral center).

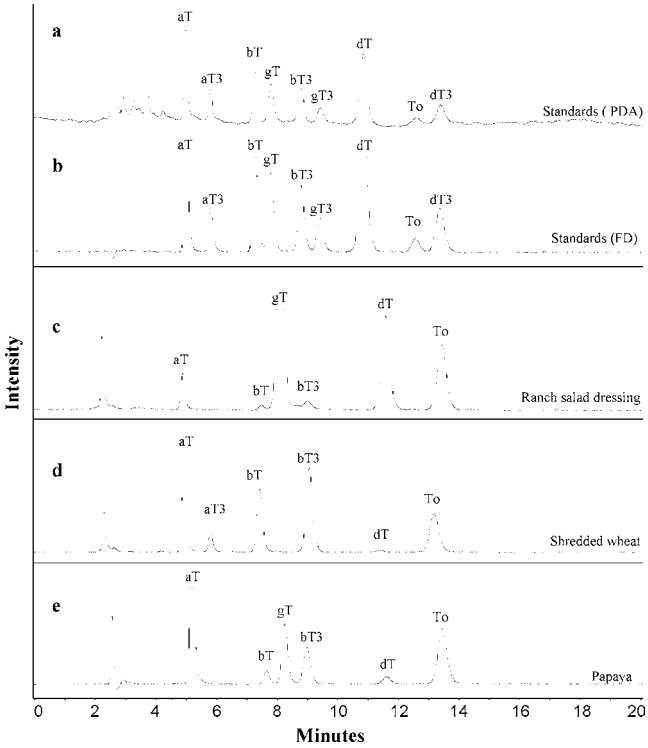


Figure 2. HPLC traces of standards monitored by absorbance at 295 nm (**a**) or by fluorescence (**b**), and extracts of ranch salad dressing high in soy oil (**c**), shredded wheat flour (**d**), and papaya fruit (**e**) monitored by fluorescence. Fluorescence traces were obtained using 296 nm for excitation and 336 nm for emission. FD, fluorescence detection; PDA, photo diode array; for other abbreviations, see **Figure 1**.

to the expression of vitamin E activity as αT equivalents. However, a recent report from the Institute of Medicine (IOM) (*2*) recommended that only αT be credited with vitamin E activity and that the use of αT equivalents be discontinued. Furthermore, of the eight stereoisomers that are included in synthetic αT, used for fortification and supplements, only those with an *R* conformation at C-2 (*RRR*, *RRS*, *RSR*, and *RSS*) are believed to have vitamin E bioactivity, and no bioactivity at all is attributed to the *S* conformers at C-2 (*4*).

While αT is the most active E vitamer in the traditional rodent fertility test, the most efficient at trapping reactive oxygen species (ROS), and the predominant E vitamer in the circulation, many health benefits also derive from the other forms of vitamin E (*4*, *5*). For example, γT and not αT scavenges reactive nitrogen oxide species (RNOS) to produce 5-nitro γT from nitrogen dioxide (*6*, *7*) or from the highly reactive peroxynitrite radicals generated in vivo from phagocytes during inflammation (*8*, *9*). γT but not αT acts as an antiinflammatory agent by inhibiting cyclooxygenase-catalyzed prostaglandin E2 formation (*10*, *11*), inhibits protein kinase C activity, aids in cell signaling (*2*), and is metabolized to a natriuretic factor (*12*). γT and particularly δT were found to have even higher hypocholesteremic properties than αT3 (*13*). T3 and δT but not other E vitamers were reported to kill breast cancer cell lines through apoptosis (*14*, *15*) and to reduce tumors in animals (*16*). γT exposure through the diet was recently correlated with lower cancer risk (*17*−*22*). Because of this very specific pharmacodynamic profile of each E vitamer but also because of their very different metabolic and pharmacokinetic patterns, food concentrations of individual Ts or T3s need to be known.

The total and relative amounts of T and T3 vary greatly in foods. Dietary E vitamers are found predominantly in nuts, seeds, grains (particularly their oils), and green leafy vegetables (mainly αT) but are lower in fruits (*23*). γT is the most prevalent E vitamer in vegetable oils from corn, soybean, or sesame and represents the major E vitamer (about 70%) in the U.S. diet (*10*). Wheat germ and rice bran are rich sources of βT3, while coconuts contain αT3 and represent the only γT3-containing fruit (*24*). Palm oil is unique due its high content of δT3 (*25*). Food composition and levels are influenced by processing, growing, and environmental conditions, i.e., higher in shaded than sunny locations and variable depending on season of harvest, storage, and food preparation (*25*). E vitamers remain stable in the absence of oxygen, such as in the anaerobic treatment used in canning processes (*26*), but up to 65% can be lost through the chemical treatment of foods (*25*). Boiled foods have lower E vitamer contents than fried foods because the latter retain vitamer-rich frying oils, which explains the high vitamin E content of french fries and potato chips (*25*).

Saponification prior to food extraction, although used traditionally (*27*−*29*), has recently been discouraged (*30*) because acylated E vitamers escape analysis and analyte degradation can occur during these alkaline hydrolysis conditions even in the presence of pyrogallol or other preservatives (*31*−*34*). E vitamer measurements are preferably carried out by normal-phase high-pressure liquid chromatography (NP-HPLC) with fluorescence detection (FD) after extraction with a highly lipophilic solvent

such as hexane (*32*). As compared to other techniques, this methodology offers high speed, selective separation by removing potentially interfering lipids in the early part of the chromatogram before the analytes are eluted, and sensitive and analyte specific monitoring using excitation and emission wavelengths of 290−298 and 326−330 nm, respectively (*27*, *30*, *32*). Previous reports on E vitamers in foods often do not include all eight vitamers in the analysis (*35*) or apply nonoptimal analytical techniques, i.e., no internal standards, no authentic standards for calibration, systems leading to coeluting analytes, or use of insensitive detection modes (*32*). The U.S. Department of Agriculture National Nutrient Database (USDANND, http://www.nal.usda.gov/fnic/foodcomp/search) is a widely used source of information on the vitamin E content of foods, but complete values are generally available only for αT. The current version (Standard Reference 18) includes other T values for only a few items, and T3 levels are not listed at all. A recent report on E vitamers in foods includes complete analyses for fruits and vegetables but not for other foods (*35*).

In order to extend this information to all E vitamers and to gain knowledge about a variety of local foods, we analyzed all naturally occurring Ts and T3s in 127 commonly consumed foods in Hawaii by direct extraction followed by sensitive and very selective NP-HPLC with FD and photodiode array (PDA) monitoring. In addition, acylated T in fortified cereals was measured by reversed-phase (RP) HPLC.

