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Daily intake of 4-nonylphenol in Taiwanese

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Abstract

Alkylphenol polyethoxylates (APEO), alkylphenols combined with ethylene oxide, are a class of nonionic surfactants. APEO have been widely used for industrial, agricultural and household applications, and are biodegraded to more persistent and estrogen-active products, namely, nonylphenol (NP), octylphenol (OP), butylphenol (BP), nonylphenol monoethoxylate (NP_1EO) and nonylphenol diethoxylate (NP_2EO). This study determined NP levels in commonly consumed foodstuffs to assess daily intake of NP in a Taiwanese population.

This study analyzes 318 of samples from 25 types of commonly consumed foodstuffs in northern, central, southern and eastern regions of Taiwan and estimates daily intake of NP in 466 subjects. Moreover, daily NP intake for 3915 additional subjects was estimated by analyzing data from the Nutrition and health survey in Taiwan (NAHSIT).

The foodstuff samples were analyzed for five alkylphenol compounds simultaneously by HPLC with fluorescence detection. Additionally, the average compositions of typical foods consumed in Taiwan were investigated. In combination with alkylphenol levels in these foodstuffs, daily intake of NP in Taiwanese was calculated.

The average daily intake of NP for the 466 subjects was 28.04 ± 25.32 μg/day. Estimated daily intake of NP, based on NP levels in this study as well as the NAHSIT data, was 31.40 μg/day. Rice was the most commonly consumed source of NP, the proportion was 21.46% among daily intake of NP and the following were aquatic products and livestock, which percentage were 17.97% and 17.38%, respectively. Additionally, oysters had the highest NP levels (235.8 ± 90.7 ng/g) in four regions of Taiwan, followed by salmon (123.8 ± 116.2 ng/g). This study suggested that the average daily NP intake in Taiwan is 4-fold and 8.5-fold higher than daily intake in Germany and New Zealand, respectively and rice was the major source of NP intake.

Keywords: Nonylphenol; Foodstuff; Daily intake; Taiwan

1. Introduction

Alkylphenol polyethoxylates (APEO) are the most widely used classes of nonionic surfactants in industrial, agricultural and household applications (Ying et al., 2002). Cheng and Ding (2002) indicated that nonylphenol polyethoxylates (NPEO) levels of commercial liquid detergents in Taiwan range from 0.2% to as high as 21%. Approximately 60% of used APEO is discharged into the aquatic environment (Renner, 1997; Sole et al., 2000) then biodegrades to shorter-chain and more persistent products (Giger et al., 1984; Jonkers et al., 2001) such as nonylphenol (NP), octylphenol (OP), nonylphenol monoethoxylate (NP_1EO) and nonylphenol diethoxylate (NP_2EO). Octylphenol has a relative potency (compared to 17β-estradiol) of 0.0000370 in a rainbow trout hepatocytes. The relative potencies of NP and NP_2EO are 0.0000090 and 0.0000060, respectively (Jobling and Sumpter, 1993). Fabrication of plastic and rubber products often requires extensive use of 2,4-di-*tert*-butylphenol (BP) as an antioxidant (Nerín et al., 2003; Mutsuga et al., 2003). These estrogen-like products are known to widely contaminate biota and foodstuffs (Ahel et al., 1993; Tsuda et al., 2000).

As a result of potential interference with estrogens by APEO and bioaccumulation in organisms, APEO was banned for use in detergents by the 2000 OSPAR convention (PARCOM recommendation, 1992) and the 2005 European Commission

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(European Chemicals Bureau, 2002). However, APEO is still used as an emulsifier in pesticide and as an antioxidant in plastics (Tanghe et al., 1999; Howe et al., 2001). Biodegraded APEO products are detectable in rivers, groundwater, soil, sewage and seawater in many countries (Ding and Tzing, 1998; Thibaut et al., 1998; Jorgen et al., 2002; Ferguson et al., 2001).

Alkylphenol polyethoxylates is produced on a large scale and is widely used in numerous industrial manufacturing processes or domestic detergents in Taiwan. However, because of deficiencies in the sanitation system, levels of alkylphenol compounds in Taiwanese rivers and sediments are higher than in some other countries (Ding and Wu, 2000; Wang et al., 2001).

The majority of APEO intake is through commonly available foodstuffs. Guenther et al. (2002) reported that NP is ubiquitous in foods. Yang and Ding (2005) indicated that fresh fruits and vegetables reveal several ng/g of NP and OP. Human milk also contains NP and OP (Ye et al., 2006; Otaka et al., 2003). Most disturbingly, various plastic food containers and wrappings have been found to allow migration of alkylphenol residue into foodstuffs (Inoue et al., 2001; Loyo-Rosales et al., 2004). Guenther et al. (2002) in a study of German adults reported a daily NP intake of 7.5 μg/day and Thomson et al. (2003) reported that the average daily intake of alkylphenols in New Zealand males above 25 years old is 3.6 μg/day.

