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Original Article

Dioxin and furan contamination of deodorizer distillates and natural vitamin E supplements

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Abstract

Thirty-five samples of vitamin E supplement softgels, obtained from store shelves, were analyzed for 17 dioxin and furan congeners. Of these samples, 14 were identified as natural vitamin E, containing D-α-tocopherol, as well as lesser amounts of β-, α-, and δ-tocopherols, and the remaining 21 were labeled as synthetic vitamin E, containing a mixture of D- and L-α-tocopherol. The supplements were collected during the years of 2002 and 2004. The seven natural vitamin E supplements collected in 2002 were found to contain significant quantities of dioxins and furans, with an average total toxic equivalence (TEQ) of 0.79 pg/g, compared to 0.10 g/g for the 2004 natural vitamin E supplements. The 21 synthetic vitamin E supplements collected during the same time period showed little or no contamination, with an average TEQ of 0.057 pg/g. Eight samples of deodorizer distillate, from which natural vitamin E is derived, were also collected and analyzed. The distillates exhibit an overall congener pattern similar to that found in the natural vitamin E, but at a much higher average TEQ of 3.4 pg/g. This suggests the possibility of carryover of contamination to the vitamin E samples from the deodorizer distillate during the extraction process. The natural vitamin E supplements collected in 2004 have much lower levels of contamination, suggesting that improved extraction processes may be in use, effectively reducing contamination.
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Keywords: Vitamin E; Tocopherol; Deodorizer distillate; Bleaching clay; Dioxin; Furan; PCDD; PCDF

1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), commonly referred to as dioxins, include a group of 17 lipophilic compounds that are considered highly carcinogenic when the chlorines are in the 2,3,7 and 8 positions. Over 95% of human exposure to these compounds comes from dietary intake (Eljarrat et al., 2002). Cases illustrating the presence of these compounds in various foods and animal feeds have been published both domestically (Ferrario et al., 2002) and internationally (Jacobs et al., 2002; Wu et al., 2001; Aoki, 2001). Another group (Schecter and Li, 1997) investigated PCDD/PCDF levels in fast foods. The majority of exposure concerns have been predominantly from animal feeds, which ultimately appear in the human diet in such commodities as milk, eggs, poultry, beef, and fish. The European Union has established maximum allowable dioxin levels for various foodstuffs for human consumption and for components of animal feeds, and has recently revised these values (The Commission of the European Communities, 2006). The United States is in the process of acquiring data by sampling foodstuff, vitamins, feeds, and feed ingredients. The US Food and Drug Administration (FDA) has established a monitoring program to obtain baseline data for dioxins and furans in these consumables.

Vitamin supplements collected in the survey include those containing vitamin E. Vitamin E supplements are available as natural vitamin E, containing mixed tocopherols, and as synthetic vitamin E, which is a mixture of D- and L-alpha tocopherol. Studies have indicated that the natural form has a higher degree of biopotency; the currently accepted ratio of biopotency of natural to synthetic vitamin E is 1.36 (Chopra and Bhagavan, 1999). One study has indicated that natural vitamin E is retained more than two times better than the synthetic form (Burton et al. (1998). These studies indicate that natural vitamin E,

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consumed either in the diet or ingested as a supplement, would be more efficacious than synthetic vitamin E.

Natural vitamin E is a fat-soluble vitamin that consists of mainly D-α-tocopherol, with β-, γ- and δ-forms in lesser quantities, each having its own biological activity (Traber and Packer, 1995). Sources of natural vitamin E in the diet include vegetable oils, nuts, grains, and green leafy vegetables (Office of Dietary Supplements, National Institutes of Health Clinical Center, 2005). According to the US Institute of Medicine (2000), most North American adults get enough vitamin E from their diets without taking additional supplements. Low-fat diets may, however, result in a decreased vitamin E intake. Additionally, many disorders such as Crohn's disease can cause malabsorption, and may lead to vitamin E deficiency, which may result in nerve damage. The reports of the effects of vitamin E supplementation on coronary artery disease and cancer prevention have shown mixed results and additional studies are underway (Office of Dietary Supplements, 2005).

The source of natural vitamin E used in supplements is a substance known as deodorizer distillate, a by-product of the refinement of vegetable oils. In the process of purification, vegetable oils undergo distillation, and substances with lower boiling points are distilled off and collected. Some of these substances in the oil may give the oil an odor and/or color. Therefore, the mixture of substances collected is called deodorizer distillate. Most natural vitamin E is extracted from soybean distillate because of its relatively high concentration of tocopherols (11–14%) and great abundance due to high production levels (Mendes et al., 2002). Tocopherols are separated from the other components of deodorizer distillate by a variety of methods, including supercritical fluid extraction (Mendes et al., 2005), solvent extraction (Buczenko et al., 2003), molecular distillation (Batistella et al., 2002), and esterification (Yang et al., 2002).