MATERIALS AND METHODS

Apparatus. HPLC analyses were carried out on a quaternary solvent delivery liquid chromatography system with FD (model FD100, GTI/SpectroVision, Concord, MA) and multiple channel PDA monitoring (model Surveyor, Thermo, San Jose, CA). Absorbance readings were obtained from a model BioSpec1601 spectrophotometer (Shimadzu, Kyoto, Japan). Vortexing was carried out on a Genie 2 model (Fisher Scientific, Santa Clara, CA). Lyophilization was performed using a model Freezermobile (The Virtis Co., Inc., Gardiner, NY). Centrifugation was performed with a model Alegra 21R centrifuge (Beckman Coulter, Fullerton, CA).

All solvents used for HPLC and absorbance readings were analytical grade or HPLC grade from Fisher Scientific (Fair Lawn, NJ). Butylated hydroxytoluene (BHT) and all other chemicals were purchased from Sigma Chemicals Co. (St. Louis, MO). αT3, βT3, γT3, and δT3 were obtained from EMD Bioscience Inc. (La Jolla, CA) (toco tris) or Davos Life Science PTE, Ltd. (Singapore). αT, α-tocopheryl acetate (αTac), γT, and δT were purchased from Sigma Chemicals Co. Tocol (To) and βT were a gift from DSM, formerly Hoffmann-LaRoche (Basel, Switzerland), and Dr. Robert Cooney, Cancer Research Center of Hawaii, respectively. All E vitamers were >96% pure according to HPLC analysis.

Food Collection and Preparation. Commonly consumed foods in Hawaii with insufficient or no information on E vitamer content were collected from local markets. All foods were treated as if intended for consumption to mimic typical household conditions. For frequently consumed foods, more than one item was purchased within a 2−3 day period from different markets (see **Table 1**). Foods were stored in a refrigerator or freezer for a few days depending on typical local customs, and then, fresh vegetables (broccoli, dandelion greens, spinach, beet greens, turnip greens, and collard greens) were boiled 5−10 min according to typical local methods prior to work up in the laboratory. Upon arrival in the laboratory, food materials were cut into pieces of 1−2 cm and frozen in sealed nitrogen-flushed plastic bags at −20 °C for 8−24 h. Whenever possible, we freeze-dried foods to preserve the labile analytes and to rupture cell compartments; this rupturing by lyophilization resulted in better extraction efficiency as compared to extraction of fresh materials (*36*−*38*). The lyophilized foods were packed into sealed nitrogen-flushed plastic bags and then analyzed, or the plastic bags were stored in a desiccator until extracted.

E Vitamer Analysis. A representative sample of each food item was homogenized with a tissue grinder or a mortar and pestle. To was added as an internal standard to 1.00 g of pulverized aliquots of the lyophilized material representing the entire food, and the mixture was extracted three times with 25 mL of hexane containing 20 mg/L BHT. After centrifugation at 4 °C for 5 min, the clear hexane phases were mixed and concentrated under reduced pressure to 1 mL. A 20 μL amount of that extract was injected directly into the HPLC system or after dilution with hexane if too concentrated. Oils were weighed, To was added as an internal standard, and hexane containing 20 mg/L BHT was added to give solutions with a final vitamin E concentration expected to be within the measurable range.

The NP-HPLC system consisted of a Spherex 5 OH analytical and guard column (diol, 250 mm × 4.6 mm i.d., 5 μm and 30 mm × 4.6 mm i.d., 5 μm; Phenomenex, Torrance, CA) and a mobile phase consisting of hexane:dioxane = 95:5 (v/v) containing 0.025% BHT (250 mg/L), which was kept at a flow rate of 1.3 mL/min. A RP-HPLC system was used to analyze acylated E vitamers and consisted of a Spherex C18 analytical and guard column (150 mm × 3.2 mm i.d., 3 μm and 4 mm × 3 mm i.d.; 10 μm; Phenomenex) and a mobile phase consisting of MeOH/CH_2Cl_2/MeCN (65:25:10; v:v:v) with 0.25 g/L BHT and 2 mL/L bis-tris-propane; elution was performed at a flow rate of 0.3 mL/min (*39*). PDA detection at 296 nm was followed online by FD using 296 nm for excitation and 336 nm for emission.

Peak areas were used for quantitation after adjusting for internal standard recovery and water content in order to express final values in mg/kg as consumed. External authentic standards were used for calibration using the following specific absorbance values in ethanol for concentration determinations (λ_{nm}; E-1%): αT (292; 75.8), αTac (285; 44.0), βT (296; 89.4), γT (298; 91.4), δT (298; 87.3), αT3 (292.5; 91), βT3 (294; 87.3), γT3 (296; 90.5), and δT3 (297; 88.1) (*40*). Calibration curves were extremely linear for all vitamers in the range 0.1−5.0 μg/mL using FD (r^2 > 0.992) and 1−80 μg/mL (r^2 > 0.988). The limit of quantitation using FD was 0.1 μg/mL (ca. 10-fold higher using PDA) for all analytes, which translated into 5.0 mg/kg in oils, 0.5 mg/kg in oily foods, and 0.1 mg/kg in freeze-dried foods (ca. 0.01 mg/kg fresh food) due to the differential food amount applied in the assay; food amounts for extraction were adjusted to the expected E vitamer concentration. Validation was performed by analyzing various brands of soy oil (*30*), which resulted in values in good agreement with those reported previously (*23*−*25*); repeated analyses of soy oil as an external standard during this study afforded coefficients of variation for all E vitamers at ≤8, ≤10, ≤11, ≤17, and ≤37% for concentrations >246, 246−67, 66−27, 27−8, and <8 mg/kg, respectively. Pearson's correlations were calculated using Excel:Mac 2001 software (Microsoft Corp., Redmond, WA).

RESULTS

A NP-HPLC system was applied that baseline-separated all eight analytes and also the internal standard To (**Figure 2**). Monitoring by fluorescence resulted in very sensitive measurement of all compounds of interest without interference (detection limit of freeze-dried samples at 0.01 mg/kg fresh weight). Quantitation could be performed by PDA monitoring if levels exceeded the dynamic range of FD. PDA monitoring was less sensitive but had a highly linear calibration curve up to very high concentrations (20 mg/mL; r^2 > 0.98). Acylated analytes other than αTac were not observed in any of the samples using RP-HPLC/PDA.

Table 1 provides average levels and ranges of Ts and T3s in the 79 food items analyzed (some in replicates, data not shown). The sum of all E vitamers (total E vitamers, TEV) in the analyzed foods ranged from 0.6 to 827.7 mg/kg, T ranged from nondetectable to 541.8 mg/kg, and T3 ranged from nondetectable to 432.2 mg/kg.