However, systematic data regarding alkylphenols concentrations in foodstuffs and daily intake of alkylphenols in a mass population are scarce. The present study analyzes NP, OP, BP, NP_1EO and NP_2EO levels in foods commonly consumed in Taiwan and estimates daily levels of alkylphenol intake.

2. Methods

2.1. Chemicals and reagents

4-Nonylphenol (NP) and 4-*tert*-octylphenol (OP) were purchased from Fluka (Japan). The purity of NP and OP was all >90%. 2,4-Di-*tert*-butylphenol (BP) with a purity of over 98% was purchased from Merck (Germany). *p-n-*

Table 1
Concentrations of NP, OP, BP, NP_2EO and NP_1EO in Taiwan foodstuff samples (unit: ng/g wet weight)

Samples	*N*	NP	OP	BP	NP_2EO	NP_1EO
Fresh water fish	37	56.5±56.5*	6.9±10.8	9.6±12.4	n.d.	n.d.
Milkfish	19	59.0±69.7	6.8±10.7	9.3±7.3	n.d.	n.d.
Tilapia	18	54.5±44.6	7.0±11.0	9.9±16.1	n.d.	n.d.
Saltwater fish	61	62.8±88.7	26.9±36.7	10.5±11.9	52.7±147.4	5.4±14.1
Salmon	13	123.8±116.2	63.7±47.5	25.6±11.3	241.9±252.2	9.8±29.4
Cod	12	94.7±122.3	39.8±38.9	15.0±11.9	n.d.	n.d.
Sailfish	13	27.8±23.5	6.5±5.8	3.4±1.9	n.d.	n.d.
Mackerel	13	36.5±25.7	10.3±11.9	3.8±2.6	n.d.	n.d.
Silver pomfret	10	16.2±12.1	9.0±5.0	n.d.	n.d.	n.d.
Shellfish	31	162.4±116.1	6.4±6.2	10.1±9.2	n.d.	4.7±3.5
Oyster	16	235.8±90.7	6.3±6.0	7.4±6.3	n.d.	5.2±4.7
Clam	15	75.4±76.1	6.5±6.5	13.3±11.1	n.d.	n.d.
Other sea foods	18	16.7±21.1	14.7±19.7	3.0±1.0	n.d.	n.d.
Shrimp	9	17.3±12.4	17.9±16.7	2.8±0.7	n.d.	n.d.
Squid	9	16.1±27.3	11.7±22.2	3.2±1.3	n.d.	n.d.
Poultry						
Chicken	17	71.3±79.2	23.0±39.2	12.0±11.9	n.d.	n.d.
Duck	13	19.3±13.7	2.8±0.6	3.6±3.4	n.d.	n.d.
Livestock						
Pig	16	47.3±41.4	3.0±1.4	3.5±3.4	n.d.	n.d.
Beef	17	69.1±64.7	4.5±5.1	3.6±3.3	n.d.	n.d.
Dark green vegetables						
Lettuce	9	7.5±4.2	4.5±2.4	4.7±2.0	n.d.	n.d.
Green vegetables						
Cabbage	15	31.0±22.7	4.5±3.9	n.d.	n.d.	n.d.
Rice						
Rice	16	39.7±21.0	3.2±1.2	19.3±18.9	n.d.	n.d.
Noodles						
Noodle	9	5.8±3.0	n.d.	3.7±1.6	n.d.	n.d.
Other protein						
Egg	9	18.6±8.5	3.9±1.8	3.1±1.5	n.d.	n.d.
Milk	9	16.9±14.2	2. 9±0.8	n.d.	n.d.	n.d.
Soybean milk	9	7.3±4.2	n.d.	2.9±1.1	n.d.	n.d.
Fruit						
Total	32	24.1±15.8	5.9±5.4	2.8±1.1	n.d.	n.d.
Watermelon	15	22.0±11.8	3.4±1.9	3.2±1.7	n.d.	n.d.
Pineapple	9	27.4±17.9	7.7±5.6	n.d.	n.d.	n.d.
Guava	8	24.3±20.9	9.1±7.8	n.d.	n.d.	n.d.

*Mean±SD.
Calculated assuming that non-detected congener concentrations are equal to half of the limit of detection.
NP: n.d. <8.9 ng/g, OP: n.d.<5.4 ng/g, BP: n.d. <5.2 ng/g, NP_2EO: n.d. <8.7 ng/g, NP1EO: n.d. <8.1 ng/g.