Other substances derived from deodorizer distillate include phytosterols used in the manufacture of margarine, salad dressing, and snack bars (Spizzirri and John, 2004), and in the pharmaceutical industry as a starting material for the production of vitamins and hormones (Mendes et al., 2002).

It has previously been found that substances such as chlorinated pesticides and polychlorinated biphenyls (PCBs) are removed from vegetable oils during the distillation step and are concentrated in the deodorizer distillate (Chaudry et al., 1976; Mounts et al., 1969; Smith et al., 1968, and Young and Kamps, 1982). Therefore, deodorizer distillate has been banned for direct use in animal feeds in the United States due to the elevated levels of these contaminants. Samples of deodorizer distillate were recently found to contain elevated levels of dioxins and furans as well (Ferrario et al., 2002). Since natural vitamin E and other products are derived from deodorizer distillate, the possibility of carryover of these contaminants should be considered.

Table 1
Analytes and toxic equivalency factors (TEFs)[a]

Congener	Abbreviation	WHO TEF
2,3,7,8-Tetrachlorodibenzo-*p*-dioxin	2,3,7,8-TCDD	1
1,2,3,7,8-Pentachlorodibenzo-*p*-dioxin	1,2,3,7,8-PeCDD	1
1,2,3,4,7,8-Hexachlorodibenzo-*p*-dioxin	1,2,3,4,7,8-HxCDD	0.1
1,2,3,6,7,8-Hexachlorodibenzo-*p*-dioxin	1,2,3,6,7,8-HxCDD	0.1
1,2,3,7,8,9-Hexachlorodibenzo-*p*-dioxin	1,2,3,7,8,9-HxCDD	0.1
1,2,3,4,6,7,8-Heptachlorodibenzo-*p*-dioxin	1,2,3,4,6,7,8-HpCDD	0.01
Octachlorodibenzo-*p*-dioxin	OCDD	0.0001
2,3,7,8-Tetrachlorodibenzofuran	2,3,7,8-TCDF	0.1
1,2,3,7,8-Pentachlorodibenzofuran	1,2,3,7,8-PeCDF	0.05
2,3,4,7,8-Peentachlorodibenzofuran	2,3,4,7,8-PeCDF	0.5
1,2,3,4,7,8-Hexachlorodibenzofuran	1,2,3,4,7,8-HxCDF	0.1
1,2,3,6,7,8-HexachlorodibenzofuranF	1,2,3,6,7,8-HxCDF	0.1
2,3,4,6,7,8-Hexachlorodibenzofuran	2,3,4,6,7,8-HxCDF	0.1
1,2,3,7,8,9-Hexachlorodibenzofuran	1,2,3,7,8,9-HxCDF	0.1
1,2,3,4,6,7,8-Heptachlorodibenzofuran	1,2,3,4,6,7,8-HpCDF	0.01
1,2,3,4,7,8,9-Heptachlorodibenzofuran	1,2,3,4,7,8,9-HpCDF	0.01
Octachlorodibenzofuran	OCDF	0.0001

[a]World Health Organization TEF values (Van den Berg and Birnbaum, 1998).

During the course of the FDA survey for dioxins and furans, samples of natural ($n = 14$) and synthetic ($n = 21$) vitamin E supplements were collected for analysis. Eight samples of deodorizer distillate were collected from vegetable oil refineries, along with eight samples of bleaching clay used in the refining process. Bleaching clay is routinely used in the purification of vegetable oils and has been reported as a possible source of dioxin/furan contamination (Ferrario et al., 2000).

These samples were analyzed for 17 of the most toxic dioxins and furans, listed in Table 1. Shown in the table are names, abbreviations, and the toxic equivalent factors (TEFs) that have been determined by the World Health Organization (Van den Berg and Birnbaum, 1998). The most toxic of these are 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) and 1,2,3,7,8-pentachlorodibenzo-*p*-dioxin (1,2,3,7,8-PeCDD), both with an assigned TEF value of 1. The toxicities of the other congeners are relative to this value. For example, the octa-chlorinated congeners are only 0.0001 times as toxic as 2,3,7,8-TCDD. Total toxicities for samples are determined by multiplying the amount found in pg/g sample times the TEF for each congener. The resulting toxic equivalents (TEQs) are added together to obtain the total TEQ for the sample.

2. Materials and methods

2.1. Sample collection

All vitamin E supplements were collected during the years 2002 and 2004 as part of the normal workplan assignments as directed by the US Food and Drug Administration's Center for Food Safety and Applied Nutrition (CFSAN) for the ongoing dioxin monitoring program. The supplements were in the form of gelatin capsules and were collected in bottles from store shelves

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