In bread items, we found individual Ts and T3s at levels up to 8 mg/kg, the lowest being δT and the highest being βT3, with TEV levels ranging from 20 to 22 mg/kg. The cereal group

Table 1. Mean and Range of E Vitamer Levels in Foods Analyzed[a]

food item mean [*n*] (range)	mg/kg (wet weight)								
	αT	βT	γT	δT	αT3	βT3	γT3	δT3	TEV
bread									
wheat [4]	2.3 ± 1.0 (1.4–3.2)	2.6 ± 0.7 (2.1–3.5)	5.9 ± 5.9 (2.4–14.7)	2.5 ± 3.3 (0.4–7.3)	0.4 ± 0.2 (0.2–0.6)	7.9 ± 1.2 (6.6–9.2)	0.2 ± 0.1 (<0.01–0.2)	<0.01 ±<0.01 (<0.01–0.1)	21.6 ± 11.3 (13.1–38.9)
white [4]	2.7 ± 1.2 (1.6–4.3)	1.1 ± 0.2 (0.9–1.3)	7.1 ± 3.0 (4.5–11.2)	2.3 ± 2.2 (0.5–4.8)	0.2 ± 0.2 (<0.01–1.5)	6.8 ± 3.2 (4.0–9.8)	0.1 ± 0.1 (<0.01–0.1)	<0.01 ± 0.1 (<0.01–0.10)	20.3 ± 2.2 (11.4–32.2)
cereals									
cereals [13][b]	7.7 ± 4.2 (1.1–13.2)	1.8 ± 1.7 (<0.01–6.0)	1.3 ± 2.6 (<0.01–9.7)	0.7 ± 1.1 (0.1–3.9)	1.6 ± 1.4 (0.1–5.4)	9.2 ± 11.9 (0.01–43.2)	1.0 ± 1.3 (<0.01–5.1)	0.1 ± 0.1 (<0.01–0.3)	23.4 ± 13.5 (3.9–56.2)
cookies									
chocolate chip cookies [5]	18.7 ± 11 (10.8–36.8)	3.0 ± 1.3 (1.9–5.1)	70.5 ± 35.6 (46.6–130.1)	30.3 ± 14.8 (17.6–53.9)	6.7 ± 14.5 (<0.01–32.7)	8.2 ± 4.6 (3.9–15.3)	1.0 ± 0.9 (<0.01–1.9)	0.1 ± 0.2 (<0.01–0.4)	138.4 ± 61.2 (91.2–242.6)
fig bars [2]	8.4 ± 3.5 (6.0–10.9)	1.7 ± 0.4 (1.4–2.0)	29.6 ± 3.1 (27.4–31.9)	9.6 ± 1.7 (8.4–10.8)	0.6 ± 0.3 (0.3–0.8)	3.5 ± 3.2 (1.2–5.7)	0.6 ± 0.2 (0.4–0.7)	0.1 ±<0.0 (0.1–0.1)	54 ± 12.4 (45.3–62.8)
fish									
raw Ahi (block) [3]	6.4 ± 2.4 (5.0–9.2)	<0.01 ± <0.0 (<0.01–<0.01)	0.1 ± <0.01 (<0.01–0.10)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	6.5 ± 2.4 (5.0–9.3)
raw Ahi (filet)	1.6	<0.01	<0.01	<0.01	<0.01	<0.01	0.3	<0.01	2.0
raw Atlantic Salmon (filet) [3]	37.7 ± 13.3 (23.5–49.9)	<0.01 (<0.01–<0.01)	0.6 ± 0.1 (0.5–0.7)	<0.01 (<0.01–<0.01)	0.1 ± 0.1 (<0.01–0.2)	0.2 ± 0.3 (<0.01–0.5)	0.5 ± 0.6 (<0.01–1.1)	<0.01 (<0.01–<0.01)	39.1 ± 12.8 (25.3–50.5)
raw Atlantic Salmon (steak)	32.4	<0.01	0.2	<0.01	0.1	<0.01	<0.01	<0.01	32.7
fruit									
canned pumpkin [2]	7.5 (5.1–9.8)	0.4 (0.3–0.5)	2.7 (2.5–2.9)	0.4 (0.4–0.4)	<0.01 (<0.01–<0.01)	1.5 (1.2–2.2)	0.2 (0.1–0.2)	<0.01 (<0.01–<0.01)	12.9 (9.7–16.1)
jack fruit	10.4	<0.01	<0.01	<0.01	0.1	0.2	0.1	<0.01	10.7
mango [2]	7.9 ± 0.9 (7.2–8.6)	0.1 ± <0.01 (0.1–0.1)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	0.1 ± 0.1 (<0.01–0.1)	0.2 ± <0.01 (0.2–0.2)	<0.01 ± <0.01 (<0.01–<0.01)	<0.01 ± <0.01 (<0.01–<0.01)	8.4 ± 1.0 (7.7–9.1)
papaya (green)	0.7	0.1	0.6	0.1	<0.01	0.3	<0.01	<0.01	1.8
papaya (red) [2]	1.1 ± 0.4 (0.8–1.3)	0.1 ± <0.01 (0.1–0.1)	0.2 ± 0.1 (0.2–0.3)	0.1 ± <0.01 (<0.01–0.1)	<0.01 ± <0.01 (<0.01–<0.01)	0.3 ± <0.01 (0.3–0.3)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	1.8 ± 0.6 (1.4–2.2)
papaya (yellow) [2]	6.8 ± 8 (1.2–12.5)	0.6 ± 0.7 (0.1–1.0)	1.4 ± 1.3 (0.5–2.3)	0.4 ± 0.4 (0.1–0.8)	0.1 ± 0.1 (<0.01–0.2)	2.3 ± 2.6 (0.5–4.2)	0.2 ± 0.1 (0.1–0.2)	<0.01 (<0.01–<0.01)	11.8 ± 13.2 (2.5–21.2)
margarine									
margarine, 60% fat [2]	33.7 ± 6.7 (28.9–38.4)	4.7 ± <0.5 (4.7–4.7)	214.9 ± 35.4 (189.9–240.0)	88.5 ± 23.2 (72.1–104.9)	<0.5 ± <0.5 (<0.5–<0.5)	16.1 ± 1.0 (15.4–16.8)	<0.5 (<0.5–<0.5)	1.2 ± 0.2 (1.0–1.3)	359.1 ± 66.1 (312.4–405.8)
margarine, 70% fat	63.4	5.8	285.5	82.2	<0.5	19.0	<0.5	0.7	456.6
mayonnaise									
fat-free mayonnaise	0.7	<0.5	1.9	1.5	<0.5	<0.5	0.8	<0.	4.9
meat									
Manapua (char siu)	1.2	0.4	0.9	1.4	0.1	1.5	0.2	6.1	11.7
Manapua (pork)	2.8	0.3	8.6	3.2	0.1	0.6	0.9	1.0	17.4
nuts									
boiled peanuts	36.9	1.2	25.0	7.2	<0.5	3.2	1.1	<0.5	74.6
dry-roasted macadamia nuts [2]	0.8 ± 1.1 (<0.5–1.5)	0.3 ± 0.2 (0.2–0.5)	0.3 ± 0.1 (0.2–1.4)	<0.5 (<0.5–<0.5)	18.0 ± 6.9 (13.1–22.8)	0.7 ± 0.2 (0.6–0.8)	1.7 ± 1.1 (1.0–2.5)	<0.5 (<0.5–<0.5)	21.8 ± 9.1 (15.4–28.2)
dry-roasted peanuts [2]	69.1 ± 0.8 (68.6–69.6)	25.6 ± 2.5 (23.8–27.3)	41.7 ± 9.3 (35.1–48.3)	12.9 ± 2.9 (10.9–14.9)	<0.5 (<0.5–<0.5)	10.0 ± 0.9 (9.4–10.7)	1.4 ± 2.0 (<0.5–2.9)	0.2 ± 0.3 (<0.5–0.4)	161.0 ± 4.1 (158.0–163.9)
honey-roasted almonds	185.3	2.1	4.8	0.9	2.4	4.1	1.3	<0.5	200.9
mixed nuts, oil-roasted, with peanuts [2]	69.6 ± 6.6 (64.9–74.3)	8.4 ± 2.8 (6.4–10.4)	47.2 ± 1.3 (46.3–48.1)	6.1 ± 2.3 (4.5–7.7)	1.0 ± 0.3 (0.8–1.2)	6.0 ± 0.5 (5.7–6.3)	2.0 ± 0.8 (1.4–2.6)	<0.5 (<0.5–<0.5)	140.3 ± 14.5 (130.0–150.6)
oil									
corn oil [2]	222.1 ± 66.2 (175.3–268.9)	19.0 ± 12.0 (10.4–27.5)	436.4 ± 135.0 (340.9–531.9)	69.4 ± 25.2 (51.6–87.2)	9.8 ± 13.9 (<0.5–19.7)	40.0 ± 5.5 (36.1–43.9)	31.0 ± 3.9 (28.2–33.7)	<0.5 ± <0.5 (<0.5–<0.5)	827.7 ± 187.4 (695.2–960.2)
safflower oil [4]*	391.2 ± 190.9 (122.9–575.2)	17.1 ± 10.1 (7.0–31.1)	14.1 ± 7.3 (3.4–19.5)	15.3 ± 11.6 (<0.5–25.5)	1.3 ± 2.7 (<0.5–5.4)	10.2 ± 12.4 (<0.5–25.7)	10.6 ± 7.4 (3.8–18.20	37.0 ± 59.6 (<0.5–125.0)	497.0 ± 276.8 (142.9–816.4)
sesame oil [2]	65.0 ± 2.4 (48.2–82.4)	78.0 ± 11.0 (50.3–156.7)	382.0 ± 0.1 (382.4–383.3)	<0.5 (<0.5–<0.5)	<0.5 (<0.5–<0.5)	<0.5 (<0.5–<0.5)	<0.5 (<0.5–<0.5)	<0.5 (<0.5–<0.5)	525.0 ± 8.5 (465.7–585.4)
sunflower oil	541.8	20.8	41.8	15.3	<0.5	19.9	<0.5	<0.5	639.6
pasta									
fried noodles (chow mein) [2]	9.2 ± 9.2 (2.7–15.8)	1.4 ± 0.1 (1.3–1.4)	20.1 ± 20.4 (5.6–34.5)	17.9 ± 11.1 (10.1–25.8)	1.2 ± 0.8 (0.6–1.8)	2.8 ± 0.7 (2.3–3.3)	0.7 ± 0.9 (<0.5–1.3)	<0.5 (<0.5–<0.5)	53.3 ± 43.2 (22.7–83.8)
fried noodles (top ramen) [4]	16.3 ± 4.8 (9.4–20.4)	1.7 ± 1.2 (0.7–3.3)	4.0 ± 3.2 (0.3–7.8)	5.5 ± 5.4 (<0.5–10.8)	13.2 ± 5.7 (6.5–18.9)	8.1 ± 3.7 (4.4–12.9)	36.4 ± 21.1 (13.4–60.3)	5.0 ± 0.9 (3.7–5.8)	90.3 ± 29.6 (53.8–122.9)
spaghetti/ marinara pasta sauce [4]	10.9 ± 3.1 (8.9–15.4)	0.5 ± 0.1 (0.4–0.7)	6.4 ± 3.1 (2.6–9.3)	1.9 ± 1.3 (0.5–3.5)	<0.01 (<0.01–<0.01)	3.8 ± 2.2 (2.1–7.1)	<0.01 ± 0.1 (<0.01–0.2)	0.4 ± 0.7 (<0.01–1.5)	24.0 ± 7.9 (16.4–34.4)

Table 1. Continued

food item mean [*n*] (range)	mg/kg (wet weight) αT	βT	γT	δT	αT3	βT3	γT3	δT3	TEV
salad dressing									
blue cheese salad dressing [2]	16.3 ± 8.1 (10.6–22.0)	1.2 ± 1.7 (<0.5–2.3)	64 ± 22.1 (48.4–79.7)	36.6 ± 7.3 (31.4–41.8)	<0.5 (<0.5–<0.5)	2.4 ± 1.1 (1.6–3.1)	0.6 ± 0.8 (<0.5–1.2)	<0.5 (<0.5–<0.5)	121.0 ± 33.9 (97.0–145.0)
French salad dressing [2]	19.3 ± 26.0 (0.8–37.7)	1.4 ± 1.7 (0.2–2.6)	68.2 ± 92.0 (3.2–133.3)	18.4 ± 23.3 (1.9–34.8)	<0.5 (<0.5–<0.5)	3.2 ± 4.6 (<0.5–6.5)	0.8 ± 1.1 (<0.5–1.5)	0.2 ± 0.1 (0.2–0.3)	111.5 ± 148.7 (6.4–216.7)
Italian salad dressing [2]	40.7 ± 8.8 (34.5–47.0)	4.4 ± 3.4 (2.0–6.7)	167.8 ± 127.8 (77.4–258.1)	62.6 ± 39.5 (34.7–90.6)	0.9 ± 0.5 (0.6–1.3)	12.5 ± 0.3 (12.3–12.7)	1.4 ± 2.0 (<0.5–2.8)	0.3 ± 0.5 (<0.5–0.7)	290.6 ± 182.7 (161.4–419.8)
Oriental salad dressing [2]	22.1 ± 25.1 (4.3–39.8)	2.6 ± 2.2 (1.0–4.1)	141.4 ± 169.1 (21.8–260.9)	2.8 ± 3.9 (<0.5–5.6)	3.8 ± 5.3 (<0.5–7.6)	3.5 ± 1.7 (2.3–4.6)	<0.5 (<0.5–<0.5)	<0.5 (<0.5–<0.5)	176.0 ± 199.5 (35.0–317.1)
ranch salad dressing [2]	2.4 ± 0.7 (1.9–2.9)	0.3 ± 0.1 (0.2–0.3)	10.1 ± 0.4 (9.9–10.4)	6.2 ± 1.3 (5.3–7.1)	<0.5 (<0.5–<0.5)	0.4 ± 0.6 (<0.5–0.9)	0.2 ± 0.2 (<0.5–0.3)	<0.5 ± 0.5 (<0.5–0.1)	19.6 ± 20.5 (18.3–21.0)
sesame and miso salad dressing	57.9	5.5	<0.5	15.5	2.1	12.5	<0.5	<0.5	93.5
sesame ginger salad dressing [2]	9.0 ± 10.6 (1.5–16.5)	0.1 ± 0.1 (<0.5–0.2)	40.1 ± 56.1 (0.4–79.8)	2.4 ± 3.4 (<0.5–4.8)	11.8 ± 15.6 (0.8–22.80)	0.4 ± 0.6 (<0.5–0.8)	1.2 ± 1.8 (<0.5–2.5)	15.3 ± 21.6 (<0.5–30.6)	80.3 ± 73.7 (28.2–132.4)
thousand island dressing [4]	16.3 ± 12.0 (6.1–32.8)	1.2 ± 0.8 (<0.5–1.8)	35.9 ± 20.2 (20.2–64.8)	16.7 ± 12.5 (2.2–32.0)	0.5 ± 1.0 (<0.5–2.1)	7.3 ± 11.1 (1.3–23.9)	0.3 ± 0.7 (<0.5–1.4)	0.8 ± 1.2 (<0.5–2.7)	79.0 ± 36.0 (49.0–123.2)
snack									
frozen, ready-to-bake pie crust, baked	15.0	3.8	100.7	40.0	<0.01	5.7	0.8	<0.01	166.0
graham cracker pie crust	12.6	3.1	65.0	38.7	0.7	14.1	<0.01	0.3	134.5
microwave popcorn [2]	25.4 ± 2.4 (23.7–27.1)	2.6 ± 0.2 (2.5–2.8)	127.7 ± 5.4 (123.9–131.6)	49.9 ± 4.9 (46.4–53.3)	1.8 ± 0.4 (1.5–2.1)	7.1 ± 0.5 (6.8–7.4)	3.8 ± 0.6 (3.4–4.3)	1.5 ± 1.1 (0.8–2.3)	219.8 ± 11.3 (211.9–227.8)
potato chips, plain, salted [4]	37.5 ± 14.3 (24.2–55.9)	1.2 ± 0.9 (<0.5–1.9)	50.7 ± 64.8 (1.7–145.4)	0.8 ± 1.0 (<0.5–2.1)	0.2 ± 0.5 (<0.5–0.9)	9.0 ± 7.6 (<0.5–18.4)	1.3 ± 1.7 (<0.5–3.5)	0.6 ± 0.9 (<0.5–1.9)	101.3 ± 64.2 (57.5–194.2)
potato chips, plain, salted, hydrogenated oils [2]	76.5 ± 25.3 (58.6–94.4)	1.0 ± 0.6 (0.6–1.4)	53.6 ± 39.8 (25.5–81.7)	2.7 ± 2.3 (1.1–4.4)	0.8 ± 0.2 (0.7–1.0)	12.9 ± 9.8 (6.0–19.8)	4.4 ± 3.9 (1.6–7.1)	<0.5 (<0.5–<0.5)	152.0 ± 81.4 (94.4–209.5)
spice									
raw tamarind	<0.01	0.2	0.1	<0.01	0.3	<0.01	<0.01	<0.01	0.6
vegetable									
beet greens[c]	13.9	0.1	0.5	<0.01	0.9	2.6	0.7	<0.01	18.7
canned tomato soup [2]	2.1 ± 0.2 (1.9–2.2)	0.3 ± <0.01 (0.2–0.3)	0.1 ± 0.1 (0.1–0.2)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	1.1 ± <0.01 (1.1–1.1)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	3.5 ± 0.3 (3.3–3.7)
canned, whole leaf spinach [2]	20.9 ± 3.7 (18.3–23.5)	0.4 ± 0.2 (0.3–0.5)	2.5 ± 2.0 (1.0–3.9)	0.3 ± 0.2 (0.2–0.4)	0.6 ± 0.2 (0.5–0.7)	3.5 ± 0.9 (2.8–4.1)	0.3 ± 0.2 (0.2–0.4)	5.1 ± 7.2 (<0.01–10.2)	33.5 ± 0.3 (33.3–33.7)
carrot juice	1.0	0.1	0.1	<0.01	<0.01	0.4	0.3	0.1	1.9
chili, hot pepper paste	12.1	1.4	2.5	0.2	0.3	2.7	<0.01	0.2	19.4
Chinese cabbage [2]	2.6 ± 0.8 (2.0–3.1)	<0.01 (<0.01–<0.01)	<0.01 ±<0.01 (<0.01–0.1)	0.1 ± 0.2 (<0.01–0.3)	<0.01 (<0.01–<0.01)	0.3 ± 0.1 (0.2–0.4)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	3.0 ± 1.2 (2.2–3.9)
dandelion greens[b]	7.0	0.1	3.1	<0.01	0.1	1.1	<0.01	0.1	11.6
fresh spinach [2][b]	11.4 ± 2.0 (10.0–12.8)	0.1 ± <0.01 (<0.01–0.1)	0.6 ± 0.3 (0.4–0.8)	<0.01 ± 0.1 (<0.01–0.1)	0.1 ± 0.2 (<0.01–0.2)	1.4 ± 0.5 (1.1–1.8)	0.1 ± 0.1 (<0.01–0.2)	0.1 ± 0.21 (<0.01–0.3)	13.8 ± 1.5 (12.8–14.9)
frozen broccoli[c]	4.8	0.1	0.5	<0.01	<0.01	0.1	<0.01	<0.01	5.4
frozen collards[c]	18.5	0.3	<0.01	<0.01	0.1	1.5	0.3	<0.01	20.7
frozen turnip greens[c]	19.2	0.3	0.1	<0.01	<0.01	2.1	<0.01	<0.01	21.7
red bell pepper [2]	20.8 ± 14.1 (10.8–30.8)	0.5 ± 0.3 (0.3–0.7)	0.1 ± 0.1 (<0.01–0.1)	<0.01 01	<0.01 (<0.01–<0.01)	1.1 ± 0.3 (0.8–1.3)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	22.5 ± 14.8 (12.1–32.9)
sweet potatoes	2.1	0.1	0.1	<0.01	<0.01	<0.01	0.1	<0.01	2.3
taro root [2]	4.1 ± 14.1 (3.4–4.7)	<0.01 ± 0.3 (<0.01–0.1)	<0.01 ± 0.1 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	1.2 ± 0.3 (1.0–1.3)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	5.4 ± 14.8 (4.8–5.9)
tomato	5.5	0.1	1.9	<0.01	<0.01	1.4	<0.01	<0.01	9.0
turnip greens[c]	19.6	0.1	0.5	<0.01	0.6	0.6	0.7	<0.01	22.1
other									
fried tofu	18.1	1.4	29.7	11.2	0.1	2.4	0.4	0.1	63.5
poi [2]	2.0 ± 0.4 (1.7–2.3)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	0.2 ± 0.1 (0.2–0.3)	<0.01 (<0.01–<0.01)	<0.01 (<0.01–<0.01)	2.3 ± 0.3 (2.0–2.5)

[a] The range describes the lowest and highest values found in different foods; *n* = number of different foods; ±, standard deviation. [b] Rac αTac present due to fortification in Complete Wheat Bran Flakes (813 mg/kg), Product 19 (971 mg/kg), Raisin Nut Bran (ND), Whole Grain Total (239 mg/kg), Total Corn Flakes (594 mg/kg), Total Raisin Bran (470 mg/kg), and Special K (289 mg/kg). The rac αTac content was multiplied with 0.45 to convert to natural αT (*2*). [c] Boiled for 5–10 min as needed for consumption prior to analysis.

had TEV levels ranging from 4 to 56 mg/kg, with βT3 as the most predominant vitamer with levels up to 43 followed by αT with levels ranging from 1 to 13 mg/kg. In the cookie group, mean TEV levels ranged from 54 to 138 mg/kg; γT was the

Table 2. Foods from Hawaii Ranked According to αT and TEV Levels

rank	food name	αT (mg/kg)	αT/ TEV	RDA (g)[b]	food name	TEV (mg/kg)	αT/ TEV
1	sunflower oil	542	0.85	28	corn oil[a]	828	0.27
2	safflower oil[a]	391	0.79	38	sunflower oil	640	0.85
3	cereals[a,c]	261	0.94	58	sesame oil[a]	525	0.12
4	corn oil[a]	222	0.27	68	safflower oil[a]	497	0.79
5	honey-roasted almonds	185	0.92	81	margarine, 70% fat	457	0.14
6	potato chips[a]	77	0.50	196	margarine, 60% fat[a]	359	0.09
7	mixed nuts, oil-roasted[a]	70	0.50	216	Italian salad dressing[a]	291	0.14
8	dry-roasted peanuts[a]	69	0.43	217	cereals[a]	276	0.94
9	sesame oil[a]	65	0.12	231	microwave popcorn[a]	220	0.12
10	margarine, 70% fat	63	0.14	237	honey-roasted almonds	201	0.92
11	sesame and miso salad dressing	58	0.62	259	Oriental salad dressing[a]	176	0.13

[a] Mean levels for foods with multiple samples (see **Table 1** for details). [b] Grams of food needed to obtain the recommended dietary allowance of 15 mg αT/day (*2*). [c] Presence of αTac determined by RP-HPLC; we assumed rac αTac and used a multiplication factor of 0.45 to convert to natural αT (*2*).

most prevalent vitamer with mean levels ranging from 30 to 71 mg/kg. In the fish group, mean TEV levels ranged from 2 to 39 mg/kg; αT was the most prevalent vitamer with mean levels ranging from 2 to 38 mg/kg. The margarines had mean TEV levels ranging from 359 to 457 mg/kg; γT was the most prevalent vitamer with mean levels ranging from 215 to 286 mg/kg, while αT3 and γT3 were not detected. The nut group contained mean TEV levels ranging from 22 to 201 mg/kg; αT yielded 0.8−185 mg/kg, while δT3 yielded 0.0−0.2 mg/kg. The salad dressings contained mean TEVs ranging from 20 to 291 mg/kg with γT predominating, except mainly αT when soy oil was the main ingredient. The vegetables had mean TEV levels ranging from 2 to 152 mg/kg with mainly αT (1.0−77 mg/kg). The oil group had the highest T and T3 values (TEV 497−828 mg/kg). αT and γT were the most predominant E vitamers in the analyzed foods with mean levels reaching 542 and 436 mg/kg, respectively. Least abundant was αT3 followed by γT3, δT3, βT3, βT, and δT with mean levels up to 18.0 mg/kg, 36.4 mg/kg, 37.0 mg/kg, 40.0 mg/kg, 78.0 mg/kg, and 88.5 mg/kg, respectively. The fruit group had the lowest levels of TEV, with mean levels ranging from 1.8 mg/kg (papaya) to 12.9 mg/kg (canned pumpkin). αT was the main vitamer, ranging from 0.7 (green papaya) to 10.4 mg/kg (jackfruit).

RP-HPLC was applied for fortified foods because, contrary to NP-HPLC, αTac could be accurately quantified without coeluting interferences. We found αTac levels in Complete Wheat Bran Flakes (813 mg/kg), Product 19 (971 mg/kg), Whole Grain Total (239 mg/kg), Total Corn Flakes (594 mg/kg), Total Raisin Bran (470 mg/kg), and Special K (289 mg/kg) but not in Raisin Nut Bran (ND). Because fortifications are usually carried out with racemic (rac) αTac mixtures (*26*), we converted αTac into αT with vitamin E activity by multiplying with 0.45 (*2*). After this conversion, we found that, on average, the six fortified cereals that we analyzed contained a total of 261 mg/kg αT (**Table 2**), which represented 94% of all E vitamers in these cereals. The high levels were largely due to fortification because the mean natural αT content was only 7.7 mg/kg.

Table 2 shows the ranking of the analyzed foods according to αT levels, which is quite different from the ranking according to TEV concentrations. The αT/TEV ratio shows the contribution of αT to the total E vitamer concentration. In the 11 richest αT foods, the ratio ranged from 0.85 to 0.12. The amount of these foods needed to achieve the current recommended intake of 15 mg/day αT (*2*) ranged from 28 to 259 g.

Pearson's correlations (**Table 3**) between TEV (excluding fortifications by acylated E vitamers) and tocopherols were found to be very high in the analyzed foods, ranging between 0.60 (βT) and 0.87 (γT), whereas those between TEV and T3s were lower, ranging from 0.32 for δT3 to 0.65 for βT3 and <0.10 for αT3. Among the E vitamers, high correlations were found between the pairs γT/δT ($r = 0.75$), γT/βT ($r = 0.59$), γT/βT3 ($r = 0.56$), and δT/βT3 ($r = 0.54$). Correlations among all other E vitamers were small (**Table 3**). When compared to fat components as assessed from the food labels [total, saturated (SAT), monounsaturated (MU), and polyunsaturated (PU)], very high correlations with TEV were observed (total = 0.87, PU = 0.83, SAT = 0.69, and MU = 0.67). Among the individual E vitamers, Ts were highly correlated with the fat components ($r = 0.35-0.78$), particularly with αT and γT (αT/PU, $r = 0.78$; γT/total, $r = 0.74$; γT/MU, $r = 0.68$; and γT/SAT, $r = 0.67$). T3s were less well-correlated with the fat components, and αT3 showed the lowest correlation coefficients.

Table 3. Correlations among E Vitamers and between E Vitamers and Fat in the Foods Analyzed[a]

	TEV	αT	βT	γT	δT	αT3	βT3	γT3	δT3
TEV									
αT	0.68								
βT	0.60	0.40							
γT	0.87	0.25	0.59						
δT	0.73	0.17	0.17	0.75					
αT3	0.08	0.01	0.00	0.06	−0.01				
βT3	0.65	0.38	0.22	0.56	0.54	0.18			
γT3	0.51	0.26	0.16	0.43	0.42	0.30	0.40		
δT3	0.32	0.44	0.14	0.06	0.17	0.10	0.08	0.31	
fat									
total	0.87	0.62	0.60	0.74	0.58	0.16	0.50	0.37	0.31
SAT	0.69	0.39	0.50	0.67	0.49	0.18	0.38	0.33	0.14
MU	0.67	0.35	0.59	0.68	0.45	0.17	0.35	0.27	0.07
PU	0.83	0.78	0.51	0.58	0.50	−0.03	0.46	0.35	0.51

[a] Total = total fat; SAT = saturated fat; MU = monounsaturated fat; PU = polyunsaturated fat; other abbreviations, see **Table 1**.

DISCUSSION

The applied NP-HPLC system resulted in baseline separation of all eight analytes including the internal standard To. This is a major achievement because previous LC methods separated one or more analytes only partially or did not include an internal standard (*32*). The NP mode proved to be a major advantage relative to a RP system because the lipids were eluted from the column before the analytes (*41*). This resulted in a clean column for each consecutive injection, avoided the need to recondition the stationary phase due to accumulation of sample matrix (lipids), and led to fast turnaround times. The on-line combination of FD and PDA detection in our HPLC system was very

useful because the data obtained from the latter could be used for analysis of high E vitamer levels that exceeded the FD quantitation range. This avoided the need for reanalysis after sample dilution and accelerated the turnaround times. Saponification, although recommended by some reports (*27*), was not performed due to our earlier observation on degradation of E vitamers in that process despite the use of BHT or vitamin C as a preservative (*30*). This is in agreement with others (*30*−*32*) and also allowed us to quantitate acylated E vitamers such as αTac, which is used widely for fortification.

The richest sources of αT (≥58 mg/kg) among the analyzed foods were vegetable oils or foods rich in these oils (nuts, margarine, salad dressings, and potato chips) and fortified cereals (**Table 2**). As others have reported, E vitamers occur mainly in oils of seeds with Ts present mainly in nuts and vegetable oils and T3s occurring predominantly in monocotyledonic plants (cereals, rice, barley, and oats) (*27*, *42*). A daily intake of 15 mg of αT, as recommended for adults by the IOM (*2*), would be achieved by 28 g of sunflower oil, but larger amounts would be needed for foods that are poorer sources of αT (see **Table 2**). The ranking according to αT does not correlate well with the ranking according to TEV ($r < 0.1$ if the T richest foods in **Table 2** are considered), which highlights the difference in food distribution of the E vitamers. This is further underscored in the correlation matrix in **Table 3** with low correlation coefficients between the individual E vitamers, even between the predominant food E vitamers αT and γT ($r = 0.25$). Interestingly, γT was more associated with SATs and MUs, whereas αT was more correlated with PUs. This might be due to the biological function of these vitamers because αT is better at scavenging ROS and, therefore, the best protection against fat oxidation (*5*, *42*).

Our findings for αT were similar to those reported by the USDANND for some items (wheat bread, white bread, papaya, boiled peanuts, mixed nuts, oil roasted with peanuts, safflower oil, Italian salad dressing, and canned, whole leaf spinach, chewy Chips Ahoy, mango, dry-roasted peanuts, safflower oil, sunflower oil, fresh spinach, and turnip) but were more than 20% different for almost half of the analyzed foods. **Table 1** shows E vitamer values for 24 foods that were not included in the USDANND or other reports (for example, chewy chocolate chip cookies and reduced fat chocolate chip cookies, refrigerated chocolate chip cookie dough, vanilla wafers, raw ahi tuna, raw wild salmon, jack fruit, 70% margarine, manapua, honey-roasted almonds, top ramen, Oriental salad dressing, fresh carrot juice, chili hot pepper paste, and plain potato chips made with hydrogenated oils.). For most of the analyzed food items and almost all of the local foods, βT, γT, δT, and all T3 values are reported here for the first time. Also, the USDANND, even the newest SR18 version, often does not distinguish between different food varieties, which we did in our analyses, for example, for papaya (red, yellow, and green).

Six of the foods that we analyzed were also included in a recent E vitamer analysis of fruits and vegetables (*35*); similar results were obtained for tomatoes, sweet potatoes, and Chinese cabbage, but we found lower values for marinara sauce, fresh spinach, and frozen broccoli. This could be due to the natural variability of these foods or to differences in the storage or preparation methods.

When we analyzed the same food repeatedly, either from different brands or from markets, we found considerable variability (>50 mg/kg standard deviation) in cookies, corn and safflower oils, French, Italian, and sesame ginger salad dressings, and potato chips. Studies investigating dietary exposure should consider this variability and also variation in levels of other foods if consumed in large amounts; in the latter case, even relatively small standard deviations could contribute to marked variation in daily E vitamer intake.

Fortified foods contained αTac levels that were in good agreement with the amounts shown on the label except for Total Whole Grain and Total Corn Flakes, where we found significantly lower levels. Because we were unable to repeat these analyses, αTac results for these items should be considered unconfirmed. Cereal fortification led to very high αT levels (>97% of total αT) after conversion from the analyzed αTac concentration [using a factor of 0.45 (*2*)]. It raised fortified cereals to the third and eighth highest rank regarding αT and TEV concentrations, respectively, among all analyzed foods, superior to nuts and even some oils (**Table 2**). Only 58 g of fortified cereals is needed to meet the recommended intake of 15 mg/day αT, which is easy to achieve and, in contrast to most other αT-rich foods, is not associated with high energy intake.

Each of the E vitamers possesses a very specific pharmacokinetic and pharmacodynamic profile (*2*), and consequently, dietary exposure of each vitamer needs to be determined individually. T3s but not Ts were found to lower serum cholesterol levels in animals and humans (*43*, *44*). αT3 was better than αT in regards to trapping peroxyl radicals (*45*) or as general antioxidants (*46*). More recently, γT and particularly δT were found to have even higher hypocholesteremic properties than αT3 (*13*). T3 and δT but not other E vitamers were reported to kill breast cancer cell lines through apoptosis (*14*, *15*) and to reduce tumors in animals (*16*). γT but not αT reduced inflammation via cyclooxygenase inhibition in vitro and in vivo (*47*), lowered C-reactive protein (*48*), and was identified to be metabolized in humans to LLU-α, a specific natriuretic factor (*12*, *49*). γT but not αT plasma levels could be increased by sesame seeds (*30*, *50*) in animals and humans leading to enhanced vitamin E bioactivity (*50*). Clinical relevance for the activity of an individual E vitamer is evidenced by the elevated 5-nitro γT levels in plasma of subjects with coronary heart disease or with atherosclerotic plaques (*51*). General cancer protective activities by γT are inconsistent and depend on the site and stage of this disease (*52*), but very promising and consistent findings have been reported for γT and partly for αT regarding the prevention of prostate cancer in cell, animal, and, most importantly, also in prospective and intervention studies (*17*−*22*). In male smokers, a 32% reduction in prostate cancer incidence and a moderate reduction in colorectal cancer was demonstrated in response to daily 400 I.U. vitamin E given as rac αTac (*53*, *54*). However, this benefit by supplementation disappeared during postintervention follow-up (*55*). In contrast, participants with high circulating baseline concentrations of αT and γT from dietary exposure experienced a 51 and 43% lower prostate cancer risk, respectively (*21*). Most strikingly, in prospective studies, the highest quintile of serum γT experienced a 5-fold decreased prostate cancer risk vs the lowest quintile (*22*). While these and other epidemiologic data indicate increasingly a protective effect of dietary E vitamers against chronic disorders, intervention trials could generally not confirm these findings (reviewed in *56*−*60*). Most recent long-term studies in diseased or nondiseased populations confirm this (*61*, *62*). The HOPE trial with over 9500 cardiovascular disease (CVD) and diabetes mellitus patients taking daily 400 I.U. vitamin E as *RRR*-αTac showed no effects on major cardiovascular events or cancer even in the extended trial with a total of 7 years of intervention; contrary to expectations, heart failure even increased relative to the control (*61*). Similarly, healthy women

(ca. 20000) taking supplements of 600 I.U. of natural-source vitamin E every other day did not experience a reduction in heart disease, stroke, cancer, or all causes of death when followed for a mean of 10.1 years (*62*). Also, Alzheimer's disease risk was not affected by vitamin E supplements, but again, once consumed in the diet, a significant reduction in risk was observed (40 or 34% for every 5 mg of γT or αT, respectively) (*63*, *64*).

Dietary supplements contain predominantly αT as the sole E vitamer, mostly as acetate ester and in doses often over 20-fold higher than currently recommended (*1*). It is known that αT consumption can impair the bioavailability of other E vitamers (*65*) (reviewed in *66*). Therefore, other E vitamers, especially γT, might be lowered in the circulation upon high exposure to αT and thereby diminish desirable health effects. Such imbalances are avoided by dietary vitamin E exposure because foods contain a natural E vitamer mixture not just αT. For example, the U.S. population's highest dietary exposure among the E vitamers is γT due to soy oil intake (*67*). Exposure to the natural E vitamer mixture might be needed to achieve desired health effects. Therefore, we suggest that future research should consider all E vitamers individually, or at least the sum of all E vitamers (TEV), not only αT. TEV values based on weight (mg/kg) bear a small error ($\leq$6.5%) due to molecular weight differences of the individual E vitamers. This error can be avoided by using food concentrations based on molar units (mmol/kg), but these units are less commonly used in the nutrition field.

In addition, dietary E vitamer consumption is correlated with exposure to other known and unknown compounds that might act individually or in concert with E vitamers to produce (synergistic) benefits. Support for this hypothesis comes from the significant increase in circulating γT levels after sesame intake (*30*) or the cardioprotective effect from nut ingestion that approaches the effects achieved with the use of lipid-lowering medication (reviewed in *68*). In addition to the presence of all E vitamers, other macro- and micronutrients in nuts (or oils) such as lignans, arginine, folate, fiber, trace minerals, tannins, and/or polyphenols, alone or in combination with E vitamers, might account for these findings. This highlights the superior outcome of dietary over supplemental E vitamer exposure. Because of the specificity in bioactivities, each of the E vitamers should be considered as an individual agent in the evaluation of vitamin E effects; therefore, food composition tables need to carry all E vitamers. Science is continuously undergoing change, and evaluating intakes of only αT ignores the other important E vitamers. Inclusion of all E vitamers will avoid masking effects and misleading outcomes and will allow recalculations if a new grouping of individual E vitamers is desired. These new data will be used to update the food composition table used for dietary studies in Hawaii and, thus, will allow better estimates of E vitamer intakes. Ultimately, they will lead to a higher likelihood of discovering important associations in epidemiologic and other studies.

ABBREVIATIONS USED

λ_{nm}, wavelength in nanometer; rac, racemic; αT, α-tocopherol; αTac, α-tocopheryl acetate; αT3, α-tocotrienol; αTE, αT equivalent ($\alpha T \times 1.0 + \beta T \times 0.5 + \gamma T \times 0.1 + \delta T \times 0.03 + \alpha T3 \times 0.33$) (3); βT, β-tocopherol; βT3, β-tocotrienol; CVD, cardiovascular disease; δT, δ-tocopherol; δT3, δ-tocotrienol; FD, fluorescence detection; γT, γ-tocopherol; γT3, γ-tocotrienol; HPLC, high-pressure liquid chromatography; MS, mass spectrometry; MU, monounsaturated; NP, normal phase; PDA, photodiode array; PU, polyunsaturated fat; RP, reversed phase; RNOS, reactive nitrogen oxide species; ROS, reactive oxygen species; SAT, saturated fat; T, tocopherols; T3, tocotrienols; To, tocol; TEV, total E vitamers (sum of all T and all T3 but excluding acylated E vitamers); USDANND, U.S. Department of Agriculture National Nutrient Database (http://www.nal.usda.gov/fnic/foodcomp/search/).

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